Anomalous time behaviors of remanent and normal magnetizations in spinel spin glass Ga_{0.8}Fe_{0.2}NiCrO₄

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Complex spin glass behavior is found to be present in a spinel system $Ga_{0.8}Fe_{0.2}NiCrO_4$ where the cations occupy two different sites, namely the tetrahedrally coordinated A site and the octahedrally coordinated B site. The complex findings include anomalous time (*t*) variations of isothermal remanent magnetization (IRM), thermoremanent magnetization (TRM), zero field cooled magnetization [M(ZFC)], and field cooled magnetization [M(FC)]. Surprising results, like M(FC) < M(ZFC), M(FC) decreasing and changing faster than a slow increasing M(ZFC) with *t*, the M(FC), M(ZFC) not moving (i.e., not changing with *t*) toward a common equilibrium value, M_{eq} , and the IRM, TRM increasing with *t*, also exist for certain *H* (external magnetic field), *T* (temperature), *t* values. We have tried to understand the above and other observed results on the basis of the possible separate time behaviors of the A- and B-site magnetizations in the system.

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I. INTRODUCTION

We report here anomalous time (t) dependencies of isothermal remanent magnetization (IRM), thermoremanent magnetization (TRM), zero field cooled (ZFC) magnetization [M(ZFC)], and field cooled (FC) magnetization [M(FC)] in a spinel spin glass (SG) $Ga_xFe_{1-x}NiCrO_4$ (x=0.8). Some of these dependencies look too abnormal when compared with the results existing in the literature. A SG system has many ground states and it drifts with t from one ground state one possible arrangement (pattern) of randomly frozen (SG frozen) moments] to the other while attempting to reach some equilibrium ground state (GS). It is this drift which is responsible for the t dependence of various M, M_r (magnetizations) or what may also be called as normal magnetizations) and remanent magnetizations [i.e., M(ZFC), M(FC), IRM, TRM]).¹⁻³ Normally in a SG system, M(FC) has very slow variation, generally increasing, with t, as it is supposed to be near the system's equilibrium GS magnetization, M_{eq} , and M(ZFC) [<M(FC)] increases at a faster rate toward M(FC)[i.e., M_{eq}]. On the other hand IRM, TRM decay with t, which looks natural as, unlike M(ZFC), M(FC) case, the external magnetic field, H, =0 there.^{1–8} However in the present system, i.e., spinel ferrite Ga_{0.8}Fe_{0.2}NiCrO₄, to our surprise we find results, like M(FC) < M(ZFC) and IRM, TRM increasing with t (even though H=0), which look highly puzzling at first sight. These results are H, T (temperature), t dependent and in the following sections we describe the details. The system with x=0.8 has been chosen for the study as its SG transition temperatures are in a convenient T range. Even though the SG nature of spinel ferrites is known, 9,10 their M, $M_r - t$ behaviors have systematically not been investigated. Therefore the important results reported in the present work have remained unobserved so far. The present study thus adds to the physics of spinel ferrites, which are an important class of compounds, and provides an advancement to the understanding of their magnetism. It also shows that great care should be taken in interpreting any of their properties which could be influenced by their magnetism, like transport properties. We studied the present system after observing, in a recent study,¹ an oscillatory M(FC) vs *t* and a maximum in M(ZFC) vs *t* in a crystallographically anisotropic (orthorhombic) SG pseudobrookite (Fe₂TiO₅). We have explained those results on the basis of the two site occupancy of cations in pseudobrookite.¹ Similar to pseudobrookite, the present system is also a two site system. However whereas in pseudobrookite the crystallographic *c* axis provides the quantization (*z*) direction due to crystal field anisotropy, in the present crystallographically isotropic (cubic) system, Weiss field, \mathbf{H}_W , provides the quantization direction. This, as discussed later, gives rise to additional anomalies in the present system's behavior.

II. EXPERIMENT

The system $Ga_xFe_{1-x}NiCrO_4$ was prepared by the conventional ceramic technique. High purity oxides NiO, Cr_2O_3 , Fe_2O_3 , and Ga_2O_3 , intimately mixed in molar proportions, were wet ground. These mixtures, in powder form, were pressed into circular pellets and fired at 1000 °C for 35 h in a muffle furnace. The pellets were furnace cooled, ground into fine powder, repelletized and refired at 1300 °C for 10 h. The above-mentioned sequence was repeated and final sintering was carried out at 1400 °C for 20 h. The pellets were furnace cooled and used as powder samples.

X-ray powder diffraction patterns were recorded using Nifiltered Cu K_{α} radiation (λ =1.5405 Å). The observed peaks were well defined and could be indexed in terms of a single phase cubic spinel structure. The lattice parameter, *a*, was found to decrease linearly with *x*, being *a*=8.275±0.005 Å for *x*=0.8. In the following descriptions, we are concerned only with *x*=0.8 system. The spinel crystal structure has two cation sites with tetrahedral (A-site) and octahedral (B-site) oxygen coordinations.^{11,12} On the basis of the site preference energy of the individual cations,^{11,13} the expected cation dis-

tribution for the studied system is $(Ga_{0.8}^{3+}Fe_{0.2}^{3+})_A$ [Ni²⁺Cr³⁺]_BO₄. Our x-ray intensity and Mössbauer spectroscopy measurements confirm this cation distribution for the system. The x-ray intensity data were obtained both from the area under the peaks and by a pulse counting technique with a slow scanning speed. The presence of Ga³⁺ at the A site was confirmed by comparing the observed and calculated intensity ratios (220)/(440) and (422)/(440).^{11,14} Mössbauer spectra were recorded in a transmission geometry at different temperatures with H=0, 12, and 40 kOe. They show A-site location of Fe³⁺ ions with $M_A < M_B$ and no canting of moments on A site (M_A , M_B =A-site, B-site magnetization). ac susceptibility (χ_{ac}) and M, M_r measurements were made using a commercial ac susceptometer [21 Hz, 0.5 Oe (rms) ac field] and vibrating sample magnetometer. The M(ZFC) value obtained at high H (\sim 55 kOe) was used to calculate the magnetic moment per formula unit, n_B ,¹¹ which showed a canting of moments at B site with average canting angle, $\overline{\theta}_{B}$, equal to 65° at 4.2 K and 70° at 50 K.

III. RESULTS AND DISCUSSION

Frustration [simultaneous presence of competing (opposite) instructions at a spin site concerning its orientation direction] and disorder [random distribution of cations (magnetic and diamagnetic ions) in the lattice are the basic requirements for a SG system. In a spinel ferrite this requirement gets satisfied in the following manner. There are three exchange interactions present in a spinel lattice, J_{AA} (between A site ions), $J_{\rm BB}$ (between B site ions), and $J_{\rm AB}$ (between A and B site ions). All the three exchange interactions are negative, which makes J_{AB} competitive to J_{BB} , J_{AA} . Thus the orientation instruction of J_{AB} to a spin is opposite to that of $J_{\rm BB}$ or $J_{\rm AA}$. However, generally $J_{\rm AB} \gg J_{\rm BB}$, $J_{\rm AA}$ (Refs. 11 and 12) which causes antiparallel alignment of A-, B-site moments (ferrimagnetic ordering). A SG freezing (random direction alignment of moments) occurs when diamagnetic ions are present in the lattice and magnetic and diamagnetic ions are randomly distributed. This happens in $(Ga_{0.8}Fe_{0.2})_A[NiCr]_BO_4$ where Fe^{3+} $[S_{Fe^{3+}}$ (Fe³⁺ spin value) =5/2] and diamagnetic Ga^{3+} ($S_{Ga^{3+}}=0$) are randomly distributed on A site and magnetic ions with dissimilar moments, Ni²⁺ (S_{Ni²⁺}=1) and Cr³⁺ (S_{Cr³⁺}=3/2), are randomly distributed on B site. Such a random distribution makes J_{AB} comparable to $J_{\rm BB}$, $J_{\rm AA}$ and the resultant spin direction different at different sites¹⁰ causing SG freezing of moments. However SG freezing occurs via cluster formation¹⁵ and there are four transition temperatures, namely T_{CF} , T_C , T_{M1} and T_{M2} ; normally $T_{CF} > T_C > T_{M1} > T_{M2}$, but near the tricritical point, T_C , T_{M1} may be quite close. This is so-called reentrant SG behavior.¹⁵ As one cools the lattice, at T_{CF} magnetic clusters are formed in the material's otherwise paramagnetic state. The magnetic ordering inside the clusters is ferrimagnetic and they remain uncoupled from each other until T_C is reached. Assigning a spin S_{cl} to a cluster, at T_C the z component of \mathbf{S}_{cl} , $(\mathbf{S}_{cl})_z$, of all clusters get magnetically ordered and the x, y components, $(\mathbf{S}_{cl})_x$, $(\mathbf{S}_{cl})_y$, average out to zero; T_C =system's ferrimagnetic Curie temperature. On further



FIG. 1. (A) Temperature (*T*) dependence of magnetization (*M*) recorded using external magnetic field H=70 Oe. Curve a [b] there, and also in inset, belongs to the zero field cooled (ZFC) [field cooled (FC)] case. The inset shows *T* dependence of *M* (recorded using H=40 kOe) and of ac susceptibility (χ_{ac}). The horizontal (vertical) arrows there, and also in (B), indicate the *y* (*x*) axes to which the curves belong. The $\chi_{ac}-T$ curve has been recorded for ZFC sample, and H=0 during measurement. (B) M-H variation recorded at T=150 K (curve a) and 300 K (curve b). Inset (a) [(b)] shows hysteresis curve recorded at 4.2 K [50 K] and inset (c) shows 50 K hysteresis curve's central portion; the hysteresis loop's branch numbers are marked there. All the curves in (B) have been recorded for ZFC sample.

cooling, at T_{M1} (\mathbf{S}_{cl})_z remain magnetically ordered but (\mathbf{S}_{cl})_x, (\mathbf{S}_{cl})_y freeze in SG configuration (random direction pointing on the average). Finally at T_{M2} , all three components, (\mathbf{S}_{cl})_z, (\mathbf{S}_{cl})_x, (\mathbf{S}_{cl})_y, get randomly frozen in SG configuration. Figure 1 shows these transitions clearly for Ga_{0.8}Fe_{0.2}NiCrO₄. Curves a, b in Fig. 1(A) show the magnetic irreversibility, M_{irr} , behavior. For doing M_{irr} [branching of ZFC (a) -FC (b) curves] measurement, sample is zero field (H=0) cooled from room temperature (RT) [i.e., from above T_{CF} (which is ~170 K as discussed in the following)] to the required temperature (4.2 K) and a desired field H (70 Oe) applied there and then M vs T recorded, with H present, up to RT. This is ZFC curve [curve a; M (ZFC) vs T curve]. With the same Hpresent, the sample is then field cooled from RT to 4.2 K and M vs T recorded with H present and T increasing. This is FC



FIG. 2. Temperature (*T*) variation of isothermal remanent magnetization (IRM) and thermoremanent magnetization (TRM), recorded using *H* (external field) =40 kOe (curves a) and 70 Oe (curves b). Horizontal arrows indicate the *y* axes to which the curves belong and for these measurements, H has been applied and removed in the desired way as described in the text. The IRM, TRM-*T* data recorded using H=20 kOe are similar in nature to the 40 kOe data.

curve (curve b; M(FC) vs T curve). It is seen from the curves a, b, that as the lattice cools, we get $T_C \sim 110$ K (where M starts increasing), $T_{M1} \sim 85$ K [where M_{irr} (a-b curve branching) starts] and $T_{M2} \sim 30$ K (below which M_{irr} becomes strong). Such a behavior has been seen in other SG systems also. Curve c in Fig. 1(A) inset shows χ_{ac} vs T and the T_{M1} , T_{M2} peaks are clearly seen there; the T_C peak is merged with the T_{M1} peak's high temperature tail.⁵ It may be noted that the T_{M1} , T_{M2} peak positions, and also to some extent the T_C peak position, get affected by H and therefore their values may not be identical in χ_{ac} and low field M, vs T, data. At high H, SG transition temperatures change significantly. Curves a, b in Fig. 1(A) inset are M(ZFC), M(FC) vs T curves recorded using H=40 kOe and we see from them that T_{M1} has decreased to ~65 K; the T_C and T_{M2} cannot be determined there owing to monotonic increase of M with T. The M-T, H behavior of Fig. 1(A) is similar to what has been seen in pseudobrookite¹ and other SG systems^{4,5} but $M_{\rm irr}$ behavior using a field as high as 40 kOe has not been investigated there.

Below T_C (~110 K), the M-H curves show magnetic hysteresis. Figure 1(B) shows typical hysteresis curves recorded at 4.2 K [inset (a)] and 50 K [inset (b)]. The 50 K



FIG. 3. Isothermal remanent magnetization (IRM) and thermoremanent magnetization (TRM) recorded as a function of time (*t*) using H (external field) =70 Oe at T=50 K (curves a) and 4.2 K (curves b). Horizontal [vertical] arrows indicate the y [x] axes to which the curves belong. For these measurements, H has been applied and removed in the desired way as described in the text. The above description is valid for the main figures [(a) and (b)] and also for their insets which show enlarged view of some portions of the main figures' curves.

hysteresis curve has very small loopwidth and inset (c) gives an enlarged view of its central portion showing a loopwidth of ~50 Oe. Above T_C , magnetic hysteresis is not observed but the M-H variation is nonlinear. Figure 1(B) curve a shows a typical nonlinear M-H curve recorded at 150 K. This nonlinearity is found to persist up to ~ 170 K above which linear M-H curves are obtained; for instance Fig. 1(B) curve b shows a typical linear M-H curve recorded at 300 K (RT). The nonlinear M-H variation, without any hysteresis, indicates the presence of uncoupled magnetic clusters¹⁵ in the $T_C < T < 170$ K range giving $T_{CF} \sim 170$ K. Above T_{CF} , clusters are absent and the system is truly paramagnetic as is shown by the linear M-H variation. Mössbauer measurements too show the presence of magnetic clusters below ~ 170 K and their absence above that temperature.

Thus the Fig. 1 results are able to show all the four transition temperatures, viz. T_{CF} , T_C , T_{M1} , and T_{M2} . The T_C , T_{M1} , and T_{M2} transitions are seen more conspicuously in Fig. 2 in IRM, TRM vs *T* measurements. For the IRM measurement, the sample is zero field cooled from RT to 4.2 K where *H* is



FIG. 4. Time (*t*) dependence of isothermal remanent magnetization (IRM) and thermoremanent magnetization (TRM) using *H* (external field) =40 kOe at T=50 K (curves a) and 4.2 K (curves b). Horizontal [vertical] arrows indicate y [x] axes to which the curves belong. For these measurements, *H* has been applied and removed in the desired way as described in the text. The above description is valid for the main figures [Fig. (a), Fig. (b)], Fig. (a) inset (i) and Fig. (b) inset. Data recorded using H=20 kOe, 10 kOe are similar in nature to the 40 kOe data. Inset (i) of Fig. 4(a) and Fig. 4(b) shows enlarged view of some portions of the main figures' curves. Inset (ii) of Fig. 4(a) shows TRM vs *t* curve recorded at 4.2 K using H=40 kOe but *H* applied at 115 K (just above T_C), for sample field cooling, and not at room temperature (RT) as has been done for all other TRM vs *t* curves of Figs. 3 and 4. Details are discussed in the text.

applied and removed. The remanent magnetization, IRM, vs T is then recorded. For the TRM measurement, the sample is field cooled in H from RT to 4.2 K where H is removed and remanent magnetization (TRM) vs T measured. Figure 2(a) [2(b)] shows IRM [TRM] vs T variation for H=70 Oe (curve b) and 40 kOe (curve a). In all the curves, a change of slope is seen at the transition temperatures (T_C , T_{M1} , T_{M2}). However these changes are sharper in curves a (H=40 kOe) and

the T_{M2} transition is seen spectacularly there. This spectacular slope change is similar to what we have seen in pseudobrookite SG.¹ It may be noted that the IRM, TRM vs *T* measurements provide unshifted (*H*=0) values of the transition temperatures.

Having seen the SG nature of the system and obtained its transition temperatures, we now show some very surprising time dependencies of its magnetizations. These measure-

ments have been made at T=4.2 K ($T < T_{M2}$) and 50 K $(T_{M2} < T < T_{M1})$. Figure 3 shows the IRM, TRM vs t for H =70 Oe. As above, for the IRM [TRM] measurement, the sample is ZFC [FC in H] from RT to the measurement temperature, T_m , (4.2 or 50 K) and H applied and removed [H removed] at that temperature before recording IRM [TRM] vs t. As seen in Fig. 3, both IRM and TRM show a decrease with t, normally a fast initial decrease followed by a slow decrease. However this is not the case when the above data are recorded for H=40 kOe (Fig. 4). In Fig. 4 whereas the 4.2 K behavior is similar to what is seen in Fig. 3, at 50 K both IRM and TRM show an initial decrease followed by an increase with t. This increase is surprising for a remanent magnetization where H=0 during measurement. Such a behavior, to our knowledge, has not been reported before. We have tried to understand these results on the basis of the two site (A-, B-site) occupancy of cations in the spinel lattice, where the magnetizations of the two sites, M_A and M_B , are antiparallel. Thus there will be two remanent magnetizations, M_{rA} and M_{rB} , corresponding to A and B sites, respectively, with $M_{rB} > M_{rA}$ since, as stated before, $M_B > M_A$. Therefore the observed remanent magnetization, M_r , is given by, M_r $=M_{rB}-M_{rA}$ where M_r represents IRM or TRM. As the lattice is SG frozen at 4.2, 50 K, the M_{rA} , M_{rB} can have complicated t dependencies like exponential or algebraic or logarithmic or stretched exponential or a combination of these.¹ Therefore M_r vs t can be quite complex. Assuming M_{rA} , M_{rB} vs t curves to be exponential, having their own initial values and decay rates, and $M_r = M_{rB} - M_{rA}$, in Fig. 5 we have shown some possible, computed, t variations of M_r . For instance, in Fig. 5(A) the curve c[c'] shows a situation where M_r decreases [increases] with t and the corresponding M_{rB} , M_{rA} vs t variations are shown by the curves a, b [a',b'], respectively. Similarly in Fig. 5(B), curve c shows a situation where M_r is t independent and c' where M_r shows an initial decrease and then an increase with t. The corresponding M_{rB} , M_{rA} vs t variations are given there by the curves a, b (for curve c) and a', b' (for curve c'). These computed variations match with the results of Figs. 3 and 4. Thus the surprising result of IRM, TRM increasing with t seems to be a consequence of the different t behaviors of M_{rA} , M_{rB} [i.e. (IRM)_A, $(IRM)_B$ or $(TRM)_A$, $(TRM)_B$]. Mathematically, for exponential t dependencies for M_{rA} , M_{rB} , we have

$$M_{r}(t) = M_{rB}(t) - M_{rA}(t) = M_{rB}(0)e^{-\lambda_{B}t} - M_{rA}(0)e^{-\lambda_{A}t},$$
(1)

where $M_{rA}(0)$, $M_{rB}(0)$ are the t=0 values of M_{rA} , M_{rB} and λ_A , λ_B , respectively, represent the decay rates of M_{rA} , M_{rB} . The different *t* behaviors of M_{rA} , M_{rB} arise due to a difference in the λ_A , λ_B values. This difference is physically possible for a spinel lattice owing to the different nearest neighbor (nn) cation environments for A and B sites, which makes the two sites magnetically inequivalent, and any weak A–B coupling, which augments the independence of A, B site behaviors. In a spinel lattice, an A site cation has 12 B site cations as nearest neighbors (nns) and a B site cation has 6 A site and 6 B site cations as nns.^{11,12} Further the weakness of A–B coupling in Ga_{0.8}Fe_{0.2}NiCrO₄ is brought out by the



FIG. 5. Typical time (t) variation of remanent magnetization (M_r) , computed for various possible time behaviors of A- and Bsite remanent magnetizations $(M_{rA} \text{ and } M_{rB})$. For these computations, M_{rA} , M_{rB} vs t curves have been assumed to be exponential, having their own initial values and decay rates, and $M_r=M_{rB}$ $-M_{rA}$. Details are described in the text.

presence of B-site canting (large $\overline{\theta}_{\rm B}$) in the system.^{16,17} It may be noted that a time dependent λ_A , λ_B can make M_r vs t oscillatory in nature. Such is found to be the case with M(ZFC), M(FC) vs t, described in the following, for certain H, T, where $[M(ZFC)]_A$, $[M(ZFC)]_B$ and $[M(FC)]_A$, $[M(FC)]_{B}$ growth rates become time dependent. It may be noted that Fig. 4(a) curve b (called curve I) and Fig. 4(a)inset (ii) curve (called curve II) have been recorded in identical conditions except that for curve I, like all other TRM curves of Figs. 3 and 4, sample was field cooled from RT whereas for curve II, the sample has been field cooled from 115 K (i.e., from just above T_C). As seen there, the initial sharp fall of curve I is absent in curve II. Similarly the curves' nature also depended on the waiting time t_w , before H is applied after zero field cooling, and the cooling rate used in ZFC, FC measurements. These parameters have been





kept uniform in all our measurements reported here to facilitate the curves' comparison.

Figures 6 and 7 show the M(ZFC), M(FC) vs *t* behaviors at 4.2, 50 K for H=70 Oe, 40 kOe. For the M(ZFC) [M(FC)] measurement, the sample is zero field cooled [field cooled in *H*] from RT to T_m (4.2 or 50 K) and *H* applied [not removed] there. With *H* present, M(ZFC) [M(FC)] vs *t* is recorded. Several types of *t* variations are seen in Figs. 6 and 7, like *M* vs *t* almost constant (*t* independent), increasing, increasing and then decreasing or vice versa, and oscillating; here *M* means M(ZFC) or M(FC). As in pseudobrookite case,¹ we can write

$$\begin{split} M(t) &= M_{\rm B}(t) - M_{\rm A}(t) \\ &= \left[M_{\rm B}(\infty) - M_{\rm A}(\infty) \right] - \left[M_{\rm B}(\infty) - M_{\rm B}(0) \right] e^{-\zeta_{\rm B} t} \\ &+ \left[M_{\rm A}(\infty) - M_{\rm A}(0) \right] e^{-\zeta_{\rm A} t}, \end{split}$$
 (2)

where $M_{\rm B}(0)$, $M_{\rm A}(0)$ and $M_{\rm B}(\infty)$, $M_{\rm A}(\infty)$ are the t=0 and t $=\infty$ values of $M_{\rm B}$, $M_{\rm A}$; $M_{\rm B}$ is $[M(ZFC)]_{\rm B}$, $[M(FC)]_{\rm B}$ and $M_{\rm A}$ is $[M(ZFC)]_A$, $[M(FC)]_A$. ζ_B , ζ_A , respectively, represent the growth rate of $M_{\rm B}$ and $M_{\rm A}$. Thus, as in the case of M_r (Fig. 5), different ζ_A , ζ_B values can produce the results of Figs. 6 and 7.¹ Various ζ_A , ζ_B values will occur depending on H, T and cooling conditions (ZFC or FC) and like λ_A , λ_B , difference in ζ_A , ζ_B values is physically possible. As explained in Ref. 1, an oscillatory M vs t arises due to a t dependent ζ_A , $\zeta_{\rm B}$ i.e., a *t* dependent drift rate when the SG system drifts, as mentioned before, among its various ground states. If $\Delta_{\rm B}$ is the energy barrier separating two ground states, then the drift rate, and also ζ_A , ζ_B , will depend on Δ_B/kT .¹ When Δ_B $\sim kT$, any small difference in $\Delta_{\rm B}$ seen by the drifting system, when it moves from one GS to the other, is felt by the system making $\Delta_{\rm B}/kT$, and so the drift rate (i.e., $\zeta_{\rm A}$, $\zeta_{\rm B}$), t dependent. In the case of pseudobrookite, such a situation occurred





at 4.2 K.¹ However in the present system, this situation (i.e., oscillatory behavior) is existing both at 4.2 and 50 K (Figs. 6 and 7). This means that $\Delta_{\rm B} \sim kT$ both at 4.2 and 50 K in presence of *H*. In the present system at 50 K, as discussed before, longitudinal magnetic ordering $[(\mathbf{S}_{\rm cl})_z \text{ ordering}]$ is present since $T_{M2} < 50 \text{ K} < T_{M1}$. This provides anisotropy, for spin reorientation, which enhances $\Delta_{\rm B}$ by adding to system's disorder induced exchange anisotropy. The presence of *H* further enhances $\Delta_{\rm B}$ by providing its own preferential direction for any spin orientation drift. This makes $\Delta_{\rm B} \sim kT$ at 50 K. When the system is cooled to 4.2 K, both $\Delta_{\rm B}$ and kT decrease; $\Delta_{\rm B}$ decreases owing to the absence of preferential longitudinal $[(\mathbf{S}_{\rm cl})_z]$ ordering direction at 4.2 K as 4.2 K $< T_{M2}$. This seems to be making $\Delta_{\rm B} \sim kT$ at 4.2 K also. In

pseudobrookite,¹ $\Delta_{\rm B} \sim kT$ at 4.2 K and $\langle kT$ at 30 K. This is because at 30 K though kT has increased, $\Delta_{\rm B}$ has not owing to the absence of $(\mathbf{S}_{\rm cl})_z$ ordering at 30 K. In pseudobrookite,¹ SG freezing is governed by the crystal field anisotropy and at 30 K, **a**-, **b**-axis spin components are paramagnetic and **c**-axis component is frozen in a random way; **a**,**b**,**c** = crystal axes. Thus there is no preferential magnetic ordering direction of any kind at 30 K. Another strange looking result is seen in Fig. 7(b) inset (ii) where 50 K M(ZFC), M(FC) vs *t* curves are plotted together. As seen there, (i) M(FC) [curve a'] is changing faster than M(ZFC) [curve b'] after a certain *t*, (ii) M(FC) < M(ZFC) after certain *t*, and (iii) M(ZFC), M(FC) do not seem to be drifting toward a common $M_{\rm eq}$ as $t \rightarrow \infty$. Similar results are seen at 4.2 K also

[Figs. 7(a) and 7(b)]. All these results [(i), (ii), (iii)] look abnormal as they are opposite to what is observed in a normal SG system. However we can understand these results by realizing that for our system, $M(FC) = [M(FC)]_{B} - [M(FC)]_{A}$ and $M(ZFC) = [M(ZFC)]_B - [M(ZFC)]_A$. Thus even though $[M(FC)]_{A,B}$ and $[M(ZFC)]_{A,B}$ may be behaving in a normal way, i.e., $[M(FC)]_{A,B} > [M(ZFC)]_{A,B}$, $[M(FC)]_{A,B}$ having slow t variation and $[M(ZFC)]_{A,B}$ increasing at a faster rate toward $[M(FC)]_{A,B}$, this need not be the case with $[M(FC)]_{B}$ - $[M(FC)]_{A}$ and $[M(ZFC)]_{B}$ - $[M(ZFC)]_{A}$ which we actually measure. They can have any variation, especially at large H, like 40 kOe, where $[M(ZFC)]_{A,B}$ is expected to be comparable to $[M(FC)]_{A,B}$ (since larger the *H*, closer are the M(FC), M(ZFC) values in a normal SG system). Thus depending on the $[M(FC)]_A$, $[M(FC)]_B$, $[M(ZFC)]_A$, $[M(ZFC)]_{B}$ growth rates, all the Fig. 7(b) inset (ii) results can be theoretically reproduced. These results indicate the

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extent to which the time behavior of a SG system can be complex.

IV. CONCLUDING REMARKS

To summarize, we find surprising time behaviors for the spinel spin glass $Ga_{0.8}Fe_{0.2}NiCrO_4$. This is a two site system where the cations are distributed on two sites, A and B, in the lattice. The observed abnormal looking results have been understood on the basis of the separate A-, B-site time behaviors. These results and their understandings add to our knowledge of spinel ferrite physics and are significant as the spinel ferrites, along with garnets, form an important class of magnetic compounds.^{9–14,17} They also show that great care must be taken while interpreting the *t* behaviors of certain SG systems, like say high- T_c superconducting SG systems, ^{15,18} which could have more than one cation site in their lattice; T_c =critical temperature.

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