Aging dynamics in a reentrant ferromagnet: $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ graphite bi-intercalation compound

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The aging dynamics of a reentrant ferromagnet $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ graphite bi-intercalation compound has been studied using ac and dc magnetic susceptibility. This compound undergoes successive transitions at the transition temperatures T_c (=9.7 K) and T_{RSG} (=3.5 K). The relaxation rate $S(t)$ $[=d\chi_{ZFC}(t)/d\ln t]$ exhibits a characteristic peak at t_{cr} close to a wait time t_W below T_c, indicating that the aging phenomena occur in both the reentrant spin glass phase below T_{RSG} and the ferromagnetic (FM) phase between T_{RSG} and T_c. The aging state in the FM phase is fragile against a weak magnetic-field perturbation. The time (t) dependence of $\chi_{ZFC}(t)$ around $t \approx t_{cr}$ is well approximated by a stretched exponential relaxation, $\chi_{ZFC}(t) \approx \exp[-(t/\tau)^{1-n}]$, around $t = t_W$. The exponent n depends on t_W, T, and H. The relaxation time τ ($\approx t_{cr}$) exhibits a local maximum around 5 K, reflecting a frustrated nature of the FM phase. It drastically increases with decreasing temperature below T_{RSG}.

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I. INTRODUCTION

In random spin systems with competing ferromagnetic and antiferromagnetic interactions, the spin frustration effect occurs, leading to a spin-glass (SG) phase at low temperatures. This situation may change when there are a majority of the ferromagnetic interactions and a minority of the antiferromagnetic interactions to create a substantial spin frustration effect. The system (so-called reentrant ferromagnet) exhibits two phase transitions at T_{RSG} and T_c ($T_c > T_{RSG}$): the reentrant spin glass (RSG) phase below T_{RSG} and the ferromagnetic (FM) phase between T_{RSG} and T_c. Experimental studies of the dynamic magnetic properties have been carried out for reentrant ferromagnets such as $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$,[1–4] $Cr_{78}Fe_{22}$,[5] $(Fe_{0.65}Ni_{0.35})_{0.882}Mn_{0.118}$,[6] $CdCr_{2x}In_{2(1-x)}S_4$ ($x=0.90$, 0.95, and 1.00),[7–9] and $Fe_{0.7}Al_{0.3}$.[10] For $Cr_{78}Fe_{22}$,[5] $(Fe_{0.65}Ni_{0.35})_{0.882}Mn_{0.118}$,[6] and $Fe_{0.7}Al_{0.3}$,[10] the RSG phase exhibits aging phenomena, which are very similar to those observed in the SG phase of SG systems. No aging phenomenon has been observed in the FM phase. For $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$,[1–4] in contrast, not only the RSG phase but also the FM phase exhibit aging phenomena. A strikingly increased fragility to the magnitude of the magnetic field is observed when passing from the low-temperature RSG region into the FM phase. The effect of the probing field H on the relaxation rate has to be carefully considered. The dramatic decrease of the limiting field around T_{RSG} may explain why other experiments on reentrant ferromagnets have not resolved an aging behavior in the FM phase. For $CdCr_{2x}In_{2(1-x)}S_4$ with $x=0.90$, 0.95, and 1.0,[7–9] the aging behavior of the low-frequency ac susceptibility (absorption χ'') is observed in both the FM and RSG phases.

Experimental studies of the aging dynamics have been limited to a macroscopic measurement such as the time evolution of zero-field-cooled (ZFC) susceptibility, thermoremnant susceptibility, and the absorption of the ac magnetic susceptibility. Recently Motoya *et al.*[10] have studied time-resolved small-angle neutron scattering of $Fe_{0.70}Al_{0.30}$ in order to probe the microscopic mechanism of slow dynamics. The Lorentzian form of the scattering pattern and the temperature variation of the inverse correlation length below T_{RSG} show that the system is composed of only finite-sized clusters. The size of the clusters gradually decreases with decreasing temperature.

$Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ graphite bi-intercalation compound (GBIC) is one of typical three-dimensional (3D) Ising reentrant ferromagnets. It has a unique layered structure where the $Cu_{0.2}Co_{0.8}Cl_2$ intercalate layer ($=I_1$) and $FeCl_3$ intercalate layers ($=I_2$) alternate with a single graphite layer (G), forming a stacking sequence ($-G-I_1-G-I_2-G-I_1-G-I_2-G-\cdots$) along the c axis. In the $Cu_{0.2}Co_{0.8}Cl_2$ intercalate layer, two kinds of magnetic ions (Cu^{2+} and Co^{2+}) are randomly distributed on the triangular lattice. The static and dynamic magnetic properties have been reported in a previous paper.[11] This compound undergoes successive transitions at the transition temperatures T_c (=9.7 K) and T_{RSG} (=3.5 K). A prominent nonlinear susceptibility is observed between T_{RSG} and T_c.

In this paper we report our experimental study of the aging dynamics of the RSG and FM phases of $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC using dc and ac magnetic susceptibility measurements. Our system is cooled from 50 K to T ($<T_c$) in the absence of an external magnetic field. This ZFC aging protocol process is completed at $t_a=0$, where t_a is defined as an age (the total time after the ZFC aging protocol process). Then the system is aged at T under $H=0$ until $t_a=t_W$, where t_W is a wait time. The aging behavior of the ZFC magnetic susceptibility $\chi_{ZFC}(t)$ has been measured under various aging processes: (i) a wait time t_W ($2.0\times10^3 \leqslant t_W \leqslant 3.0\times10^4$ sec), T ($1.9 \leqslant T \leqslant 9$ K), and H ($1 \leqslant H \leqslant 60$ Oe) as parameters and (ii) the T-shift and H-shift perturbations. The relaxation rate defined by $S(t)=d\chi_{ZFC}/d\ln t$ exhibits a peak at a characteristic time t_{cr} close to t_W below T_c, indicating the occurrence of the aging phenomena in both the RSG and FM phases. We will also show that the t dependence of $\chi_{ZFC}(t)$ around $t=t_W$ is well described by a stretched exponential relaxation

$[\chi_{ZFC}(t) \approx \exp[-(t/\tau)^{1-n}]$ (see Sec. II), where n is an exponent and τ is a relaxation time nearly equal to t_{cr}. We will show that n, τ, and t_{cr} depend on T, t_W, and H. The local maximum of τ and t_{cr} around 5 K is observed, reflecting the frustrated nature of the FM phase. A partial rejuvenation of the system occurs in $S(t)$ under the positive shift in the FM phase.

II. BACKGROUND: STRECHED EXPONENTIAL FORM OF $\chi_{ZFC}(t)$

After the SG system is cooled to T ($<T_{SG}$) through the ZFC aging protocol at $t_a=0$, the size of domain defined by $R_T(t_a)$ grows with the age of t_a and reaches $R_T(t_W)$ just before the field is turned on at $t=0$ or $t_a=t_W$.[12] After $t=0$, a probing length $L_T(t,t_W)$ corresponding to the maximum size of excitation grows with t, in a similar way as $R_T(t_a)$. The quasiequilibrium relaxation occurs first through local spin arrangements in length $L_T(t,t_W) \ll R_T(t_W)$, followed by nonequilibrium relaxation due to domain growth, when $L_T(t,t_W) \approx R_T(t_W)$, so that a crossover between the short-time quasiequilibrium decay and the nonequilibrium decay at longer observation times is expected to occur near $t \approx t_W$.

Theoretically[13,14] and experimentally [15–25] it has been accepted that the time variation of $\chi_{ZFC}(t)$ may be described by a product of a power-law form and a stretched exponential function

$$\chi_{ZFC}(t) = M_{ZFC}(t)/H = \chi_0 - At^{-m} \exp[-(t/\tau)^{1-n}], \quad (1)$$

where the exponent m may be positive and is very close to zero, n is between 0 and 1, τ is a characteristic relaxation time, and χ_0 and A are constants. In general, these parameters are dependent on t_W. This form of $\chi_{ZFC}(t)$ incorporates both the nonequilibrium aging effect through the stretched exponential factor $[\exp[-(t/\tau)^{1-n}]$ [Eq. (1) with $m=0$] in the crossover region ($t \approx t_W$ and $t > t_W$) between the quasiequilibrium state and nonequilibrium state and an equilibrium relaxation response at $t \ll t_W$ through a pure power-law relaxation (t^{-m}). Note that Ogielski[13] fits his data by a stretched exponential multiplied by a power function. For $0.6 < T/T_{SG} < 1$, Ogielski[13] fits it by a power law with a different temperature dependence of exponent m. When $t \ll \tau$, $\chi_{ZFC}(t)$ is well described by a power-law form given by At^{-m}. However, in the regime of $t \approx \tau$, the stretched exponential relaxation is a very good approximation in spite of a finite m, which is very small.

For all temperatures, $\chi_{ZFC}(t)$ increases with increasing t and the relaxation rate $S(t)$, which is defined by

$$S(t) = \mathrm{d}\chi_{ZFC}(t)/\mathrm{d}\ln t = t\mathrm{d}\chi_{ZFC}(t)/\mathrm{d}t, \quad (2)$$

exhibits a maximum (S^0_{max}) at t_{cr} that is close to t_W, where t_{cr} is given by

$$t_{cr} = \tau(\xi/2)^{1/(1-n)}, \quad (3)$$

with

$$\xi = [1 - 2m - n + (1-n)^{1/2}(1 - 4m - n)^{1/2}]/(1-n), \quad (4)$$

where $4m+n<1$. Note that the value of t_{cr} is uniquely determined only by the values of n and m. When $m=0$, we have $t_{cr}=\tau$ and S^0_{max} $[=A(1-n)/e]$ with $e=2.7182$.

III. EXPERIMENTAL PROCEDURE

We used the same sample of $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC that was used in the previous paper.[11] The details of the sample characterization and synthesis were presented in the references of the previous paper.[11] The dc magnetization and ac susceptibility of $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC were measured using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS XL-5) with an ultralow-field capability option. The remnant magnetic field was reduced to zero field (exactly less than 3 mOe) at 298 K for both dc magnetization and ac susceptibility measurements. The ac magnetic field used in the present experiment has a peak magnitude of the ac field (h) and a frequency ($f=\omega/2\pi$); $0.05 \leqslant h \leqslant 0.3$ Oe and $0.01 \leqslant f \leqslant 10$ Hz. Each experimental procedure for measurements is presented in the text and figure captions.

In our measurement of the time (t) dependence of the ZFC magnetization (M_{ZFC}), the time required for the ZFC aging protocol and subsequent wait time were precisely controlled. Typically it takes 240 ± 3 sec to cool the system from 10 K to T ($<T_c$). It takes another 230 ± 3 sec until T becomes stable within the uncertainty (± 0.01 K). The system is kept at T and $H=0$ for a wait time t_W (typically $t_W = 1.5 \times 10^4$ or 3.0×10^4 sec). At time $t=0$, external magnetic field H is applied along any direction perpendicular to the c axis. The time for setting up a magnetic field from $H=0$ to $H=1$ Oe is 68 ± 2 sec. In the ZFC measurement, the sample is slowly moved through the pickup coils over the scan length (4 cm). The magnetic moment of the sample induces a magnetic flux change in the pickup coils. It takes 12 sec for each scan. The data at t are regarded as the average of M_{ZFC} measured over the scanning time t_s between the times $t-(t_s/2)$ and $t-(t_s/2)$. Thus the time window Δt is a scanning time (t_s). The measurement was carried out at every interval of t_s+t_p, where t_p is a pause between consecutive measurements. Typically we used (i) the time window $\Delta t=36$ sec for the measurements with three scans and $t_p=45$ sec or 30 sec and (ii) the time window $\Delta t=12$ sec for the measurements with one scan and $t_p=1$ or 2 sec.

The actual ZFC process is uniquely determined by the apparatus (Quantum Design MPMS XL-5). We used the same cooling process for all the measurements. This process consists of the following three parts: (i) rapid cooling from high temperatures (≈ 10 K) to temperatures ($=T+\Delta T_1$ with $\Delta T_1 \geqslant 0.5$ K), where T is the measured temperature, (ii) undercooling down to temperatures ($=T-\Delta T_2$ with $\Delta T_2 \approx 0.5$ K), and (iii) overshooting up to $T+\Delta T_3$ with $\Delta T_3 \approx 0.3$ K and slowly approaches T from above. Note that similar cooling process has been used by Zotev *et al.*[26] After T is finally reached within ± 0.01 K, the system is kept at T in the absence of H for the wait time t_W. The question is whether the time required for the system to reach a final

temperature T should be added to t_W ($=1.5\times10^4$ and 3.0×10^4 sec in our case). We find that the details of the cooling process above T do not affect the aging dynamics at T. The undercooling process below T is also not significant, because of its large ΔT_2. The subsequent overshooting because of its small ΔT_3, however, is considered to play a significant role in the aging dynamics and is not neglected in the measurements. In our case of $\Delta T_3 \leqslant 0.3$ K at $T=3.5$ K, the time taken for this process ($\Delta T_3 \to 0$) is less than 60 sec, which is negligibly short compared to t_W. Therefore, it follows that the access time does not influence the aging behavior. This is consistent with a prediction from the droplet model for the SG phase.[27] The spin configuration imprinted at T below a spin freezing temperature T_{SG} is preserved after the T shift from T to $T+\Delta T$ only if $|\Delta T|$ is close to zero. Then the SG short-range order continuously grows after the T shift with a rate which depends on the temperature [see Sec. IV E for the effect of $S(t)$ under the T shift].

IV. RESULT

A. $\chi''(\omega,T)$ and $\chi_{ZFC}(T)$

In the previous paper[11] the magnetic properties of $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC have been extensively studied from our results on dc susceptibility (χ_{ZFC}, χ_{FC}) and ac magnetic susceptibility (χ' and χ''). In Fig. 1(a), as a typical example we show the T dependence of the absorption $\chi''(\omega,T)$ at various f, where $h=50$ mOe. It is concluded from this figure that our system undergoes two phase transitions at $T_{RSG}=3.5$ K and $T_c=9.7$ K. There are a RSG phase below T_{RSG} and an FM phase between T_{RSG} and T_c.

In order to demonstrate the evidence of the aging behavior in χ_{ZFC}, we measured the T dependence of χ_{ZFC} ($=M_{ZFC}/H$) in the following two ZFC protocols. The system was quenched from 50 to 1.9 K in the absence of H before the measurement. The susceptibility χ_{ZFC} was measured with increasing T from 1.9 K to $T_1=7.0$ K in the presence of H (=1 Oe), where T_1 is in the FM phase. The system was kept at T_1 for a wait time $t_{W1}=1.5\times10^4$ sec. Subsequently χ_{ZFC} was measured with further increasing T from T_1 to 20 K. The system was annealed at 50 K for 1.2×10^3 sec in the presence of H (=1 Oe). The susceptibility χ_{FC} was continuously measured from 20 to 1.9 K. In Fig. 1(b) we show the T dependence of χ_{ZFC} and χ_{FC} ($H=1$ Oe) thus obtained (denoted as the ZFC-1 and FC-1 processes). We find that there is a remarkable increase of χ_{ZFC} with increasing t during the one stop at $T_1=7.0$ K. As T again increases from T_1, χ_{ZFC} starts to decrease with increasing T and merges with χ_{ZFC}^{ref} without the stop as a reference at $T\approx7.3$ K [see the inset of Fig. 1(b)]. The state at $T=7.3$ K is uncorrelated with that at T_1 since the temperature difference ΔT is large so that the domain size $R_{T1}(t_{W1})$ is much larger than the overlap length $L_{\Delta T}$ which will be defined in Sec. IV E.

In another measurement (denoted as the ZFC-2 and FC-2 processes), the system was kept at $T_2=3.5$ K in the RSG phase for $t_{W2}=1.5\times10^4$ sec during the ZFC process. In the inset of Fig. 1(b) we show the T dependence of χ_{ZFC} in the ZFC-2 process. There is a remarkable increase of χ_{ZFC} during the one stop at T_2. As T again increases from T_2, χ_{ZFC} starts to increase with a derivative $d\chi_{ZFC}/dT$ that is positive and much smaller than that of χ_{ZFC}^{ref}. This susceptibility χ_{ZFC} merges with χ_{ZFC}^{ref} at $T\approx3.9$ K. The state at $T=3.9$ K is uncorrelated with that at T_2 since the temperature difference ΔT is large so that the domain size $R_{T2}(t_{W2})$ is much larger than $L_{\Delta T}$. The T dependence of χ_{FC} in the FC-2 process is the same as that in the FC-1 process. Note that the gradual increase of χ_{ZFC} with increasing T from the stop temperature is also observed in the SG phase of the 3D Ising spin glasses $Fe_{0.5}Mn_{0.5}TiO_3$ (Ref. 28) and $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC (Ref. 29). Such an aging behavior in χ_{ZFC} may be common to the RSG and SG phases. This is in contrast to the gradual decrease of χ_{ZFC} with increasing T from the stop temperature in the FM phase.

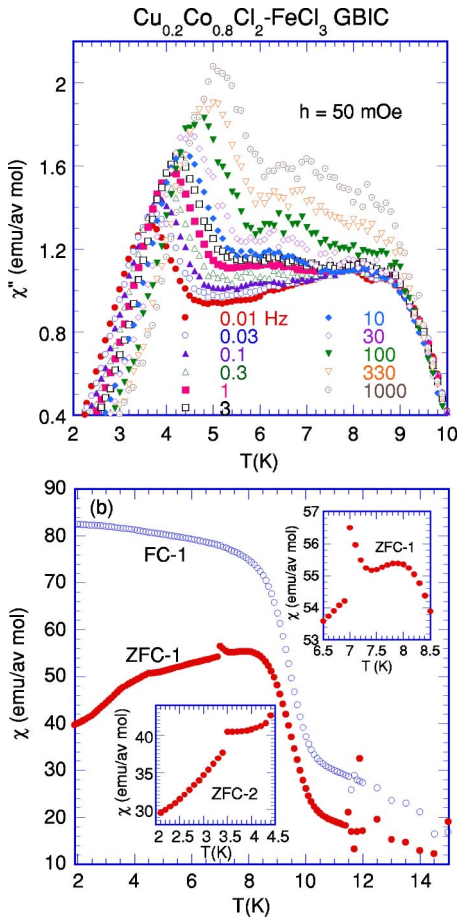

FIG. 1. (Color online) (a) $\chi''(\omega,T)$ vs T at various f ($0.01\leqslant f\leqslant1000$ Hz). $h=50$ mOe. (b) T dependence of χ_{ZFC} (●) and χ_{FC} (○) at $H=1$ Oe for $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC. The measurement was carried out after the ZFC aging protocol: annealing of the system at 50 K for 1.2×10^3 sec at $H=0$ and quenching from 50 to 1.9 K. During the ZFC measurement with increasing T from 1.9 to 20 K, the system was aging at $T_1=7.0$ K (ZFC-1 measurement) and $T_2=3.5$ K (ZFC-2 measurement) for wait time $t_{W1}=t_{W2}=1.5\times10^4$ sec in the presence of $H=1$ Oe. After the ZFC measurement, the system was annealed at $T=50$ K for 1.2×10^3 sec in the presence of H (=1 Oe). The FC measurement (denoted as FC-1 and FC-2) was carried out with decreasing T from 20 to 1.9 K. The T dependence of FC-1 susceptibility is the same as that of FC-2 susceptibility.

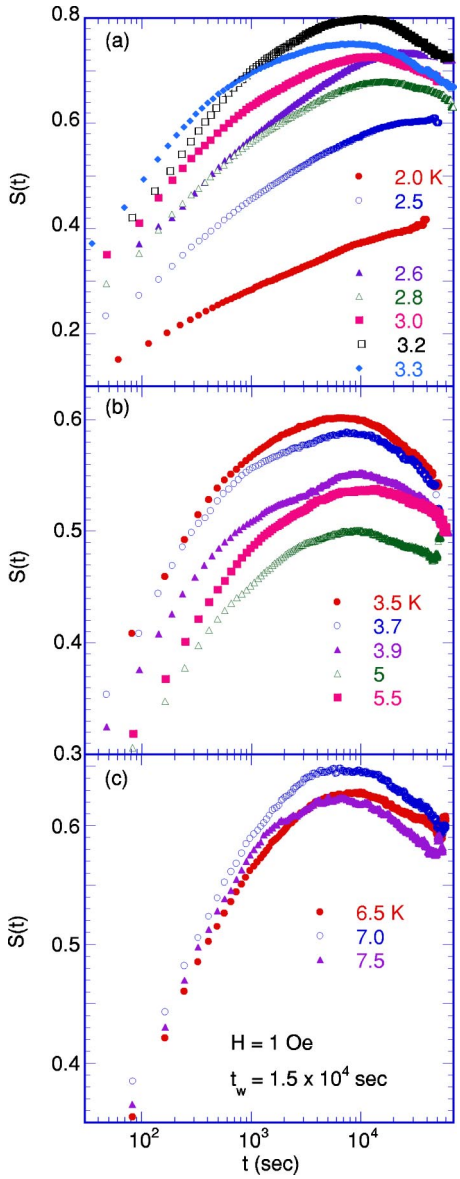

FIG. 2. (Color online) (a)–(c) Relaxation rate S $[=d\chi_{ZFC}/d\ln t]$ vs t at various T. $H=1$ Oe. $t_W=1.5\times10^4$ sec. H is applied along a direction perpendicular to the c axis. The ZFC aging protocol: annealing of the system at 50 K for 1.2×10^3 sec at $H=0$, quenching from 50 K to T, and then isothermal aging at T and $H=0$ for a wait time t_W. The measurement was started at $t=0$ when the field H was turned on.

B. T dependence of $S(t)$

We have measured the t dependence of χ_{ZFC} under the condition with various combinations of T, H, and t_W. Figures 2 and 3 show the t dependence of the relaxation rate $S(t)$ at various $T(2.0\leqslant T\leqslant 9.0$ K) for $t_W=1.5\times10^4$ sec and 3.0×10^4 sec, respectively, where $H=1$ Oe. The relaxation rate $S(t)$ exhibits a broad peak at a characteristic time t_{cr} in the FM phase as well as the RSG phase, indicating the aging behaviors. We find that $\chi_{ZFC}(t)$ is well described by a stretched exponential relaxation form given by Eq. (1) with $m=0$ for $10^2\leqslant t\leqslant 3.0\times10^4$ sec. The factor t^{-m} in Eq. (1) is almost independent of t around $t=t_W$, since m is nearly equal

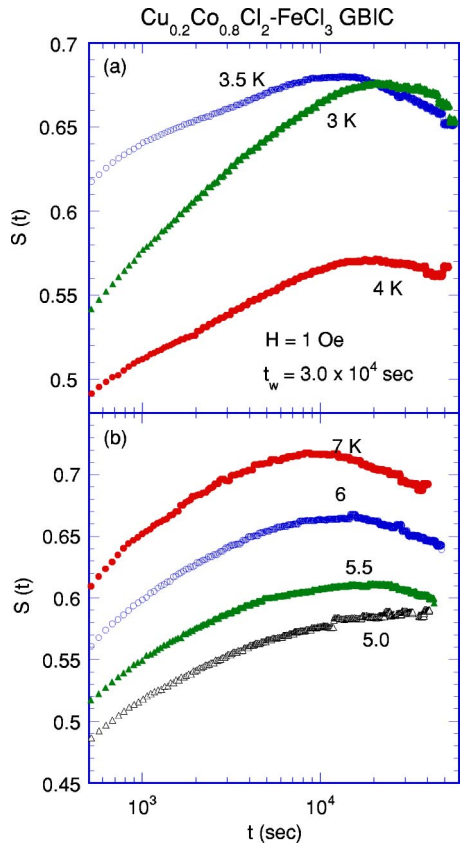

FIG. 3. (Color online) (a) and (b) S vs t at various T. $H=1$ Oe. $t_W=3.0\times10^4$ sec.

to zero. This assumption does not mean $m=0$ in the present system. Note that the least-squares fit of the data of χ_{ZFC} vs t around $t=t_W$ to Eq. (1) does not work well in determining the values of n and m as adjustable parameters: the value of n is very sensitive to very small change in m. The least-squares fits of these data to Eq. (1) with $m=0$ yields the parameters τ, n, and A. Figure 4(a) shows the T dependence of t_{cr} and τ for $t_W=1.5\times10^4$ sec, where $H=1$ Oe. The T dependence of t_{cr} almost agrees well with that of τ for $2\leqslant T\leqslant 9$ K. This indicates that the relaxation is dominated by a stretched exponential relaxation with $m=0$ (see Sec. II). The relaxation time τ $(\approx t_{cr})$ shows a broad peak centered around 5.5 K between T_c and T_{RSG} and a local minimum around T_{RSG}. It increases with further decreasing T below T_{RSG}. As far as we know, there has been no report on the broad peak of t_{cr} (or τ) in the FM phase of reentrant ferromagnets. The existence of the broad peak around 5.5 K suggests the frustrated nature of the FM phase in our system. The drastic increase of t_{cr} (or τ) below T_{RSG} with decreasing T is a feature common to the SG phases of typical SG systems. Note that at $T=2$ K no peak in $S(t)$ is observed for $1\times10^2\leqslant t\leqslant 6\times10^4$ sec. The value of τ is estimated as 7×10^4 sec, which is much larger than t_W. In contrast, as shown in Fig. 4(b) for $t_W=3.0\times10^4$ sec, the value of t_{cr} is larger than that of τ in the FM phase. Both t_{cr} and τ show a broad peak at T between 4 and 5 K. This peak temperature is slightly lower than that for $t_W=1.5\times10^4$ sec, suggesting that the relaxation mechanism is dependent on t_W. In the inset of

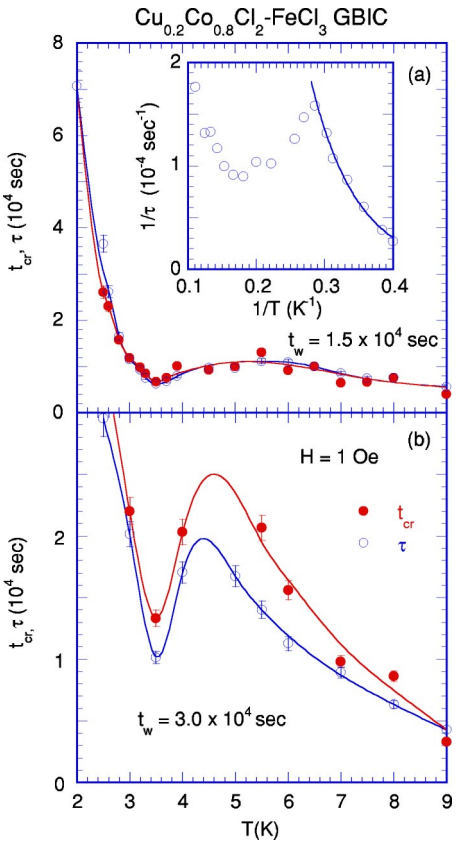

FIG. 4. (Color online) t_{cr} vs T and τ vs T. t_{cr} is a characteristic time at which $S(t)$ has a maximum S_{max}. τ is the relaxation time for the stretched exponential relaxation. $H=1$ Oe. (a) $t_W=1.5\times10^4$ sec. (b) $t_W=3.0\times10^4$ sec. The solid lines are a guide to the eye. The inset of (a) shows the plot of $1/\tau$ vs $1/T$ ($T<T_{RSG}$), where the solid line is a least-squares fit to Eq. (5). The fitting parameters are given in the text.

Fig. 4(a) we show the plot of $1/\tau$ as a function of $1/T$. In the limited temperature range ($2.5\leqslant T\leqslant 3.3$ K), $1/\tau$ can be approximated by an exponential T dependence

$$1/\tau = c_1 \exp(-c_2 T_{RSG}/T), \tag{5}$$

with $c_1=(1.16\pm0.32)\times10^{-2}$ sec^{-1} and $c_2=4.2\pm0.2$. Our value of c_2 is relatively larger than those derived by Hoogerbeets *et al.* ($c_2=2.5$) (Refs. 16 and 17) from an analysis of thermoremanent magnetization (TRM) relaxation measurements on four SG systems: Ag:Mn (2.6 at. %), Ag:Mn (4.1 at. %), Ag:[Mn (2.6 at. %)+Sb (0.46 at. %)], and Cu:Mn (4.0 at. %). Note that in their work the stretched exponential is taken as representative of the short time ($t<t_W$) relaxation. One must thus be careful in comparing the results.

Figure 5 shows the T dependence of S_{max} [the peak height of $S(t)$ at $t=t_{cr}$] for the following three cases: (i) $H=1$ Oe and $t_W=3.0\times10^4$ sec, (ii) $H=1$ Oe and $t_W=1.5\times10^4$ sec, and (iii) $H=10$ Oe and $t_W=2.0\times10^3$ sec. The peak height S_{max} at $H=1$ Oe exhibits two peaks around $T=T_{RSG}$ and at 7.0 K just below T_c, independent of t_W ($=1.5\times10^4$ sec or

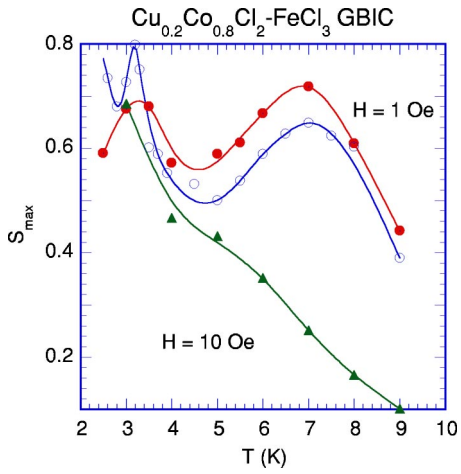

FIG. 5. (Color online) S_{max} vs T for the three cases: (i) $H=1$ Oe and $t_W=3.0\times10^4$ sec (●), (ii) $H=1$ Oe and $t_W=1.5\times10^4$ sec (○), and (iii) $H=10$ Oe and $t_W=2.0\times10^3$ sec (▲). The solid lines are a guide to the eye.

3.0×10^4 sec). Note that the peak of S_{max} at 7.0 K is strongly dependent on H: it almost disappears at $H=10$ Oe for $t_W=2.0\times10^3$ sec.

In Fig. 6(a) we show the plot of the exponent n as a function of T for $t_W=1.5\times10^4$ and 3.0×10^4 sec, where $H=1$ Oe. For $t_W=3.0\times10^4$ sec the exponent n increases with

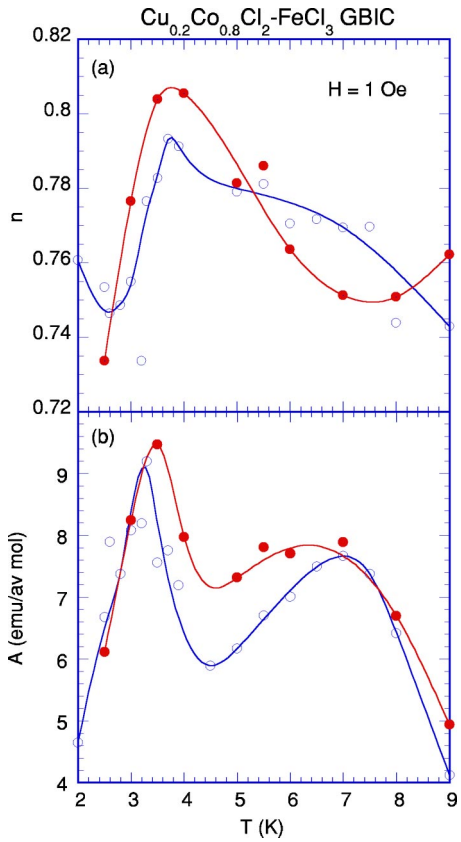

FIG. 6. (Color online) (a) n vs T and (b) A vs T, where $t_W=3.0\times10^4$ sec (●) and 1.5×10^4 sec (○). $H=1$ Oe. The solid lines are a guide to the eye.

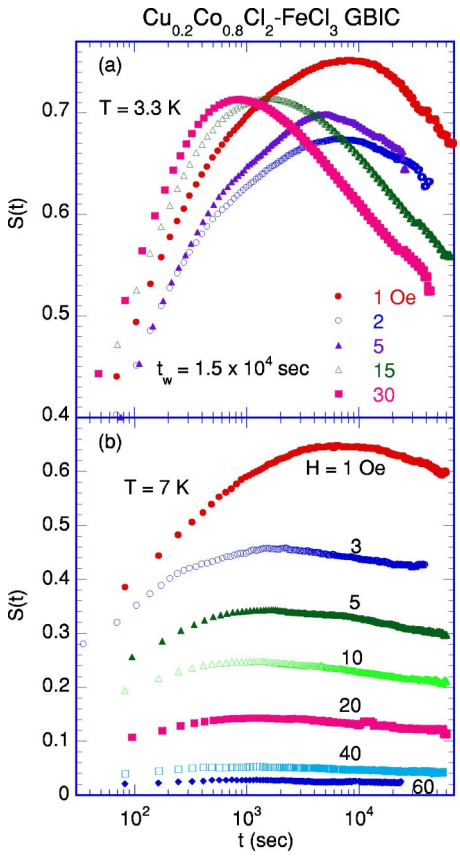

FIG. 7. (Color online) S vs t at various H. (a) $T=3.3$ K and $t_W=1.5\times10^4$ sec. (b) $T=7.0$ K and $t_W=1.5\times10^4$ sec.

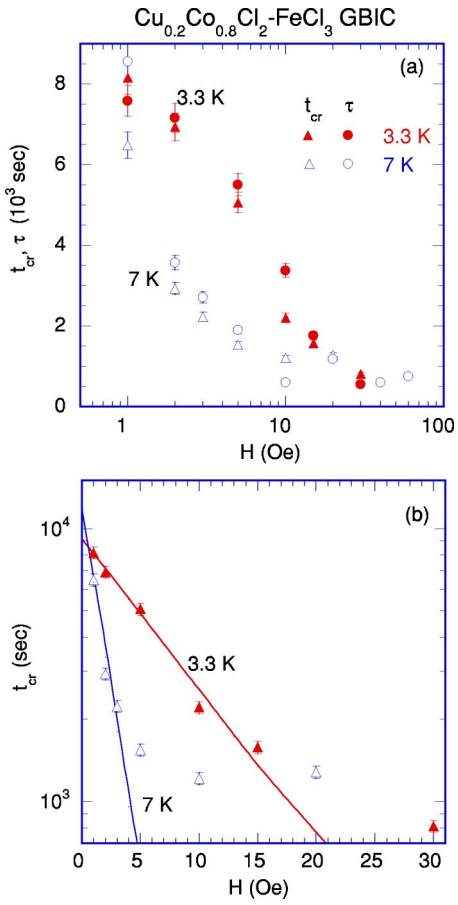

FIG. 8. (Color online) (a) t_{cr} vs H and τ vs H at $T=3.3$ and 7.0 K. (b) t_{cr} vs H at $T=3.3$ and 7.0 K. The solid lines are least-squares fits to Eq. (7) for data at low H. The fitting parameters are given in the text.

increasing T and exhibits a peak at $T=4$ K just above T_{RSG}. The exponent n decreases with further increasing T, showing a local minimum around 7.5 K. For $t_W=1.5\times10^4$ sec, the exponent n exhibits a local minimum ($n\approx0.75$) around 2.5 K ($T/T_{RSG}=2.5/3.5=0.71$) and a local maximum ($n\approx0.79$) around 4.0 K. The local minimum of n below T_{RSG} is a feature common to that of SG systems, where n has a local minimum at $T=T^*$ given by $T^*/T_{SG}\approx0.70$.[17,18] The exponent n increases as T approaches T_{SG} from the low-T side. The overall variation of n below T_{SG} is qualitatively the same as that calculated for the Sherrington-Kirkpatrick model.[30]

Figure 6(b) shows the T dependence of the amplitude A for $t_W=1.5\times10^4$ and 3.0×10^4 sec. We find that A is strongly dependent on T. Irrespective of t_W, the amplitude A shows two local maxima at 3 and 7.0 K. For comparison, we evaluate the T dependence of S^0_{max} $[=A(1-n)/e]$ with $m=0$. As the values of n and A, here we use the experimental values at each T shown in Figs. 6(a) and 6(b). We find that the T dependence of S^0_{max} thus calculated is in excellent agreement with that of S_{max} (see Fig. 5) experimentally determined from the data of $S(t)$ vs t.

C. $S(t)$ under the H shift

Figures 7(a) and 7(b) show the t dependence of $S(t)$ at various H, where $T=3.3$ and 7.0 K and $t_W=1.5\times10^4$ sec. The relaxation rate $S(t)$ shows a peak at t_{cr}. This peak greatly shifts to the short-t side with increasing H. In Fig. 8(a) we make a plot of t_{cr} and τ as a function of H at $T=3.3$ and 7.0 K, where τ is determined from the least-squares fit of the data of χ_{ZFC} vs t around $t=t_W$ to the stretched exponential relaxation with $m=0$. The value of t_{cr} is almost the same as that of τ at each T and H, indicating that the exponent m is nearly equal to zero (see Sec. II). The time t_{cr} undergoes a dramatic decrease at low fields: $H\approx2-3$ Oe at $T=7.0$ K and $H=10$ Oe at $T=3.3$ K. Note that the decrease of t_{cr} with H has been reported for the ZFC relaxation in the RSG phase of the reentrant ferromagnet $Fe_{0.3}Al_{0.7}$.[10] The increase of n and $1/\tau$ with increasing H has been reported for the TRM decay in the SG systems such as Ag:[Mn (2.6 at. %) +Sb (0.46 at. %)] (Ref. 17) and Cu:Mn (6 at. %) (Ref. 25).

The shift of t_{cr} for $S(t)$ under the H-shift process is governed by the mean domain size $L_T(t)$ and overlap lengths L_H defined by[27]

$$L_T(t)/L_0 \approx (t/t_0)^{1/z(T)}, \quad L_H/L_0 \approx (H/\Upsilon_H)^{-1/\delta}, \qquad (6)$$

respectively, where $z(T)$ and δ are the corresponding exponents for $L_T(t)$ and L_H, and Υ_H is the magnetic field corresponding to a wall stiffness Υ (a typical energy setting the scale of free energy barriers between conformations). Using

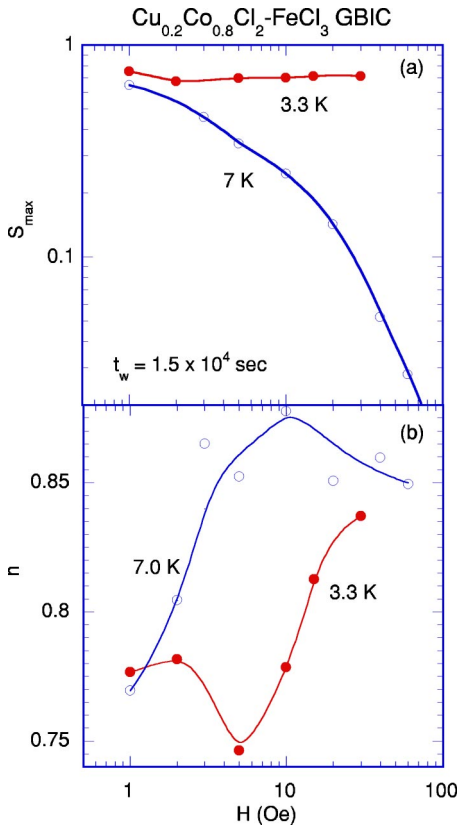

FIG. 9. (Color online) (a) S_{max} vs H and (b) n vs H at $T=3.3$ and 7.0 K. $t_W=1.5\times10^4$ sec. The solid lines are a guide to the eye.

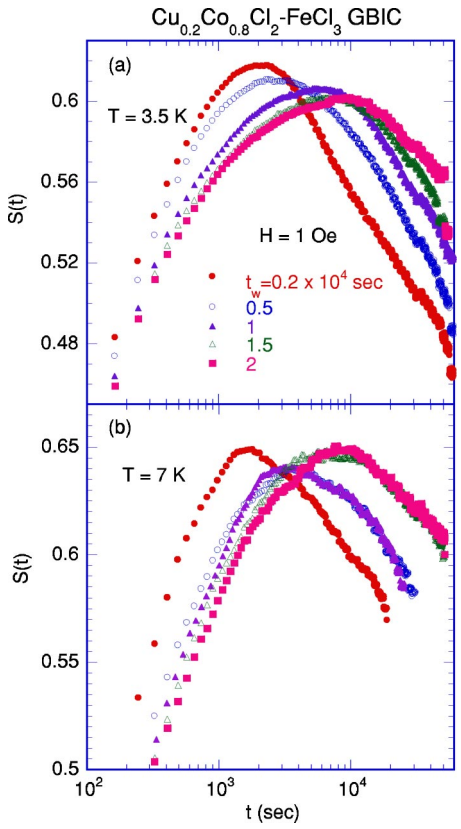

FIG. 10. (Color online) S vs t at various t_W ($2\times10^3\leqslant t_W\leqslant2.0\times10^4$ sec). $H=1$ Oe. (a) $T=3.5$ K. (b) $T=7.0$ K.

the scaling concept, the shift of t_{cr} for $S(t)$ under the H shift can be approximated by[29,31]

$$\ln(t_{cr}/t_W) = -\alpha_H H, \tag{7}$$

in the limit of $H\approx0$, where α_H is constant proportional to $z(T)\ t_W^{\delta/z(T)}$. In Fig. 8(b) we make a plot of t_{cr} vs H at $T=3.3$ and 7.0 K. We find that the data of t_{cr} vs H are well described by Eq. (7) with $\alpha_H=0.128\pm0.009$ and $t_W=(9.19\pm0.31)\times10^3$ sec for $H<15$ Oe at $T=3.3$ K and $\alpha_H=0.63\pm0.15$ and $t_W=(12.0\pm2.6)\times10^3$ sec for $H<1$ Oe at $T=7.0$ K.

In Fig. 9(a) we show the H dependence of S_{max} at $T=3.3$ and 7.0 K, where $t_W=1.5\times10^4$ sec. The value of S_{max} is almost independent of H for $0\leqslant H\leqslant40$ Oe. In contrast, the value of S_{max} at 7.0 K greatly reduces with increasing H, indicating the fragility of the aging state in the FM phase against a weak magnetic-field perturbation. For comparison, we make a plot of S^0_{max} $[=A(1-n)/e]$ as a function of H, where A and n experimentally determined are used. There is a very good agreement between S_{max} and S^0_{max}. Our result agrees well with that reported by Jonason and Nordblad[3] with the gradual decrease of S_{max} at 16 K with H and the dramatic decrease in S_{max} at 30 K with H at weak fields less than 0.2 Oe. Figure 9(b) shows the H dependence of the exponent n at $T=3.3$ and 7.0 K, where $t_W=1.5\times10^4$ sec. The H dependence of n at 7.0 K is rather different from that at 3.3 K at low H. The value of n at $T=3.3$ K has a local minimum at $H=5$ Oe, while the value of n at $T=7.0$ K has a local maximum around 10 Oe. The value of n tends to saturate to 0.82–0.84 at H higher than 30 Oe, independent of T.

D. Effect of t_W on $S(t)$

Figure 10 shows the t dependence of $S(t)$ at various t_W for $T=3.5$ and 7.0 K, where $H=1$ Oe. The relaxation rate $S(t)$ shows a peak at t_{cr}. This peak shifts to the long-t side with increasing t_W. The least-squares fit of the data of $\chi_{ZFC}(t)$ vs t for $1.0\times10^2\leqslant t\leqslant6.0\times10^4$ sec to Eq. (1) yields the exponent n, relaxation time τ, and amplitude A for the stretched exponential relaxation. Figure 11(a) shows the t_W dependence of t_{cr} and τ at $T=3.5$ and 7.0 K. At $T=7.0$ K, t_{cr} is nearly equal to τ for $0<t_W\leqslant3.0\times10^4$ sec, while at $T=3.5$ K t_{cr} is much longer than τ for $t_W\geqslant1.5\times10^4$ sec. Both t_{cr} and τ increase with increasing t_W. It has been reported that τ varies exponentially with t_W as $\tau=\tau_0\exp(t_W/t_0)$ for Ag:Mn (2.6 at. %).[17] Our data of τ vs t_W at 3.5 K are well fitted to this equation in spite of the limited t_W region ($1.0\times10^4\leqslant t_W\leqslant3.0\times10^4$ sec); $\tau_0=(3.83\pm0.012)\times10^3$ sec and $t_0=(3.07\pm0.13)\times10^4$ sec. Figure 11(b) shows the t_W dependence of n at $T=3.5$ and 7.0 K. The exponent n at $T=3.5$ K shows a local minimum at $t_W=1.0\times10^4$ sec and increases with further increasing t_W. In contrast, the exponent n at 7.0 K is smaller than that at 3.5 K for any t_W. It exhibits a local maximum around $t_W=5.0\times10^3$ sec. A similar local minimum in n vs t_W is observed in the SG phase of the spin glass $Cr_{83}Fe_{17}$ [$T_{SG}=13$ K] (Ref. 5): n at $T=8$ K shows a

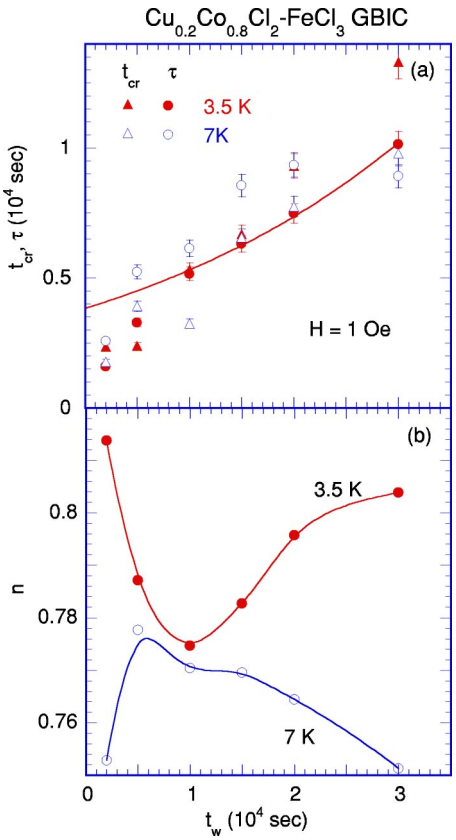

FIG. 11. (Color online) (a) t_{cr} vs t_W and τ vs t_W. The solid line is a least-squares fit to $\tau=\tau_0\exp(t_W/t_0)$ for τ at 3.5 K ($t_W \geqslant 1.0\times10^4$ sec). The fitting parameters are given in the text. (b) n vs t_W at $T=3.5$ and 7.0 K. $H=1$ Oe. The solid lines are a guide to the eye.

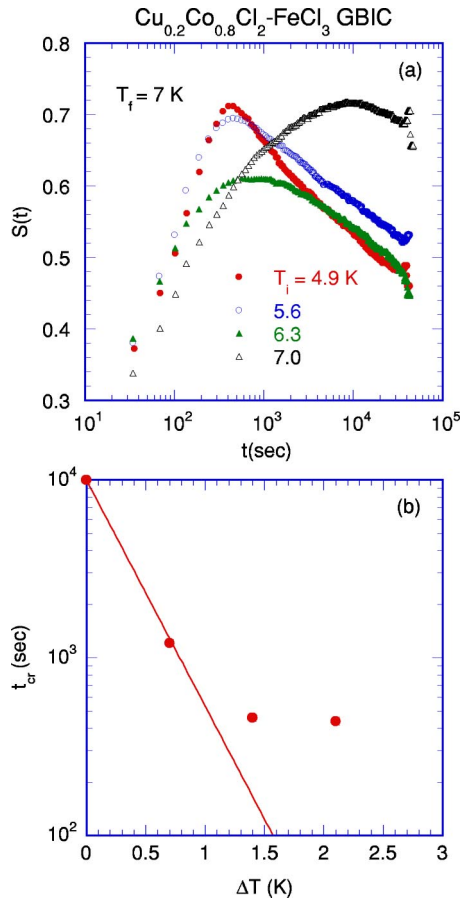

FIG. 12. (Color online) (a) t dependence of S at $T=T_f=7.0$ K under the T shift from T_i to T_f, where $T_i=4.9$, 5.6, and 6.3 K. $H=1$ Oe. The ZFC aging protocol is as follows: quenching of the system from 50 K to T_i and isothermal aging at $T=T_i$ and $H=0$ for $t_W=3.0\times10^4$ sec. The measurement was started at $t=0$, just after T was shifted from T_i to T_f and subsequently H (=1 Oe) was turned on. (b) t_{cr} vs ΔT. The solid line is a fitting curve to Eq. (9). The fitting parameters are given in the text.

local minimum around $t_W=3.0\times10^3$ sec and increases with further increasing t_W. In contrast, there is no local minimum in n vs t_W in the RSG phase of $(Fe_{0.65}Ni_{0.35})_{0.882}Mn_{0.118}$ (Ref. 6) (n increases with increasing t_W) and in the SG phase of Ag:Mn (2.6 and 4.1 at. %) (Ref. 17) (n is independent of t_W). In summary, so far there has been no theory to explain the t_W dependence of n, τ, t_{cr}, and A in both the RSG and FM phases. However, our results suggest that the RSG and FM phases are essentially nonequilibrium phases. The nature of the aging behavior in the FM phase is rather different from that in the RSG phase.

E. $S(t)$ under the T shift

Figure 12(a) shows the relaxation rate $S(t)$ measured at $T_f=7.0$ K for different initial temperature T_i (=4.9, 5.6, and 6.3 K), where $t_W=3.0\times10^4$ sec and $H=1$ Oe. The measurement was carried out as follows. After the ZFC protocol from 50 K to T_i below T_c, the system was kept at T_i for a wait time t_W. Immediately after T was changed from T_i to T_f (the positive T shift; $\Delta T>0$ where $\Delta T=T_f-T_i$), the magnetic field was applied and the t dependence of χ_{ZFC} was measured.

With increasing ΔT, the time t_{cr} shifts to the low-t side. The relaxation rate $S(t)$ has a peak at $t_{cr}=440-460$ sec at $\Delta T=2.1$ K and 1.4 K, suggesting a rejuvenation of the system during the positive T shift. For $\Delta T=0.7$ K two peaks appear at 570 sec and 1210 sec, respectively, suggesting a coexistence of two characteristic ages in the system due to the partial rejuvenation and original aging at T_i for $t_W=3.0\times10^4$ sec. This behavior is explained in terms of the overlap length $L_{\Delta T}$, which becomes smaller as ΔT becomes large. When $R_{Ti}(t_W) \ll L_{\Delta T}$ corresponding to the case of $\Delta T<\Delta T_{thresold}$, no initialization occurs in the system. There is only one type of domain. Thus $S(t)$ exhibits a peak around t_W. For $R_{Ti}(t_W) \gg L_{\Delta T}$ corresponding to the case of $\Delta T>\Delta T_{thresold}$, some domains fracture into smaller domains of dimension $L_{\Delta T}$. Thus $S(t)$ has two peaks at t_{cr} ($\approx t_W$) and a time much shorter than t_{cr}.

The shift of t_{cr} for $S(t)$ under the T-shift aging process is governed by the mean domain size $L_T(t)$ and overlap length $L_{\Delta T}$ defined by[27]

$$L_{\Delta T}/L_0 \approx (T^{1/2}|\Delta T|/\Upsilon_T^{3/2})^{-1/\zeta}, \tag{8}$$

where ζ is the corresponding exponent and Υ_T is the temperature corresponding to the wall stiffness Υ. Using the

scaling concepts, the shift of t_{cr} for $S(t)$ under the T shift can be approximated by[29,32,33]

$$\ln(t_{cr}/t_W) = -\alpha_T|\Delta T|, \tag{9}$$

in the limit of $H \approx 0$, where α_T is a constant proportional to $z(T_f)T_f^{1/2}t_W^{\zeta/z(T_i)}$. In Fig. 12(b) we make a plot of t_{cr} vs ΔT at $T_f = 7.0$ K. We find that the data of t_{cr} vs ΔT are described by Eq. (9) with $\alpha_T = 2.93 \pm 0.33$ and $t_W = (10.0 \pm 0.3) \times 10^3$ sec for $\Delta T < 0.7$ K.

Similar behaviors in $S(t)$ under the T shift have been observed in other reentrant ferromagnets. In the FM phase of $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$,[1,4] the amplitude of the maximum in $S(t)$ at $t \approx t_W$ decreases with increasing $|\Delta T|$ (both the positive and negative T shifts) and a second maximum gradually develops at a shorter time, indicating a partial rejuvenation. In the RSG phase of $(Fe_{0.65}Ni_{0.35})_{0.882}Mn_{0.118}$ double peaks in $S(t)$ vs t are observed under a temperature-cycling experiment:[6] an initial wait time t_W $(=1.0 \times 10^4$ sec) at T, followed by a temperature cycle $T \rightarrow T + \Delta T \rightarrow T$ of the duration t_{cycle} (=300 sec).

F. $\chi''(\omega, t)$

We have measured the t dependence of $\chi''(\omega, t)$ at $T = 3.3$, 3.5, 3.75, 7, and 8.5 K, where $H = 0$, $h = 0.3$ Oe, and $f = 0.05$–5 Hz. The system was quenched from 100 K to T at time (age) zero. The origin $t = 0$ is a time at which the temperature becomes stable at T within ±0.01 K. The change of $\chi''(\omega, t)$ with t below T_{RSG} is not so prominent compared to that above T_{RSG}, partly because of relatively small magnitude of χ'' below T_{RSG}. In Figs. 13(a)–13(c), we show the t dependence of $\chi''(\omega, t)$ at various f, where $T = 3.75$, 7.0, and 8.5 K. We find that the decrease of χ'' with increasing t is well described by a power-law decay of the quasiequilibrium part

$$\chi''(\omega, t) = \chi_0''(\omega) + A(\omega)t^{-\alpha}, \tag{10}$$

where $\chi_0''(\omega)$ and $A(\omega)$ are t-independent constants. The least-squares fit of the data of $\chi''(\omega, t)$ at each T to Eq. (10) yields parameters listed in Table I at $T = 3.5$, 3.75, 7, and 8.5 K. The exponent α below T_{RSG} is nearly equal to 0.07–0.084. At 7.0 K, the exponent α is dependent on f: $\alpha = 0.074$ at $f = 0.05$ Hz and $\alpha = 0.015$ at $f = 5$ Hz. The value of χ_0'' at $T = 7.0$ K tends to decrease with increasing f. Since the value of $A(\omega)$ tends to increase with increasing f, it follows that the second term of Eq. (10) does not obey the ωt-scaling law of the form $(\omega t)^{-\alpha}$.

V. DISCUSSION

A. Dynamic nature of the RSG and FM phases

The nature of the RSG and FM phases for $Cu_{0.2}Co_{0.8}Cl_2$-$FeCl_3$ GBIC is summarized as follows. The static and dynamic behaviors of the RSG phase are characterized by that of the normal SG phase: a critical exponent $\beta = 0.57 \pm 0.10$ for the SG parameter and a dynamic critical exponent $x = 8.5 \pm 1.8$ for the characteristic relaxation time.[11] The aging phenomena are clearly seen in the RSG phase, although no appreciable nonlinear magnetic susceptibility is observed.

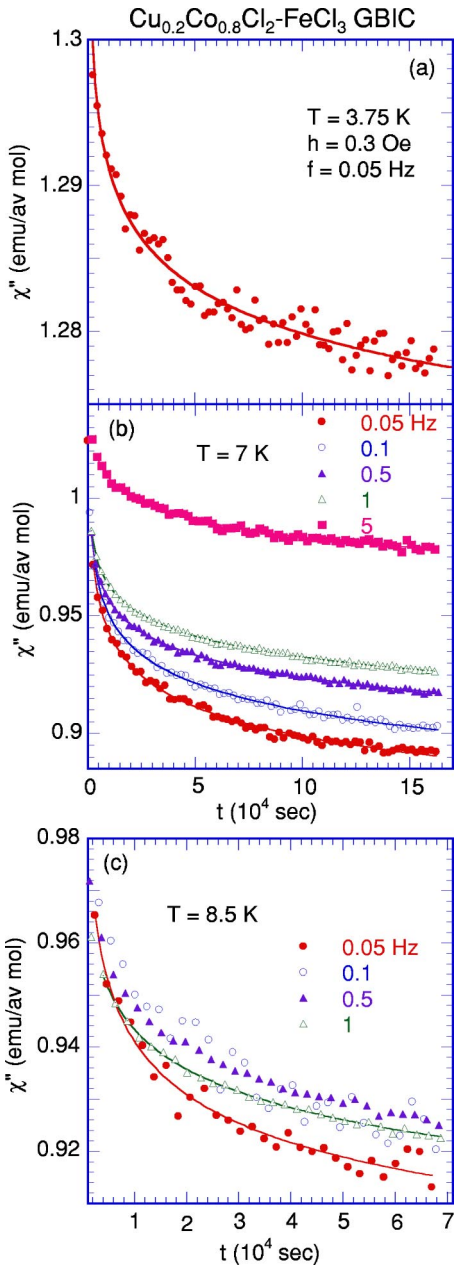

FIG. 13. (Color online) t dependence of $\chi''(\omega, t)$ at (a) $T = 3.75$ K, (b) $T = 7.0$ K, and (c) $T = 8.5$ K. $f = 0.05$, 0.1, 0.5, and 1 Hz. The time t is the time taken after the sample is quenched from 50 K to T. $H = 0$. The solid lines are the least-squares fits to Eq. (10). The fitting parameters are listed in Table I.

The exponent n for the stretched exponential relaxation exhibits a local minimum just below T_{RSG} and increases when T approaches T_{RSG} from the low-T side. The relaxation time τ for the stretched exponential relaxation drastically increases with decreasing T below T_{RSG}. In this sense, it follows that the RSG phase below T_{RSG} is a normal SG phase.

In contrast, the dynamic nature of the FM phase is rather different from that of an ordinary ferromagnet. A prominent nonlinear susceptibility is observed between T_{RSG} and T_c.[11] Aging phenomena and a partial rejuvenation effect under the T shift are also seen in the FM phase. This aging state disappears even in a weak magnetic field ($H > H_0$; H_0

TABLE I. Exponents α determined from the least-squares fits of $\chi''(\omega,t)$ at f to the power-law form given by Eq. (10). $T=3.5$, 3.75, 7, and 8.5 K.

T (K)	f (Hz)	α	A	$\chi''_0(\omega)$
3.5	0.1	0.070	0.182	1.100
3.75	0.05	0.084	0.137	1.228
7	0.05	0.074	0.334	0.662
7	0.1	0.045	0.458	0.537
7	0.5	0.042	0.386	0.606
7	1	0.029	0.505	0.489
7	5	0.015	0.770	0.265
8.5	0.05	0.147	0.148	0.835
8.5	1	0.046	0.268	0.702

$=2-5$ Oe). The relaxation time τ (or $t_{cr}\approx\tau$) as a function of T exhibits a local maximum between T_{RSG} and T_c. These results indicate the chaotic nature of the FM phase.

Similar behaviors have been observed in the reentrant ferromagnets. In $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$ (Refs. 1–4) the relaxation time diverges at a finite temperature ($\approx T_{RSG}$) with a dynamic critical exponent similar to that observed for normal SG transitions. The FM phase just above T_{RSG} shows a dynamic behavior characterized by an aging effect and chaotic nature similar to that of RSG phase. In $CdCr_{2x}In_{2(1-x)}S_4$ with $x=0.90$, 0.95, and 1.0,[7,8] the aging behavior of the low-frequency ac susceptibility is observed both in the FM and RSG phases, with the same qualitative features as in normal SG systems.

B. Scaling of $\chi_{ZFC}(t)$ and $\chi''(\omega,t)$

The t dependence of $\chi_{ZFC}(t)$ and $\chi''(\omega,t)$ arises from that of the spin autocorrelation functions $C(t_W;t+t_W)$ and $C(\Delta t_\omega;t+\Delta t_\omega)$, respectively, where $\Delta t_\omega=2\pi/\omega$.[34–37] Here we assume that $C(t_W;t+t_W)$ is defined by an addition of the quasiequilibrium term $[C_{eq}(t)]$ and aging term $[C_{ag}(t_W;t+t_W)]$.[37] In the present work, Δt_ω takes various values as $\Delta t_\omega=1/f=10^{-3}-100$ sec ($0.01\leqslant f\leqslant 1000$ Hz) for $\chi''(\omega,t)$ and as a wait time $t_W=(0.2-3.0)\times 10^4$ sec for $\chi_{ZFC}(t)$. The function $C_{eq}(t)$ is independent of t_W and is expressed by a power-law form ($\approx t^{-\alpha}$). By the appropriate choice of t and t_W (or Δt_ω), it is experimentally possible to separate the quasiequilibrium part $C_{eq}(t)$ and the aging part $C_{ag}(t_W;t+t_W)$. Our experimental results are as follows. (i) The absorption $\chi''(\omega,t)$ mainly comes from the quasiequilibrium contribution $C_{eq}(t)$. This is also supported by the fact that no ωt scaling is observed for $\chi''(\omega,t)$. The exponent α is positive and very small; typically, $\alpha=0.07$ at $T=T_{RSG}=3.5$ K in the RSG phase (see Table I). This value of α is considered to coincide with the exponent m for the stretched exponential relaxation. (ii) The susceptibility $\chi_{ZFC}(t)$ for $t\approx t_W$ and $t>t_W$ mainly comes from the aging contribution. As shown in Fig. 6, the exponent n depends on T and is 0.78 at $T\approx T_{RSG}$.

Ozeki and Ito[38] have studied the nonequilibrium relaxation of the $\pm J$ Ising model in three dimensions. They have shown that $C(t_W;t+t_W)$ obeys the power-law form $t^{-\lambda_q}$ for $t\ll t_W$, where $\lambda_q=0.070(5)$ at $T=0.92T_{SG}$. The value of λ_q is consistent with that obtained by Ogielski:[13] $\lambda_q=0.065$ at $T=T_{SG}$. The exponent λ_q at T_{SG} is described by $\lambda_q(T_{SG})=\beta/x$, where x is the dynamic critical exponent and β is the exponent of the SG order parameter. In the previous paper[11] we have reported the values of β and x as $\beta=0.57$ and $x=8.5$, leading to $\lambda_q=0.067$. This value of λ_q is in very good agreement with our value of α (=0.07) at $T=T_{RSG}$.

C. Exponents n and m of the stretched exponential relaxation

The relaxation rate $S(t)$ has a peak at $t=t_{cr}$. The value of x_{cr} ($=t_{cr}/\tau$) strongly depends on the values of n and m. The value of x_{cr} takes 1 at $m=0$ and drastically decreases with increasing m. Experimentally the value of x_{cr} is estimated from the ratio of t_{cr} to τ, where t_{cr} is the time at which $S(t)$ takes a peak and τ is from the least-squares fit of the data of χ_{ZFC} vs t around $t=t_W$ to the stretched exponential relaxation with $m=0$. The ratio x_{cr} provides a good measure to determine whether the stretched exponential relaxation is valid for our system. Experimentally we find that the ratio x_{cr} is dependent on t_W and T ($3\leqslant T\leqslant 8$ K): $x_{cr}=0.93\pm0.29$ for $t_W=1.5\times10^4$ sec and $x_{cr}=1.27\pm0.15$ for $t_W=3.0\times10^4$ sec. In our simple model, it follows that m becomes negative when $x_{cr}>1$. Furthermore, in Sec. V B, we show that $n=0.78$ and $m=0.07$ at T_{RSG}. However, these values do not satisfy the inequality ($4m+n<1$). This inequality is required for the peak of $S(t)$ to appear in the case of the stretched exponential relaxation. When $4m+n=1$, $x_{cr}=2^{-2/1-n}$, which is independent of m. The value of x_{cr} becomes zero as n tends to unity. In our system, $n=0.73-0.81$. Using the inequality $[m<(1-n)/4]$, the upper limit of m can be estimated as 0.048 for $n=0.81$ (or $1-n=0.19$).

VI. CONCLUSION

$Cu_{0.8}Co_{0.2}Cl_2$-$FeCl_3$ GBIC undergoes successive transitions at the transition temperatures T_c ($\approx$9.7 K) and T_{RSG} ($\approx$3.5 K). The FM phase of our system is characterized by the aging phenomena and nonlinear magnetic susceptibility. The relaxation rate $S(t)$ exhibits a characteristic peak at t_{cr} close to a wait time t_W below T_c, indicating the occurrence of aging phenomena in both the RSG and FM phases. The aging behavior in the FM phase is fragile against a weak magnetic-field perturbation. In the FM phase there occurs a partial rejuvenation effect in $S(t)$ under the T-shift perturbation. The time (t) dependence of $\chi_{ZFC}(t)$ around $t=t_W$ is well approximated by a stretched exponential relaxation. The relaxation time τ ($\approx t_{cr}$) exhibits a local maximum around 5 K, reflecting a frustrated nature of the FM phase. It drastically increases with decreasing temperature below T_{RSG}, as is usually seen in the SG phase of SG systems.

ACKNOWLEDGMENTS

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[1] K. Jonason, J. Mattsson, and P. Nordblad, Phys. Rev. Lett. **77**, 2562 (1996).
[2] K. Jonason, J. Mattsson, and P. Nordblad, Phys. Rev. B **53**, 6507 (1996).
[3] K. Jonason and P. Nordblad, J. Magn. Magn. Mater. **177–181**, 95 (1998).
[4] K. Jonason and P. Nordblad, Eur. Phys. J. B **10**, 23 (1999).
[5] P. D. Mitchler, R. M. Roshko, and W. Ruan, Philos. Mag. B **68**, 539 (1993).
[6] D. Li, R. M. Roshko, and G. Yang, Phys. Rev. B **49**, 9601 (1994).
[7] E. Vincent, V. Dupuis, M. Alba, J. Hammann, and J.-P. Bouchaud, Europhys. Lett. **50**, 674 (2000).
[8] E. Vincent, F. Alet, M. Alba, J. Hammann, M. Ocio, and J. P. Bouchaud, Physica B **280**, 260 (2000).
[9] V. Dupuis, E. Vincent, M. Alba, and J. Hammann, Eur. Phys. J. B **29**, 19 (2002).
[10] K. Motoya, H. Hioki, and J. Suzuki, J. Phys. Soc. Jpn. **72**, 3212 (2003).
[11] M. Suzuki and I. S. Suzuki, Phys. Rev. B **69**, 144424 (2004).
[12] L. Lundgren, in *Relaxation in Complex Systems and Related Topics*, edited by I. A. Campbell and C. Giovannella (Plenum Press, New York, 1990), p. 3.
[13] A. T. Ogielski, Phys. Rev. B **32**, 7384 (1985).
[14] G. J. M. Koper and H. J. Hilhorst, J. Phys. (Paris) **49**, 429 (1988).
[15] R. V. Chamberlin, G. Mozurkewich, and R. Orbach, Phys. Rev. Lett. **52**, 867 (1984).
[16] R. Hoogerbeets, W.-L. Luo, and R. Orbach, Phys. Rev. Lett. **55**, 111 (1985).
[17] R. Hoogerbeets, W.-L. Luo, and R. Orbach, Phys. Rev. B **34**, 1719 (1985).
[18] M. Alba, M. Ocio, and J. Hammann, Europhys. Lett. **2**, 45 (1986).
[19] L. Lundgren, P. Nordblad, and P. Svedlindh, Phys. Rev. B **34**, R8164 (1986).
[20] M. Alba, J. Hammann, M. Ocio, Ph. Refregier, and H. Bouchiat, J. Appl. Phys. **61**, 3683 (1987).
[21] P. Granberg, P. Svedlindh, P. Nordblad, L. Lundgren, and H. S. Chen, Phys. Rev. B **35**, 2075 (1987).
[22] P. Nordblad, L. Lundgren, P. Svedlindh, L. Sandlund, and P. Granberg, Phys. Rev. B **35**, 7181 (1987).
[23] R. Hoogerbeets, W.-L. Luo, and R. Orbach, Phys. Rev. B **35**, 7185 (1987).
[24] D. Chu, G. G. Kenning, and R. Orbach, Phys. Rev. Lett. **72**, 3270 (1994).
[25] D. Chu, G. G. Kenning, and R. Orbach, Philos. Mag. B **71**, 479 (1995).
[26] V. S. Zotev, G. F. Rodriguez, G. G. Kenning, R. Orbach, E. Vincent, and J. Hammann, Phys. Rev. B **67**, 184422 (2003).
[27] D. S. Fisher and D. A. Huse, Phys. Rev. B **38**, 373 (1988); **38**, 386 (1988).
[28] L. W. Bernardi, H. Yoshino, K. Hukushima, H. Takayama, A. Tobo, and A. Ito, Phys. Rev. Lett. **86**, 720 (2001).
[29] M. Suzuki and I. S. Suzuki, Eur. Phys. J. B **41**, 457 (2004).
[30] D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **32**, 1792 (1975).
[31] H. Takayama, J. Magn. Magn. Mater. **272–276**, 256 (2004).
[32] P. E. Jönsson, H. Yoshino, and P. Nordblad, Phys. Rev. Lett. **89**, 097201 (2002).
[33] P. E. Jönsson, H. Yoshino, and P. Nordblad, Phys. Rev. Lett. **90**, 059702 (2003).
[34] T. Komori, H. Yoshino, and H. Takayama, J. Phys. Soc. Jpn. **68**, 3387 (1999); **69**, 1192 (2000); **69**, Suppl. A, 335 (2000).
[35] E. Vincent, J. Hammann, M. Ocio, L.-P. Bouchaud, and L. F. Cugliandolo, in *Complex Behavior of Glassy Systems*, edited by M. Rubí and C. Pérez-Vicente (Springer-Verlag, Berlin, 1997), p. 184.
[36] M. Picco, F. Ricci-Tersenghi, and F. Ritort, Eur. Phys. J. B **21**, 211 (2001).
[37] L. Berthier and J.-P. Bouchaud, Phys. Rev. B **66**, 054404 (2002).
[38] Y. Ozeki and N. Ito, Phys. Rev. B **64**, 024416 (2001).