Nonlinear magnetic susceptibility and aging phenomena in reentrant ferromagnet: Cu$_{0.2}$Co$_{0.8}$Cl$_2$-FeCl$_3$ graphite bi-intercalation compound

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Linear and nonlinear dynamic properties of a reentrant ferromagnet Cu$_{0.2}$Co$_{0.8}$Cl$_2$-FeCl$_3$ graphite bi-intercalation compound are studied using AC and DC magnetic susceptibility. This compound undergoes successive phase transitions at the transition temperatures $T_h$ (= 16 K), $T_c$ (= 9.7 K), and $T_{RSG}$ (= 3.5 K). The static and dynamic behaviors of the reentrant spin glass phase below $T_{RSG}$ are characterized by those of normal spin glass phase with critical exponent $\beta = 0.57 \pm 0.10$, a dynamic critical exponent $x = 8.5 \pm 1.8$, and an exponent $p = (1.55 \pm 0.13)$ for the de Almeida-Thouless line. A prominent nonlinear susceptibility is observed between $T_{RSG}$ and $T_c$ and around $T_h$, suggesting a chaotic nature of the ferromagnetic phase ($T_{RSG} \leq T \leq T_c$) and the helical spin ordered phase ($T_c \leq T \leq T_h$). The aging phenomena are observed both in the RSG and FM phases, with the same qualitative features as in normal spin glasses. The aging of zero-field cooled magnetization indicates a drastic change of relaxation mechanism below and above $T_{RSG}$. The time dependence of the absorption $\chi''$ is described by a power law form ($\approx t^{-b''}$) in the ferromagnetic phase, where $b'' \approx 0.074 \pm 0.016$ at $f = 0.05$ Hz and $T = 7$ K. No $\omega t$-scaling law for $\chi'' [\approx (\omega t)^{-b''}]$ is observed.

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I. INTRODUCTION

The nature of a reentrant spin glass (RSG) phase and a ferromagnetic (FM) phase in reentrant ferromagnets has been a topic of much controversy. Spin frustration effects occur as a result of a competition between ferromagnetic interactions as a majority and antiferromagnetic interactions as a minority. As the temperature is lowered, the reentrant ferromagnet exhibits first a transition from a paramagnetic (PM) phase to a FM phase with decreasing temperature and then a second transition from the FM phase to a RSG phase. Such a reentry behavior of the reentrant ferromagnets has been extensively studied experimentally in the last two decades. There are four types of reentrant ferromagnets: (i) metallic spin glasses such as Fe$_{0.7}$Al$_{0.3}$, Fe$_{0.29}$Ni$_{0.71}$, Fe$_{1-x}$Mn$_x$P$_{16}$B$_8$Al$_3$, and Fe$_{1-x}$Mn$_x$P$_{16}$Al$_3$; (ii) insulating spin glasses such as CdCr$_2$In$_2$S$_4$ (0.90 $\leq x \leq 1$); (iii) dilute magnetic semiconductors such as Eu$_x$Si$_{1-x}$S (0.52 $\leq x \leq 0.60$); and (iv) colossal magnetoresistance materials such as Y$_{0.7}$Ca$_{0.3}$MnO$_3$. In the case of (ii) and (iii), ferromagnetic nearest neighbor interactions compete with antiferromagnetic next nearest neighbor interactions.

The nature of the RSG and FM phases in reentrant ferromagnets is basically understood in terms of a mean-field picture. The phase diagram ($T/J$ vs $J_0/J$) of the Sherrington-Kirkpatrick (SK) model with Ising spins consists of the PM phase, FM phase, and the spin glass (SG) phase, where an infinite-range Gaussian distribution of exchange interactions with variance $J$ and mean $J_0$ is assumed. For $J_0/J \leq 1$, as $T$ is lowered, the system undergoes a transition from the PM phase to the SG phase. For $J_0/J > 1$, as $T$ is lowered, the system undergoes a PM-FM transition followed by a FM-SG transition. This SG phase for $J_0/J > 1$ is called a RSG phase. However, the nature of the RSG phase is essentially the same as that of the normal SG phase for $J_0/J \leq 1$. When the Parisi’s solution for the SK model is discovered, the vertical phase boundary at $J_0/J = 1$ is added to the SK phase diagram. For $J_0/J > 1$, consequently the whole RSG phase and a part of the FM phase in the SK model are newly replaced by a RSG phase with replica symmetry breaking (RSB). This RSG phase is very different from the normal SG phase for $J_0/J \leq 1$. It is a mixed phase of the SG phase and the FM phase.

In the mean-field picture, a true reentrance from the FM phase to the normal SG phase is not predicted. There is a normal FM long-range order in the FM phase. This picture, which assumes infinite-range interactions, is not always appropriate for real reentrant magnets where the short-range interactions are large and random in sign and the spin symmetry is rather Heisenberg-like than Ising-like. Neutron scattering studies on (Fe$_{1-x}$Mn$_x$)$_7$P$_{16}$B$_8$Al$_3$ have questioned the existence of a true long-range order even in the FM phase. Aeppli et al. have proposed a phenomenological random-field picture to explain their result. In this picture, the system in the FM phase consists of regions which would order ferromagnetically and other regions forming PM clusters. The frustrated spins in the PM clusters can generate random molecular fields, which act on the unfrustrated spins in the infinite FM network. In the FM phase well above $T_{RSG}$, the fluctuations of the spins in the PM clusters are so rapid that the FM network is less influenced by them and their effect is only to reduce the
net FM moment. On decreasing the temperature toward $T_{RSG}$, the thermal fluctuations of the spins in the PM clusters become slower. The coupling between the PM clusters and the FM network becomes important and the molecular field from the slow PM spins acts as a random magnetic field. This causes breakups of the FM network into finite-sized clusters. Below $T_{RSG}$, the ferromagnetism completely disappears, leading to a SG phase. This picture is very different from the mean-field picture. The RSG phase is not a mixed phase but a normal SG phase.

Jonason et al. have shown from a dynamic scaling analysis of low-field dynamic susceptibility of (Fe$_{0.20}$Ni$_{0.80}$)$_{75}$P$_{18}$B$_{6}$Al$_{3}$ that there is a spin-glass relaxation time which diverges at a finite temperature with a dynamic critical exponent similar to that observed for an ordinary PM-SG transition. This suggests that the RSG phase is a normal SG phase. The FM phase just above $T_{RSG}$ is a reentrant ferromagnet Cu$_{2}$Co$_{0.8}$Cl$_{2}$-FeCl$_{3}$ GIC. The magnetic properties of the RSG phase and the FM phase are examined from the linear and nonlinear AC magnetic susceptibility, the magnetizations in the zero-field cooled and field cooled states, and the AC susceptibility, the field cooled susceptibility (FCC) from 1.9 to 20 K. After annealing of sample for 10 minutes at 100 K in the presence of $H$, $\chi_{FCC}$ was measured with decreasing $T$ from 20 to 1.9 K. (ii) AC susceptibility measurement. The frequency ($f$), magnetic field, and temperature dependence of the dispersion ($\Theta''/h$) and absorption ($\Theta'/h$) was measured between 1.9 to 20 K, where the frequency of the AC field is $f = 0.01 - 1000$ Hz and the amplitude $h$ is typically $h = 1$ mOe - 4.2 Oe.

III. RESULT

A. Nonlinear AC susceptibility: $\Theta'/h$ and $\Theta''/h$

We have measured the dispersion $\Theta'/h$ and absorption $\Theta''/h$ at fixed $T$ as a function of $h$, where $1$ mOe $\leq h \leq 4.2$ Oe and $f = 1$ Hz. Both $\Theta'/h$ and $\Theta''/h$ are strongly dependent on $h$, suggesting the existence of nonlinear magnetic susceptibility. We determine the $T$ dependence of nonlinear AC magnetic susceptibilities ($\chi_3$, $\chi_5$, $\chi_3''$, $\chi_5''$) from the least squares fits of the data to the power law.
FIG. 1: $T$ dependence of (a) $\Theta'_1/h$ and (b) $\Theta''_1/h$ at various $h$ for Cu$_{0.2}$Co$_{0.8}$Cl$_2$-FeCl$_3$ GBIC. $h \perp c$. $f = 1$ Hz. $H = 0$.

forms:

$$\Theta'_1/h = \chi'_1 + 3\chi'_3 h^2/4 + 10\chi'_5 h^4/16 + \cdots,$$

and

$$\Theta''_1/h = \chi''_1 + 3\chi''_3 h^2/4 + 10\chi''_5 h^4/16 + \cdots.$$  (1)

Note that for convenience we use the linear AC susceptibility $\chi'$ and $\chi''$ instead of $\chi'_1$ and $\chi''_1$. In Figs. (a) and (b) we show the $T$ dependence of the dispersion $\Theta'_1/h$ and the absorption $\Theta''_1/h$ at $f = 1$ Hz. The different curves correspond to different amplitudes of the AC field, where $h$ is varied between 1 mOe and 4.2 Oe. The susceptibility $\Theta'_1/h$ and $\Theta''_1/h$ are independent of $h$ for $h < h_0$ within experimental error, where $h_0 \approx 0.1$ Oe, implying that $\Theta'_1/h$ and $\Theta''_1/h$ coincide with the linear susceptibilities $\chi'$ and $\chi''$, respectively. Note that $\Theta''_1/h$ shows a sharp peak at 4 K, which is almost independent of $h$. This sharp peak is associated with the transition between the FM and RSG phases: $T_{RSG} = 3.5$ K.

FIG. 2: $T$ dependence of (a) $\Delta(\Theta'_1/h)$ and (b) $\Delta(\Theta''_1/h)$ at various $h$. $f = 1$ Hz. $H = 0$. $\Delta(\Theta'_1/h)$ is defined by the difference between $\Theta'_1/h$ at $h (h > 30$ mOe) and $\Theta'_1/h$ at $h = 30$ mOe. $\Delta(\Theta''_1/h)$ is defined by the difference between $\Theta''_1/h$ at $h (h > 30$ mOe) and $\Theta''_1/h$ at $h = 30$ mOe.

In Figs. (a) and (b) we show the $T$ dependence of $\Delta(\Theta'_1/h)$ and $\Delta(\Theta''_1/h)$ at various $h$, where $\Delta(\Theta'_1/h)$ and $\Delta(\Theta''_1/h)$ are defined as the differences between $\Theta'_1/h$ and $\Theta''_1/h$ at $h (h > 30$ mOe) and those at $h = 30$ mOe, respectively: $\Delta(\Theta'_1/h) = 3\chi'_3 h^2/4 + 10\chi'_5 h^4/16 + \cdots$ and $\Delta(\Theta''_1/h) = 3\chi''_3 h^2/4 + 10\chi''_5 h^4/16 + \cdots$. The differences $\Delta(\Theta'_1/h)$ and $\Delta(\Theta''_1/h)$ are strongly dependent on $h$. The difference $\Delta(\Theta''_1/h)$ has two local maxima at 16.2 K (15.8 K) and 8.6 K (7.8 K), and a local minimum at 10.3 K (10.1 K) at $h = 1$ Oe ($h = 4$ Oe). In contrast, $\Delta(\Theta'_1/h)$ has two local maxima at 15.8 K (15.6 K) and 8.3 K (7.8 K), and two local minima at 9.9 K (9.5 K) and 16.6 K (16.5 K) at $h = 1$ Oe ($h = 4$ Oe). In summary, the positions of the local maxima and minima shift to low-$T$ side as $h$ increases.

In Figs. (a) and (b) we show the plot of $\Delta(\Theta'_1/h)$ and $\Delta(\Theta''_1/h)$ as a function of $h^2$ near $T_c (= 9.7$ K) and $T_h$.
(= 16 K), respectively. Both $\Delta(\Theta_i'/h)$ and $\Delta(\Theta_1'/h)$ are strongly dependent on $h^2$ in these limited temperature regions. A linear increase of $\Delta(\Theta_1'/h)$ and $\Delta(\Theta_i'/h)$ with $h^2$ at low $h$ indicates a positive sign of $\chi'_i$ and $\chi''_i$. We find a peak in $\Delta(\Theta_i'/h) [\Delta(\Theta_i''/h)]$ at a peak field $h'_p$ [$h''_p$], which shifts to the low-$h$ side with increasing $T$. The fields $h'_p$ and $h''_p$ are defined as $d(\Theta_1'/h)/dh = 0$ and $d(\Theta_i'/h)/dh = 0$, respectively: $h'_p \approx -3\chi'_i/(5\chi'_5)$ and $h''_p \approx -3\chi''_i/(5\chi''_5)$. The existence of $h'_p$ and $h''_p$, which decrease with increasing $T$, tend to reduce to zero around 9.8 and 10.4 K, respectively.

The least squares fits of the data ($\Theta_i'/h$ vs $h^2$ and $\Theta_i''/h$ vs $h^2$) at each $T$ for 30 mOe $\leq h \leq 0.7$ Oe to Eqs. (1) and (2) yield the values of $\chi'_1$, $\chi'_3$, $\chi''_5$, $\cdots$, and $\chi''_1$, $\chi''_3$, $\chi''_5$, $\cdots$. Figure 4 shows the $T$ dependence of $\chi'_3$, $\chi'_5$, $\chi''_3$, and $\chi''_5$ thus obtained. The nonlinear susceptibility $\chi''_3$ at $f = 1$ Hz shows a positive peak at 16.0 K, a negative local minimum at 10.2 K, and two positive peaks at 8.8 and 8.3 K. The sign of $\chi''_3$ changes from negative to positive at 9.65 K and from positive to negative at 4.0 K with decreasing $T$. No anomaly is observed below 4 K. In contrast, $\chi'_5$ at $f = 1$ Hz exhibits a negative local minimum at 16.0 K, a positive peak at 10.2 K, and two negative peaks at 9.0 K and 8.3 K. The sign of $\chi'_5$ changes from positive to negative at 4.0 K with decreasing $T$. We note that the $T$ dependence of $\chi'_3$ around 10 K in our system is similar to that in stage-2 CoCl$_2$ GIC which magnetically behave like a quasi 2D ferromagnet with an extremely weak antiferromagnetic interplanar exchange interaction. In stage-2 CoCl$_2$ GIC, $\chi'_3$ exhibits a negative local minimum at 10.5 K, becomes positive below 10.2 K, and shows a positive peak below the upper critical temperature $T_{cu}$. The nonlinear susceptibility $\chi''_3$ shows a positive peak at 16.2 K, a negative local minimum at 11.0 K, and a positive peak at 8.7 K. The sign of $\chi''_3$ changes from negative to positive at 9.9 K and from positive to negative
at 5.7 K with decreasing $T$. In contrast, $\chi''_3$ shows a negative local minimum at 16.4 K, a positive peak at 11.0 K, and a negative peak at 9.0 K. The sign of $\chi''_3$ changes from negative to positive at 6.5 K with decreasing $T$.

Here we discuss the $T$ dependence of $\chi'_3$. Basically the singularity of $\chi'_3$ could reflect the breaking of spatial magnetic symmetry. The sign of $\chi'_3$ is negative for the PM phase and the SG phase and positive for the FM phase. Thus the critical temperatures $T_c$ for the PM-FM transition and $T_{RSG}$ for the FM-RSG transition could be defined as temperatures at which the sign of $\chi'_3$ changes. Using this definition, in fact we find 9.65 K as $T_c$ and 4.0 K as $T_{RSG}$ at $f = 1$ Hz for our system. Similar behaviors have been reported in other reentrant magnets. Sato and Miyako have reported that the sign of $\chi'_3$ in (Pd$_{0.9966}$Fe$_{0.0034}$)$_{0.95}$Mn$_{0.05}$ changes from negative to positive at $T_c$. Sato et al. have shown that the sign of $\chi'_3$ in Ni$_{77.5}$Fe$_1$Mn$_{22}$ changes from positive to negative at $T_{RSG}$: $\chi'_3$ has a small negative local minimum near $T_{RSG}$.

### B. Linear AC susceptibility: $\chi'$ and $\chi''$

Figures (a) and (b) show the $T$ dependence of the linear AC susceptibility $\chi'$ and $\chi''$ below 12 K at $h = 50$ mOe, where $\Theta'_h/h = \chi'_3$ and $\Theta''_h/h = \chi''_3$. The absorption $\chi''$ at $f = 0.01$ Hz shows a relatively sharp peak at 3.69 K. This peak shifts to the high-$T$ side with increasing $f$: 5.10 K at $f = 1$ kHz. In contrast, the derivative $d\chi''/dT$ at $f = 0.01$ Hz shows two negative local minima at 4.0 K corresponding to $T_{RSG}$ and 10.0 K corresponding to $T_c$. The local minimum $d\chi''/dT$ vs $T$ at 4.0 K shifts to the high-$T$ side with increasing $f$, while the local minimum at 10.0 K does not shift with increasing $f$.

Figures (c) and (d) show the $T$ dependence of $\chi'$ and $\chi''$ around $T_h$ at $h = 500$ mOe, where $\Theta'_h/h \approx \chi'$ and $\Theta''_h/h \approx \chi''$. The dispersion $\chi'$ at $f = 0.01$ Hz shows peaks at 16.2 K and 8.35 K. The peak at 16.2 K slightly changes from positive to negative at 4.0 K shifts to the high-$T$ side with increasing $f$: 16.4 K at $f = 1$ kHz. Another peak at 8.35 K does not shift with increasing $f$. The dispersion $\chi'$ at $f = 0.01$ Hz has an inflection point at 3.5 K corresponding to the positive peak of $d\chi'/dT$, and another inflection point at 9.7 K corresponding to the negative local minimum of $d\chi'/dT$. The inflection point at 3.5 K shifts to the high-$T$ side with increasing $f$.

Here we assume that the singular behavior of $\chi''$ around 4 K is due to the critical slowing down associated with the FM-RSG transition. Either the peak temperatures of $\chi''$ vs $T$ and $T\chi''$ vs $T$ or the local-minimum temperature of $d\chi''/dT$ vs $T$ around 4 K coincide with a spin freezing temperature $T_f$, at a relaxation time $\tau$ ($\approx 1/\omega$). The relaxation time $\tau$ can be described by

$$\tau = \tau^*(T_f/T_{RSG} - 1)^{-\nu z},$$

where $x = \nu z$, $z$ is the dynamic critical exponent, $\nu$ is the critical exponent of the spin correlation length $\xi$, and $\tau^*$ is the characteristic time. In Fig. we show the $T$ dependence of $\tau$ thus obtained for $d\chi''/dT$, $T\chi''$ (figure of $T\chi''$ vs $T$ is not shown), and $\chi''$. The least squares fits of the data of $\tau$ vs $T$ yield $x = 8.5 \pm 1.8$, $T_{RSG} = 3.45 \pm 0.31$ K, and $\tau^* = (4.77 \pm 0.10) \times 10^{-6}$ sec for the local-minimum temperature of $d\chi''/dT$ vs $T$, $x = 12.3 \pm 1.7$, $T_{RSG} = 2.90 \pm 0.26$ K, and $\tau^* = (1.49 \pm 0.05) \times 10^{-5}$ sec for the peak temperature of $T\chi''$ vs $T$, and $x = 16.6 \pm 1.8$, $T_{RSG} = 2.27 \pm 0.25$ K, and $\tau^* = (4.67 \pm 0.10) \times 10^{-3}$ sec for the peak temperature of $\chi''$ vs $T$. The value of $x$ for $\chi''$ vs $T$ and $T\chi''$ vs $T$ is unphysically large. In contrast, the value of $x$ for $d\chi''/dT$ vs $T$ is on the same order as that ($x = 7.9$) reported by Jonason et al. for the FM-RSG transition of (Fe$_{0.20}$Ni$_{0.80}$)$_{72}$P$_{16}$B$_{6}$Al$_{3}$. A relatively good agreement between the value of $x$ for Cu$_{0.2}$Co$_{0.8}$Cl$_2$-FeCl$_3$ GBIC and the value predicted by Ogilvies for the
FIG. 6: (a), (b) $T$ dependence of $\chi'$ ($= \chi'_1$) and $\chi''$ ($= \chi''_1$) at various $f$. $h = 50$ mOe. $H = 0$. $2 \leq T \leq 12$ K. (c), (d) $T$ dependence of $\chi'$ ($\approx \chi'_1$) and $\chi''$ ($\approx \chi''_1$) at various $f$. $h = 500$ mOe. $H = 0$. $14 \leq T \leq 20$ K.

FIG. 7: $T$ dependence of the relaxation time $\tau$ which is determined from the $f$ dependence of peak temperature of $\chi''$ vs $T$, $T\chi''$ vs $T$, and $d\chi''/dT$ vs $T$. The solid lines denote the least-squares fit of the data to Eq. 3.

3D $\pm J$ Ising SG ($x = 7.9 \pm 1.0$), suggests that the FM-RSG transition in our system is dynamically similar to an ordinary PM-SG transition. Note that the value of $\tau^*$ for $d\chi''/dT$ vs $T$ is much larger than a typical value of microscopic relaxation time $\tau_0$ (typically $10^{-10} - 10^{-12}$ sec). Such a large value of $\tau^*$ has been also reported by Kleemann et al. for Co$_{0.80}$Fe$_{0.20}$/Al$_2$O$_3$ multilayers: $\tau^* = (6.7 \pm 0.4) \times 10^{-7}$ sec and $x = 10.0 \pm 3.6$. The large $\tau^*$ suggests that the PM clusters play a significant role in the FM-RSG transition. In the random-field picture, the FM phase consists of the FM region with a longer relaxation time and the PM clusters with a shorter relaxation time. On decreasing $T$ toward $T_{RSG}$, the thermal fluctuations of the spins in the PM clusters become slower. The molecular field of the slow PM spins acting as random magnetic field causes breakups of the FM network into finite-sized clusters. It is predicted that the following dynamic scaling equation is valid for the normal SG phase:

$$T\chi'' = \omega^y\Omega(\omega\tau),$$

where $\Omega(\omega\tau)$ is a scaling function of $\omega\tau$ and assumes to take a maximum at $\omega\tau = \text{constant}$. The value $y (= \beta/x)$ is a critical exponent, where $\beta$ is a critical exponent of the order parameter. The curve of $T\chi''$ vs $T$ exhibits a peak, which shifts to the high-$T$ side as $f$ increases. The peak height of $T\chi''$ increases with increasing $f$. The least squares fit of the
FIG. 8: $f$ dependence of (a) $\chi'$ and (b) $\chi''$ at various $T$. $h = 50$ mOe.

data for the peak height of $T\chi''$ vs $f$ (for $0.01 \leq f \leq 1000$ Hz) to the form of $(\approx \Delta)^y$ yields $y = 0.066 \pm 0.001$. Then the value of $\beta$ ($= xy$) is estimated as $\beta = 0.57 \pm 0.10$, where $x = 8.5 \pm 1.8$. This value of $\beta$ is similar to that ($\beta = 0.54$) for Fe$_{0.5}$Mn$_{0.5}$TiO$_3$. In summary, the nature of the FM-RSG transition is similar to that of the normal PM-SG transition. The PM clusters for the FM-RSG transition play the same role as individual spins for the PM-SG transition.

Figures 8(a) and (b) show the $f$ dependence of $\chi'(T, \omega)$ and $\chi''(T, \omega)$ at various $T$ in the vicinity of $T_{RSG}$, respectively. The absorption $\chi''(T, \omega)$ curves exhibit different characteristics depending on $T$. Above $T_{RSG}$, $\chi''(T, \omega)$ shows a peak at a characteristic frequency, shifting to the low $f$-side as $T$ decreases. Below $T_{RSG}$, $\chi''(T, \omega)$ shows no peak for $f \geq 0.01$ Hz. It decreases with increasing $f$, following a power law form $(\approx \omega^{-\alpha''})$. This is in contrast to the $f$ dependence of $\chi''$ for conventional spin glass systems such as Fe$_{0.5}$Mn$_{0.5}$TiO$_3$: $\chi''$ increases with increasing $f$. The exponent $\alpha''$ is weakly dependent on $T$: $\alpha'' = 0.083 \pm 0.004$ at 2.5 K and $\alpha'' = 0.079 \pm 0.002$ at 3 K. According to the fluctuation-dissipation theorem, the magnetic fluctuation spectrum $P(\omega)$ is related to $\chi''(T, \omega)$ by $P(T, \omega) = 2k_B T \chi''(T, \omega)/\omega$. Then $P(T, \omega)$ is proportional to $\omega^{-1-\alpha''}$, which is similar to $1/\omega$ characteristic of typical spin glass. In contrast, $\chi'(T, \omega)$ decreases with increasing $f$ above and below $T_{RSG}$: $\chi'$ exhibits a power law form $(\omega^{-\alpha'})$. The exponent $\alpha'$ is weakly dependent on $T$: $\alpha' = 0.079 \pm 0.001$ at $T = 2.5$ K and $\alpha' = 0.094 \pm 0.001$ at $T = 3.0$ K. The value of $\alpha'$ agrees well with that of $\alpha''$. Note that $\alpha''$ is related to $\chi''$ through a so called “$\pi/2$ rule”: $\chi'' = -(\pi/2)d\chi'/d\omega$ (Kramers-Kronig relation), leading to the relation $\alpha' = \alpha''$.

Here we note the $f$ dependence of $\chi''$ above 5 K (which is not shown in Fig. 8(b)). The absorption $\chi''$ increases with increasing $f$ for $5 \leq T \leq 7.2$ K. A newly small peak is added around $f = 2$ Hz for 7.3 $\leq T \leq 9.2$ K. The absorption $\chi''$ decreases with increasing $f$ for $f \leq 70$ Hz and increases with further increasing $f$ for $9.3 \leq T \leq 9.8$ K. Above 9.9 K it decreases with increasing $f$. We find that $\chi''$ for $6.1 \leq T \leq 7.3$ K can be described by a power law form $(\approx \omega^{\beta''})$ in the limited frequency range $(0.01 \leq f < 10$ Hz). The exponent $\beta''$ increases with increasing $T$: $\beta'' = 0.04 \pm 0.01$ at 6.1 K, $0.08 \pm 0.02$ at 6.7 K, and $0.12 \pm 0.01$ at 7.2 K.

C. $\chi_{FC}$, $\chi_{ZFC}$, and $\delta \chi$ ($= \chi_{FC} - \chi_{ZFC}$)

Figures 8 shows the $T$ dependence of $\chi_{FC}$ and $\chi_{ZFC}$ at various $H$, where $H$ is applied along the $c$ plane which is perpendicular to the $c$ axis. It is strongly dependent on $H$. The susceptibility $\chi_{FC}$ at $H = 0.5$ Oe decreases with increasing $T$. It has inflection points at $T = 16.4$, 9.70, and 4.0 K where $d\chi_{FC}/dT$ exhibits negative local minima. These temperatures correspond to the transition temperatures $T_h$, $T_c$, and $T_{RSG}$. In contrast, $\chi_{ZFC}$ at $H = 0.5$ Oe exhibits two peaks at 16.2 and 8.2 K, and an inflection point at 3.2 K where $d\chi_{ZFC}/dT$ shows a positive local maximum. The peaks of $\chi_{ZFC}$ at 8.2 and 16.2 K shifts to the low-$T$ side with increasing $H$. The deviation of $\chi_{ZFC}$ at $H = 0.5$ Oe from $\chi_{FC}$ starts to occur at temperatures above 18 K. The peak of $\chi_{ZFC}$ at $T_h$ is very sensitive to the application of $H$. It shifts to the low-$T$ side with increasing $H$ and disappears above 50 Oe.

Figure 10 shows the $T$ dependence of the difference $\delta \chi$ ($= \chi_{FC} - \chi_{ZFC}$) at various $H$. The difference $\delta \chi$ has three inflection points at $T_h$, $T_c$, and $T_{RSG}$, where $d(\delta \chi)/dT$ exhibits negative local minima: 16.2 K ($\approx T_h$), 9.60 K ($\approx T_c$), and 3.40 K ($\approx T_{RSG}$). The $T$ dependence of $\delta \chi$ shown in Fig. 10 is very different from standard one observed in many reentrant ferromagnets, where $\delta \chi$ reduces to zero at $T_{RSG}$. Typical examples of $\chi_{ZFC}$ vs $T$ and $\chi_{FC}$ vs $T$ have been reported for CdCr$_2$In$_{2(1-x)}$S$_x$Au$_3$Fe$_{15}$Ni$_{77}$Mn$_{23}$ and (Fe$_{0.96}$Cr$_{0.04}$)$_{15}$Ni$_{77}$Mn$_{23}$. Both inflection points of $\delta \chi$ at $T_h(H)$ and $T_h(H)$ become less pronounced with increasing $H$. Only an inflection point at $T_{RSG}(H)$ sur-
FIG. 9: (a)-(c) $T$ dependence of $\chi_{ZF C}$ and $\chi_{FC}$ at various $H$. $H \perp c$.

vives for $H \geq 100$ Oe. Note that the inflection point at $T_{RSG}(H)$ shifts to the low-$T$ side with increasing $H$: 2.8 K at $H = 100$ Oe. The $H$ dependence of $T_{RSG}(H)$ will be discussed in Sec. III E.

D. $M^*_R$ and $M^*_T$ in the IR and TR states

We have measured the magnetization $M^*$ in the ZFC, FC, IR (isothermal remnant), and TR (thermoremnant) states in the case of $H = 5$ and 15 Oe, as a function of $T$. This magnetization $M^*$ is slightly different from usual $M$, because of different methods of measurements. The measurements of $M^*$ were carried out as follows. First the sample was quenched from 298 to 1.9 K at $H = 0$. Then $H$ (5 or 15 Oe) was applied. The measurements of $M^*_{ZF C}$ and $M^*_{IR}$ were done with increasing $T$ from 1.9 to 20 K. At each $T$, $M^*_{ZF C}$ was measured at the same $H$ and then $M^*_{IR}$ was measured 100 sec later after the field was changed from $H$ to 0 Oe. Second, the sample was annealed at 100 K for 1200 sec at $H$. The measurements of $M^*_{FC}$ and $M^*_{TR}$ were done with decreasing $T$ from 20 to 1.9 K. At each $T$, $M^*_{FC}$ was measured at $H$ and then $M^*_{TR}$ was measured 100 sec later after the field was changed from $H$ to 0 Oe.

Figure 11 shows the $T$ dependence of $M^*_{ZF C}$, $M^*_{IR}$, $M^*_{FC}$, and $M^*_{TR}$, $\delta M^* (= M^*_{FC} - M^*_{ZF C})$, $\Delta M^* (= M^*_{TR} - M^*_{IR})$ in the case of $H = 5$ and 15 Oe. Note that the $T$ dependence of $M^*_{FC}$ is not exactly the same as that of $\chi_{FC}$, because of the difference in the method of field cooling. While $\chi_{FC}$ shown in Fig. 9(a) increases with decreasing $T$, $M^*_{FC}$ shows a peak at 7.5 K between $T_c$ and $T_{RSG}$ and an inflection point at $T_{RSG}$ where $dM^*_{FC}/dT$ exhibits a positive local maximum. This result is indicative of a non-uniformity in the FM phase: frustrated spins coexists with ferromagnetically aligned spins.

The $T$ dependence of $M^*_{TR}$ is exactly the same as that of $\delta M^*$. The magnetization $M^*_{TR}$ (measured at $H = 0$) in the case of $H = 5$ Oe shows a broad peak around 10.6 K just above $T_c$, and reduces to zero at $T_h$. In contrast,
The magnetization \( M_0 \) at various \( T \) and a broad peak between \( T \) measured as \( T \) on \( M \) is plotted as a function of \( H \). The magnetization \( M_0 \) is used for 1.9 \( \leq T \leq 12 \) K and then reducing \( H \) to zero. The magnetization \( M_0 \) shows a local minimum at \( T_{\text{RSG}} \) and a broad peak between \( T_{\text{RSG}} \) and \( T_c \). It reduces to zero at \( T_c \) with increasing \( T \).

As far as we know, there has been only one report on \( M_{TR} \) (measured by a conventional method) in a reentrant ferromagnet (Fe\(_{90.05}\)C\(_{0.05}\)Ni\(_{0.05}\))\(_2\)P\(_{34}\). \( M_{TR} \) was measured as \( T \) increases after cooling down the system to the lowest \( T \) in the presence of \( H \) (FC cooling), annealing at \( T \) at a waiting time \( t_w \), and then reducing \( H \) to zero. The magnetization \( M_{TR} \) shows a local minimum at \( T_{\text{RSG}} \) and a broad peak between \( T_{\text{RSG}} \) and \( T_c \). It reduces to zero at \( T_c \) with increasing \( T \).

\( M_{IR} \) is much smaller than \( M_{TR} \). The difference \( \delta M^* \) is different from \( \Delta M^* \) between \( T_{\text{RSG}} \) and \( T_c \). In fact, the difference \( (\delta M^* - \Delta M^*) \) is larger than zero only between \( T_{\text{RSG}} \) and \( T_c \) and near \( T_h \).

FIG. 11: \( T \) dependence of \( M_{ZF C}^* \), \( M_{FC}^* \), \( M_{IR}^* \), \( M_{TR}^* \), \( \delta M^* \) \( (= M_{IR}^* - M_{ZF C}^*) \) and \( \Delta M^* \) \( (= M_{TR}^* - M_{IR}^*) \). \( H \perp c \). (a) \( H = 5 \) Oe and (c) \( H = 15 \) Oe. The definition of \( M^* \) for each state is given in the text.

\( E. \ H-T \) phase diagram

We have measured the \( T \) dependence of \( \chi' \) and \( \chi'' \) at various \( H \) for \( f = 100 \) Hz and 0.1 Hz, where \( H \) \( (0 < H \leq 2 \) kOe) is applied along the \( c \) plane perpendicular to the \( c \)-axis. Figures 12 shows the \( T \) dependence of \( \chi' \) and \( \chi'' \) at various \( H \) for \( f = 100 \) Hz. The AC field \( h \) \( (50 \) mOe) was used for \( 1.9 \leq T \leq 12 \) K and a larger \( h \) \( (500 \) mOe) was used for \( 14 \leq T \leq 19 \) K. The peak of \( \chi' \) and the shoulder of \( \chi'' \) around \( T_c \) disappears for \( H \geq 50 \) Oe, and the peaks of \( \chi' \) and \( \chi'' \) around \( T_h \) disappears for \( H \geq 7 \) Oe. The peak of \( \chi'' \) around \( T_{\text{RSG}} \) shifts to the low \( T \)-side with increasing \( H \) for \( 0 \leq H \leq 1 \) kOe. The peak of \( \chi'' \) around \( T_h \) also shifts to the low \( T \)-side with increasing \( H \) for \( 0 \leq H \leq 7 \) Oe. In Figs. 13(a) and (b) we show the \( H-T \) diagrams around \( T_{\text{RSG}} \) and \( T_h \), respectively. Here the temperature (denoted as \( T_f(H) \)) of negative local minimum of \( d\chi''/dT \) vs \( T \) at 0.1 Hz (data are not shown) is plotted as a function of \( H \). For comparison, we also show the \( H \) dependence of the negative local minimum temperatures of \( d\chi''/dT \) vs \( T \) and the peak temperatures of \( \chi'' \) vs \( T \) at \( f = 0.1 \) and 100 Hz. These lines are away from the line \( T_f(H) \). The least squares fit of the data of the line \( T_f(H) \) for \( 0 \leq H \leq 600 \) Oe to

\[ H = H_0[1 - T_f(H)/T_g]^p, \]

yields parameters \( p = 1.55 \pm 0.13 \), \( T_g = 4.26 \pm 0.11 \) K, and \( H_0 = (2.6 \pm 0.3) \) kOe. Note that the value of \( T_g \) \( (4.26 \) K) is larger than that of \( T_{\text{RSG}} \) \( (3.45 \pm 0.31 \) K). The value of exponent \( p \) is close to that \( (p = 1.50) \) for the de Almeida-Thouless (AT) line. In the mean field picture, the AT line separates the replica-symmetry (FM) phase from the replica-symmetry-breaking (RSB) phase in the \((H,T)\) line.
FIG. 12: $T$ dependence of (a) $\chi'$ and (b) $\chi''$ at various $H$. $H$ is applied along the $c$ plane perpendicular to the graphene plane ($H \perp c$). $f = 100$ Hz. $h = 50$ mOe. $h \perp c$. $1.9 \leq T \leq 12$ K. $T$ dependence of (c) $\chi'$ and (d) $\chi''$ at various $H$. $H \perp c$. $f = 100$ Hz. $h = 500$ mOe. $h \perp c$. $14 \leq T \leq 19$ K.

F. Aging: $M_{ZFC}(t,T)$, $\chi''(t,\omega)$, and $\chi'(t,\omega)$

In order to reveal a possible aging phenomenon in the RSG phase and FM phase, we have studied time ($t$) dependence of the zero-field cooled magnetization $M_{ZFC}$. First the system was cooled from 100 K to $T$ in the absence of $H$, where the remnant magnetic field is less than 3 mOe. The system was kept at $T$ for a waiting time $t_w (= 2.0 \times 10^3$ sec). A DC magnetic field ($H = 10$ Oe) was applied at $t = 0$. The magnetization ($M_{ZFC}$) was measured as a function of $t$ elapsed after the field application. Figure 13 shows the $t$ dependence of $M_{ZFC}(t)$ at $T = 3$ and 7 K. The corresponding relaxation rate $S(t) [= (1/H) dM_{ZFC}(t)/d\ln t]$ is shown in Fig. 14 as a function of $t$ for each $T$. The relaxation rate $S(t)$ at $T = 3$ K has a peak around $t_p = 4 \times 10^3$ sec which is larger than $t_w$. Note that the inflection point of $M_{ZFC}$ corresponds to the peak of $S(t)$. At $T = 4$ K, $S(t)$ shows a very flat plateau between $3.2 \times 10^3$ and $10^4$ sec, in addition to a small peak at $t_p = 1.1 \times 10^4$ sec. Such an aging behavior does not end at $T_{RSG}$, but sustains into the FM phase. The relaxation rate $S(t)$ has a peak at a time shorter than $t_w$ for $5 \leq T \leq 7$ K: $t_p = 1.6 \times 10^3$ sec at $T = 5$ K, $1 \times 10^3$ sec at 6 K, and $1.3 \times 10^3$ sec at 7 K. The peak time $t_p$ is equal to $2.1 \times 10^3$ sec at 8 K and $1.8 \times 10^3$ sec at 9 K, which are close to $t_w$. Similar behavior is observed in the FM phase of (Fe$_{0.20}$Ni$_{0.80}$)$_{75}$P$_{16}$B$_8$Al$_3$ by Jonason et al.. $S(t)$ has a peak around $t_w (= 100 - 10^4$ sec).

The relaxation rate $S(t)$ clearly shows a crossover between two asymptotic relaxation regimes: the peak time $t_p$ in the RSG phase is much longer than that in the FM phase under the same value of $t_w$. The FM phase of our system is chaotic in a similar way as the RSG

| $T(K)$ | $a$ | $M'_1$ | $M'_2$ | $M_{FC}$ | $M_{ZFC}$ |
|--------|-----|--------|--------|----------|----------|
| 5      | 0.015 ± 0.001 | 461.6 | 290.3 | 414.4 | 187.1 |
| 6      | 0.035 ± 0.001 | 296.0 | 99.0 | 399.8 | 214.6 |
| 7      | 0.030 ± 0.001 | 292.8 | 82.4 | 374.1 | 231.1 |
| 8      | 0.059 ± 0.001 | 232.4 | 26.6 | 373.7 | 229.4 |
| 9      | 0.145 ± 0.005 | 140.6 | 6.0 | 221.5 | 155.6 |

TABLE I: Least squares fitting parameters of $M_{ZFC}(t)$ to the power law form given by Eq. (6). $H = 10$ Oe. $t_w = 2.0 \times 10^3$ sec. The values of $M_{FC}$ and $M_{ZFC}$ are obtained from Fig. 13(b) at $H = 10$ Oe. $M'_1$, $M'_2$, $M_{FC}$ and $M_{ZFC}$ are in the units of emu/av mol.
FIG. 13: (a) $H$-$T$ phase diagram near $T_{RSG}$: for each $H$ the local-minimum temperatures of $d\chi''/dT$ vs $T$ at $f = 0.1$ Hz (●) and at 100 Hz (■), and $d\delta\chi/dT$ vs $T$ at $f = 0.1$ Hz (▲) and 100 Hz (○). The solid line denotes the least squares fitting curve (see the text for detail). (b) $H$-$T$ phase diagram near $T_h$: for each $H$ the local-minimum temperature of $d\delta\chi/dT$ vs $T$ (◦), and the peak temperatures of $\chi'\prime\prime$ vs $T$ at 0.1 Hz (▲) and 100 Hz (○).

phase. This is in contrast to the nonfrustrated nature of regular FM phase. The relaxation mechanism in the FM phase is different from that in the RSG phase in our system. The relaxation of $M_{ZFC}(t)$ at $T = 3$ and 4 K is described by the superposition of a stretched exponential and a constant:

$$M_{ZFC}(t) = M_1 - M_2 \exp \left[ - (t/\tau_M)^{1-n} \right]. \quad (5)$$

where $\tau_M$ is a relaxation time, $M_1$ and $M_2$ are constants, the exponent $n = 0$ corresponds to the Debye, single time-constant exponential relaxation and $n = 1$ corresponds to $t$-independent $M_{ZFC}$. The least squares fit of the data to Eq. (5) yields $n = 0.761 \pm 0.002$, $\tau_M = (6.20 \pm 0.05) \times 10^3$ sec, $M_1 = 108.72 \pm 0.17$ (emu/av mol), $M_2 = 76.93 \pm 0.53$ (emu/av mol) for $T = 3$ K, and $n = 0.819 \pm 0.001$, $\tau_M = (6.49 \pm 0.04) \times 10^3$ sec, $M_1 = 171.53 \pm 0.11$ (emu/av mol), $M_2 = 70.28 \pm 0.33$ (emu/av mol) for $T = 4$ K. In contrast, the relaxation of $M_{ZFC}(t)$ for $5 \leq T \leq 9$ K can be well described by a simple power law form and a constant:

$$M_{ZFC}(t) = M_1' - M_2' t^{-a}, \quad (6)$$

where $a$ is the exponent, and $M_1'$ and $M_2'$ are constant magnetizations. The magnetization $M_1'$ is the saturation value which $M_{ZFC}$ reaches in the limit of $t \to \infty$. The least squares fit of the data to Eq. (6) yields the parameters listed in Table I. The exponent $a$ is roughly on the same order as that reported by Li et al. for a reentrant ferromagnet ($\text{Fe}_{0.65}\text{Ni}_{0.35}/\text{Mn}_{0.118}$) ($a = 0.088 - 0.060$ for $T_c < T < T_{RSG}$). However, the value of $a$ in our system tends to increase with increasing $T$ between $T_{RSG}$...
and $T_c$, while the value of $a$ in $(\text{Fe}_{0.65}\text{Ni}_{0.35})_{0.83}\text{Mn}_{0.18}$ decreases with increasing $T$. We note that the $T$ dependence of $a$ in our system near $T_c$ is similar to that in $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$ near $T_{SG}$, in spite of the fact that $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$ is a pure spin glass and undergoes a transition between the SG phase and the PM phase at $T_{SG}$. The values of $M_{FC}$ and $M_{ZFC}$ measured at $H = 10$ Oe (see Fig. 16b) are also listed in Table II. The value of $M_{ZFC}$ is on the same order as that of $M_{FC}$ and much larger than that of $M_{ZFC}$, where $M_{FC}$ is close to thermodynamic equilibrium value. The prefactor $M_2$ decreases with increasing $T$ and tends to reduce to zero around $T_c$.

We have measured the $t$ dependence of $\chi''(t, \omega)$ at $T = 7$ and 8.5 K, where $H = 0$. The system was quenched from 100 K to $T$ at time (age) zero. Both $\chi'$ and $\chi''$ were measured simultaneously as a function of time $t$ at constant $T$.

Each point consists in the successive measurements at various frequencies ($0.05 \leq f \leq 1$ Hz). Figure 16 shows the $t$ dependence of $\chi''$ at $T = 7$ K for $f = 0.05$ and 1 Hz, respectively. The absorption $\chi''$ decreases with increasing $t$ and is well described by a power-law decay

$$\chi''(t, \omega) = \chi''_0(\omega) + A''_0(\omega)t^{-b''},$$

where $b''$ is an exponent, and $\chi''_0(\omega)$ and $A''_0(\omega)$ are $t$-independent constants. In the limit of $t \to \infty$, $\chi''(t, \omega)$ tends to $\chi''_0(\omega)$. The least squares fit of the data of $\chi''(t, \omega)$ at $T = 7$ K to Eq. (7) yields parameters listed in Table II. The exponent $b''$, which is dependent on $f$, is smaller than that of the FM phase of reentrant ferromagnet $\text{CdCr}_{1.8}\text{In}_{0.2}\text{S}_4$ ($b'' \approx 0.2$) and the SG phase of pure spin glass $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$ ($b'' \approx 0.14 \pm 0.03$). The value of $\chi''_0$ tends to decrease with increasing $f$. In contrast, the value of $A''$ tends to increase with increasing $f$. It follows that the second term of Eq. (7) cannot be described by a power form $(\omega t)'^{-b''}$, suggesting no $\omega t$-scaling law in the form of $\chi'' \approx (\omega t)^{-b''}$. This is in contrast to the $\omega t$-scaling of $\chi''$ in the FM phase of reentrant ferromagnet $\text{CdCr}_{1.8}\text{In}_{0.2}\text{S}_4$. The $t$ dependence of $\chi'$ can be also described by the power law form ($\approx t^{-b''}$) which is similar to Eq. (7). The value of $b''$ at $T = 7$ and 8.5 K which is listed in Table II is on the same order as that of $b''$. Note that similar aging behavior is also observed both in $\chi''$ and $\chi'$ below $T_{RSG}$. The change of $\chi''$ and $\chi'$ with $t$ below $T_{RSG}$ is not so prominent compared to that above $T_{RSG}$, partly because of relatively small magnitude of $\chi''$ and $\chi'$ below $T_{RSG}$.

\begin{table}[h]
\centering
\caption{Exponents $b''$ and $b'$ determined from the least squares fits of $\chi''(t, \omega)$ and $\chi'(t, \omega)$ at $f$ to the power law form by given by Eq. (6) for $\chi''(t, \omega)$ and the corresponding equation for $\chi'(t, \omega)$. $T = 7$ and 8.5 K.}
\begin{tabular}{cccc}
\hline
$T(K)$ & $f$(Hz) & $b''$ & $b'$ \\
\hline
7 & 0.05 & 0.074 $\pm$ 0.016 & - \\
7 & 0.1 & 0.045 $\pm$ 0.013 & 0.012 $\pm$ 0.005 \\
7 & 0.5 & 0.042 $\pm$ 0.008 & 0.045 $\pm$ 0.005 \\
7 & 1 & 0.029 $\pm$ 0.006 & 0.046 $\pm$ 0.006 \\
7 & 5 & 0.015 $\pm$ 0.025 & 0.070 $\pm$ 0.016 \\
8.5 & 0.05 & 0.147 $\pm$ 0.067 & - \\
8.5 & 0.5 & 0.065 $\pm$ 0.028 & - \\
8.5 & 1 & 0.046 $\pm$ 0.022 & 0.038 $\pm$ 0.011 \\
\hline
\end{tabular}
\end{table}
IV. DISCUSSION

The RSG phase below $T_{RSG}$ is not a mixed phase but a normal SG phase. The static and dynamic behaviors of the RSG phase are characterized by that of the normal SG phase: a critical exponent $\beta = 0.57 \pm 0.10$, a dynamic critical exponent $x = 8.5 \pm 1.8$, and an exponent $p = 1.55 \pm 0.13$ for the AT line. The aging phenomena are observed. The relaxation of $M_{ZF C}(t)$ obeys a stretched exponential law, which is usually used in analysis of the dynamics of the normal SG phase. No appreciable non-linear magnetic susceptibility is observed below $T_{RSG}$. These results suggest that the long-range ferromagnetic correlation completely disappears in the RSG phase.

In contrast, the FM phase of our system is very different from that of regular FM phase. The chaotic behavior observed in the FM phase is rather similar to that in the RSG phase. The prominent nonlinear susceptibility of the FM phase arises mainly from the unfrustrated ferromagnetically ordered spins (the FM network). The aging phenomena are also observed in the FM phase. A dynamic crossover of the relaxation of $M_{ZF C}(t)$ is observed around $T_{RSG}$. Above $T_{RSG}$, the relaxation of $M_{ZF C}(t)$ obeys a weak power law. These results can be well explained in terms of the phenomenological random-field picture proposed by Aeppli et al. (see Sec. III). The FM phase consists of the FM network (with slow dynamics) surrounded by frustrated spins (the PM clusters with fast dynamics). Such a nonuniformity gives rise to the chaotic nature of the FM phase. On approaching $T_{RSG}$ from the high-$T$ side, the thermal fluctuations of the spins in the PM clusters become so slow that the slow dynamics of the FM network is significantly influenced by the dynamics of the PM clusters. The coupling between the FM network and the PM clusters becomes important. The molecular fields from the slow PM spins act a random magnetic field. This causes breakups of the FM network in to finite-sized clusters. Below $T_{RSG}$, the system becomes into the RSG phase.

Next we discuss the nature of the aging phenomena in the FM phase. The features of the aging phenomena are summarized as follows. (i) $M_{ZF C}(t)$ has a power law form ($M_{ZF C} \approx -t^{-a}$) for $5 \leq T \leq 9$ K. The value of $a$ is listed in Table I. (ii) Both $\chi''(t, \omega)$ and $\chi'(t, \omega)$ at $T = 7$ and 8.5 K have power law forms ($\chi'' \approx t^{-b''}$ and $\chi' \approx t^{-b'}$) at fixed $f (=\omega/2\pi)$. The values of $b'$ and $b''$ are listed in Table I. (iii) $\chi''(\omega)$ for $6.1 \leq T \leq 7.3$ K has a power law form ($\chi'' \approx \omega^{\beta''}$), where $\beta'' = 0.04 \pm 0.01$ at 6.1 K and 0.12 $\pm 0.01$ at 7.2 K. The increase of $\chi''(\omega)$ with increasing $\omega$ is similar to that reported in conventional SG's such as Fe$_{0.5}$Mn$_{0.5}$TiO$_3$.

As is listed in Table I, the exponent $a(T)$ increases exponentially with increasing $T$ and is described by $a(T) = 0.23 \exp[-(1 - T/T_c)/0.148]$ with $T_c = 9.7$ K. Similar result on $a(T)$ has been reported by Ito et al. for the SG phase of Fe$_{0.5}$Mn$_{0.5}$TiO$_3$: $a(T) = 1.6 \exp[-(1 - T/T_{SG}(H))/0.17]$ with $T_{SG}(H) = 17.6$ K at $H = 3.2$ kOe. From Monte-Carlo simulations on the 3D $\pm J$ Ising spin glass model, Ogielski has discussed the $t$ dependence of the order parameter $q(t)$ below the spin freezing temperature $T_{SG}$. The order parameter $q(t)$ is described by a power law form $[q(t) \approx t^{-a(T)}]$, where $a(T)$ is well fitted by $a(T) = 0.07 \exp[-(1 - T/T_{SG})/0.28]$, $a = 0.07$ at $T = T_{SG}$. The similarity between our result and the computer simulation is remarkable. This implies that the growth of $M_{ZF C}$ in the FM phase (but not in the RSG phase) is essentially the same as that in the SG phase of spin glass systems.

What is the relation between the exponent $a$ for $M_{ZF C}(t)$ and the exponent ($b', b'', \beta''$) for $\chi'$ and $\chi''$? According to Lundgren et al. $M_{ZF C}(t)$ is related to the linear AC susceptibility $\chi''(\omega)$ and $\chi'(\omega)$ through the following relations

$$ (1/H)dM_{ZF C}/d\ln t = 2\chi''(\omega)/\pi, \quad (t = 1/\omega), \quad (8) $$

and

$$ 1 - q(t) = (1/H)M_{ZF C}(t) = \chi'(\omega), \quad (t = 1/\omega). \quad (9) $$

When $M_{ZF C}(t)$ is described by a power law form given by Eq. (9), $\chi'(\omega)$ and $\chi'(\omega)$ can be estimated as $\chi''(\omega) \approx \omega^a$ (or $t^{-a}$) and $\chi'(\omega) \approx \omega^a$ (or $t^{-a}$), respectively, leading to a prediction that the exponents $b''$, $b'$, and $b''$ are essentially the same as the exponent $a$. Experimentally we find $a = 0.059 \pm 0.001 (T = 8$ K$)$, $b'' = 0.074 \pm 0.016 (T = 7$ K$)$, $b' = 0.046 \pm 0.006 (T = 7$ K$, f = 1$ Hz$)$, and $\beta'' = 0.081 \pm 0.02 (T = 6.7$ K$)$. There are relatively large differences among $b''$, $b'$, and $a$, depending on the conditions. Nevertheless, we can say that these exponents are roughly the same and are on the order of 0.05 - 0.08 around 7 K. The value of these exponents is nearly equal to that of the exponent $y (=0.066 \pm 0.001)$ derived from the scaling analysis in Sec. III. Note that our result of $a$ is in good agreement with $a = 0.060 - 0.088$ for the FM phase of the reentrant ferromagnet (Fe$_{0.65}$Ni$_{0.35}$)$_{0.88}$Mn$_{0.12}$.

Finally we estimate the value of $y$. The exponent $y$ is given by $y = \beta/z$, where $x = \nu z$. When we use the scaling relation, $\beta = 2\nu \theta$, which is derived in our previous paper. $y$ is given by $y = 2\theta/z$, where $\theta$ is the energy exponent and $z$ is the dynamic critical exponent. The values of $\theta$ and $z$ are theoretically predicted: $z = 6.0 \pm 0.5$ for the 3D $\pm J$ Ising spin glass model (Ogielski$^{22}$) and $\theta = 0.19 \pm 0.01$ (Bray and Moore$^{5}$). Using these values, $y$ is estimated as $y = 0.063 \pm 0.017$, which is in good agreement with our result ($y = 0.066 \pm 0.001$).

V. CONCLUSION

The nonlinear susceptibility and aging phenomena are observed in the FM phase of the reentrant ferromagnet Cu$_{0.2}$Co$_{0.8}$Cl$_2$-FeCl$_3$ GBIC. These results indicate that the FM phase between $T_{RSG}$ and $T_c$ has a chaotic nature. The absorption $\chi''(t, \omega)$ is described by a power law
form $t^{-\beta''}$ but not by a $\omega t$-scaling form $(\omega t)^{-\beta''}$. A dynamic crossover behavior is observed around $T_{RSG}$. The time dependence of $M_{ZFC}$ has a stretched-exponential form for the RSG phase, and a power-law form for the FM phase. Further studies on aging behaviors including memory effect, rejuvenation, and wait time dependence, are required to understand the nature of the FM phase, with the same qualitative features as in conventional spin glasses.

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1. K. Motoya, S.M. Shapiro, and Y. Muraoka, Phys. Rev. B 28, 6183 (1983).
2. J. Suzuki, Y. Endoh, M. Arai, M. Furusaka, and H. Yoshizawa, J. Phys. Soc. Jpn. 59, 718 (1990).
3. J.A. Geoghegan and S.M. Bhagat, J. Magn. Magn. Mater. 25, 17 (1981).
4. K. Jonason, J. Mattsson, and P. Nordblad, Phys. Rev. B 53, 6507 (1996).
5. K. Jonason, J. Mattsson, and P. Nordblad, Phys. Rev. Lett. 77, 2562 (1996).
6. G. Aeppli, S.M. Shapiro, R.J. Birgeneau, and H.S. Chen, Phys. Rev. B 28, 5160 (1983).
7. T. Sato, T. Ando, T. Ogawa, S. Morimoto, and A. Ito, Phys. Rev. B 64, 184432 (2001).
8. T. Ogawa, H. Nagasaki, and T. Sato, Phys. Rev. B 65, 024430 (2001).
9. J.L. Dormann, A. Saifi, V. Cagan, and M. Nogues, Phys. Stat. Sol. (b) 131, 573 (1985).
10. M. Alba and J. Hammann, J. Magn. Magn. Mater. 54-57, 213 (1986).
11. S. Pouget and M. Alba, J. Phys. Condens. Matter 7, 4739 (1995).
12. V. Dupuis, E. Vincent, M. Alba, and J. Hammann, Eur. Phys. J. B 29, 19 (2002).
13. H. Maletta, G. Aeppli, and S.M. Shapiro, Phys. Rev. Lett. 48, 1490 (1982).
14. G. Aeppli, H. Maletta, S.M. Shapiro, and D. Abernathy, Phys. Rev. B 36, 3956 (1987).
15. P. Mathieu, P. Nordblad, D.N.H. Nam, N.X. Phuc, and N.V. Khiem, Phys. Rev. B 63, 174405 (2001).
16. D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 32, 1792 (1975).
17. S.F. Edwards and P.W. Anderson, J. Phys. F 5, 965 (1975).
18. G. Parisi, Phys. Rev. Lett. 43, 1754 (1979).
19. G. Toulouse, J. Phys. (France) Lett. 41, L447 (1980).
20. I.S. Suzuki, M. Suzuki, H. Satoh, and T. Enoki, Solid State Commun. 104, 581 (1997).