Abstract. Non-equilibrium aging dynamics in 3D Ising spin glass \textit{Cu}_{0.5}\textit{Co}_{0.5}\textit{Cl}_{2}\textit{FeCl}_{3} GBIC has been studied by zero-field cooled (ZFC) magnetization and low frequency AC magnetic susceptibility \((f = 0.05 \text{ Hz})\), where \(T_g = 3.92 \pm 0.11 \text{ K}\). The time dependence of the relaxation rate \(S(t) = (1/H) \frac{dM_{ZFC}}{dt}\) for the ZFC magnetization after the ZFC aging protocol, shows a peak at a characteristic time \(t_w\) near a wait time \(t_w\) (aging behavior), corresponding to a crossover from quasi equilibrium dynamics to non-equilibrium. The time \(t_w\), strongly depends on \(t_w\), temperature \((T)\), magnetic field \((H)\), and the temperature shift \((\Delta T)\). The rejuvenation effect is observed in both \(\chi'(T)\) and \(\chi''(T)\) under the \(T\)-shift and \(H\)-shift procedures. The memory of the specific spin configurations imprinted during the ZFC aging protocol can be recalled when the system is re-heated at a constant heating rate. The aging, rejuvenation, and memory effects observed in the present system are discussed in terms of the scaling concepts derived from numerical studies on 3D Edwards-Anderson spin glass model.

PACS. 75.50.Lk Spin glasses and other random magnets – 75.10.Nr Spin-glass and other random models – 75.40.Gb Dynamic properties

1 Introduction

In recent years, non-equilibrium dynamics, in particular, aging dynamics, of spin glass (SG) systems has been extensively studied theoretically \cite{1-10,11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28} and experimentally \cite{10,11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28}. When the SG system is quenched from a high temperature above the SG transition temperature \(T_g\) to a low temperature \(T\) below \(T_g\) (this process is called the zero-field cooled (ZFC) aging protocol), the initial state is not thermodynamically stable and relaxes to more stable state. The aging behaviors depend strongly on their thermal history within the SG phase. A typical experimental method for the study of the aging dynamics is the time dependence of the ZFC magnetization \((M_{ZFC})\) after the ZFC aging protocol. When a small magnetic field \((H)\) is applied after an isothermal aging at a temperature \(T\) below \(T_g\) for a wait time \(t_w\), \(M_{ZFC}\) increases as an observed time \(t\) increases. The rejuvenation (chaos) and memory effects are also significant features of the aging dynamics. These effects are typically measured from the low frequency AC magnetic susceptibility.

Such an aging dynamics of SG phase is explained mainly in terms of a real-space picture (the droplet model) \cite{11}. In this picture, the SG coherence length for equilibrium SG order, grows up slowly in aging processes. The scaling properties of age-dependent macroscopic susceptibility can be described by a growing coherence length \(L_T(t)\). The droplet model also predicts the following two. (i) The equilibrium SG states at two temperatures with the difference \(\Delta T\) are uncorrelated in length scales larger than the overlap length \(L_{\Delta T}\), i.e., so-called temperature \((T)\)-chaos nature of the SG phase. (ii) In the equilibrium and thermodynamic limits the SG phase is broken by a static magnetic field \(H\) of infinitesimal strength, thereby introduced is the crossover length \(L_H\). This length separates the mean size of SG domains \(L_T(t)\), such that they are dominated by the Zeemann energy for \(L_T(t) > L_H\) and by the SG free energy gap for \(L_T(t) < L_H\). Recently Monte Carlo (MC) simulations \cite{28} on the \(T\)- and \(H\)-shift aging processes have been carried out for the three-dimensional (3D) Ising Edwards-Anderson (EA) SG model with Gaussian nearest-neighbor interactions with zero mean and variance \(J\), where \(J\) is in the unit of energy. In the \(T\)-shift process, only a precursor of the temperature-chaos effect is observed, while the results on the \(H\)-shift process strongly support the droplet picture that the SG phase under a finite \(H\) is unstable in the equilibrium and thermodynamic limits.

In this paper we report our results on the aging dynamics (aging, rejuvenation and memory) of a 3D Ising SG, \textit{Cu}_{0.5}\textit{Co}_{0.5}\textit{Cl}_{2}\textit{FeCl}_{3} graphite bi-intercalation compound (GBIC). The aging behavior of the ZFC magnetization has been measured at various aging processes including the \(T\)- and \(H\)-shift perturbations. The rejuvenation effect of the low frequency AC magnetic susceptibility has been
measured at aging processes including the T- and H-shift perturbations. The memory of the specific spin configurations imprinted during the ZFC aging protocol can be recalled when the system is re-heated at a constant heating rate. The existence of the characteristic lengths $L_{AT}$ and $L_H$ is examined by the T- and H-shift aging protocols. The relaxation rate defined by $S(t) = (1/H) dM_{ZFC}/dt$ (see Sec. 2 [for more definitions]) exhibits a peak at a characteristic time $t_{cr}$, which is comparable to a wait time $t_w$. We show that the ratio $t_{cr}/t_w$ strongly depends on the aging process. These results are compared with those reported for typical spin glass systems such as Cu (15-13.5 at. % Mn) [12,13,14,15,22,23,25,27], Ag (11 at. % Mn) [16,21,28], CdCr$_{1-x}$In$_x$S$_4$ [17,18], Fe$_{0.5}$Mn$_{0.5}$TiO$_3$ [4,19,20], as well as results from MC simulations [2,17,18].

The equilibrium dynamics of Cu$_{0.5}$Co$_{0.5}$Cl$_2$-FeCl$_3$ GBIC has been reported in a previous paper [20]. The aging dynamics has been also studied: the $\omega$-scaling of $\chi'$ and $\chi''$ is also confirmed, where $\omega$ is the angular frequency of the AC magnetic field. This compound undergoes a SG transition at $T_g$ ($= 3.92 \pm 0.11$ K). The system shows a dynamic behavior that has some similarities and some significant differences compared to a 3D Ising SG. It shows critical slowing down with a value of the dynamic critical exponent that is rather similar to an Ising SG: $z = 6.6 \pm 1.2$, $\theta = 0.13 \pm 0.02$, and $\psi = 0.24 \pm 0.02$, where $z$, $\psi$, and $\theta$ are the dynamic critical exponent, the stiffness exponent (energy exponent) which determines the free energy of a droplet excitation, and the barrier exponent which describes how the barrier heights change with length scale. The critical relaxation time $\tau$ is well described by $\tau = \tau^* (1 - T/T_g)^{-z}$ with the dynamic critical exponent $x = 10.3 \pm 0.7$ and $\tau^* = (5.29 \pm 0.07) \times 10^{-6}$ sec. The in-field dynamics indicates, as for an Ising SG, that the SG transition is destroyed by a magnetic field. The equilibrium dynamics shows a frequency dependence that is different from an Ising SG, the absorption decreases with increasing frequency, whereas ordinary 3D Ising SG shows a increasing absorption with increasing frequency.

An aging behavior is observed that is rejuvenated by a large enough (magnetic field) perturbation.

2 Background

2.1 Scaling Properties

We present a simple review on the aging behavior of SG phase after the ZFC aging protocol, based on the droplet model [10]. This ZFC aging protocol process to the SG phase 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. Correspondingly, the size of domain defined by $R_T(t_a)$ grows with the age of $t_a$ and reaches $R_T(t_a)$ just before the field is turned on at $t = 0$ or $t_a = t_w$. The aging behavior in $M_{ZFC}$ is observed as a function of the observation time $t$. After $t = 0$, a probing length $L_T(t)$ corresponding to the maximum size of excitation grows with $t$, in a similar way as $R_T(t_a)$. When $L_T(t) \ll R_T(t_a)$, quasi-equilibrium dynamics is probed, but when $L_T(t) \gg R_T(t_a)$, non-equilibrium dynamics is probed. It is theoretically predicted that the mean SG domain-size $L_T(T)$ is described by a power law given by

$$L_T(t)/L_0 \approx (t/t_0)^{1/\psi},$$

where $L_0$ and $t_0$ are microscopic length and time scale, the exponent $1/\psi(T) = bT/T_g$ with $b = 0.16$ [2] linearly depends on $T$ except for the region near $T_g$, and $\Delta_g$ and $\psi$ are the characteristic scale of energy barrier of droplet excitations and the associated exponent, respectively. It is predicted by Komori et al. [2] that $\Delta\chi''(\omega, t)$ obeys the $\omega$-scaling law given by $\Delta\chi''(\omega, t) \approx (\omega)^{-\theta}$ with $\theta = (d - \theta(T))/z(T)$, $d = 3$ is the dimension, and $\theta = 0.20 \pm 0.03$. Experimentally, as shown in our previous paper [26], $\Delta\chi''(\omega, t)$ at 3.75 K obeys the $\omega$-scaling law, $\Delta\chi''(\omega, t) \approx (\omega)^{-\theta}$ with $\theta = 0.255 \pm 0.005$, where $\Delta\chi''(\omega, t) = \chi''(\omega, t) - (\chi''^0(\omega))$, where $(\chi''^0(\omega))$ is the stationary frequency-dependent absorption.

2.2 $S(t)$ and $\chi''(t_w; t + t_w)$

Here we present a simple review on the relation between $S(t)$ and $\chi''$. The absorption $\chi''$ is evaluated from the spin auto-correlation function $C(t_a - t; t_a) = \langle S_i(t_a - t)S_i(t_a) \rangle$ using the fluctuation-dissipation theorem (FDT) as

$$\chi''(\Delta t; t + \Delta t_w) \approx (-\pi/2T)\partial C(\Delta t_w; t + \Delta t_w)/\partial \ln t,$$

where $t_a = t + \Delta t_w$, $\Delta t_w = 2\pi/\omega$ (typically $\Delta t_w \leq 10^2$ sec) and $t$ is much larger than $\Delta t_w$. In the auto-correlation function, the over-line denotes the average over sites and over different realizations of bond disorder, and the bracket the average over thermal noises. For slow processes, the dispersion $\chi''(\Delta t_w; t + \Delta t_w)$ is approximated by

$$\chi''(\Delta t_w; t + \Delta t_w) \approx [1 - C(\Delta t_w; t + \Delta t_w)]/T.$$

In the quasi-equilibrium regime where the FDT holds, the ZFC susceptibility $\chi_{ZFC}(t_w; t + t_w)$ is described by

$$\chi_{ZFC}(t_w; t + t_w) \approx [1 - C(t_w; t + t_w)]/T,$$

where $t_a = t + t_w$ and $t_w$ is a wait time: typically, $t_w \approx 10^3 - 10^5$ sec. Then the relaxation rate $S(t)$ is described by

$$S(t) = d\chi_{ZFC}(t_w; t + t_w)/dt = (-1/T)\partial C(t_w; t + t_w)/\partial \ln t,$$

which corresponds to $(2/\pi)\chi''(t_w; t + t_w)$. It is predicted that $C(t_w; t + t_w)$ can be decomposed into a stationary part $C_{st}(t)$ and an aging part $C_{ag}(t_w; t + t_w)$. The latter is approximately described by a scaling function of $L_T(t)$ and $R_T(t_w)$. The corresponding aging part of $S(t)$ exhibits a peak at a characteristic time $t_{cr}$ ($\approx t_w$) [2,29], where $L_T(t) \approx R_T(t_w)$, showing a crossover between quasi-equilibrium region and non-equilibrium region.
3 Experimental procedure

The DC magnetization and AC susceptibility of Cu$_{0.5}$Co$_{0.5}$Cl$_2$-FeCl$_3$ GBIC were measured using a SQUID magnetometer (Quantum Design, MPMS XL-5) with an ultra low 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 $h = 0.1$ Oe and frequency $f = \omega/2\pi = 0.05$ Hz. Each experimental procedure for measurements is presented in the text and figure captions. The detail of sample characterizations and sample preparation is given in the previous paper [26].

In our measurement of the time ($t$) dependence of the zero-field cooled (ZFC) magnetization ($M_{ZFC}$), the time required for the ZFC aging protocol and subsequent wait time was precisely controlled. Typically it takes $240 \pm 3$ sec to cool the system from 10 K to 3.75 K. It takes another $t_{scanning} = 230 \pm 3$ sec until $T (= 3.75$ K) becomes stable within the uncertainty ($\pm 0.01$ K). The system is kept at $T = 3.75$ K and $H = 0$ for a wait time $t_w (0 \leq t \leq 3 \times 10^4$ sec). The time for setting up a magnetic field from $H = 0$ to $H = 5$ Oe is $68 \pm 2$ sec. In the ZFC measurement, the sample is slowly moved through the pick-up coils over the scan length (4 cm). The magnetic moment of the sample induces a magnetic flux change in the pick-up coils. It takes 12 sec for each scan. The data at $t$ is 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 $\Delta t$ 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 three scans and $t_p = 45$ sec or 30 sec for $t_w (\geq 2.0 \times 10^4$ sec), and (ii) the time window $\Delta t = 12$ sec for one scan and $t_p = 1$ or 2 sec for either $t_w (\leq 10^3$ sec) or $H \geq 100$ Oe.

4 Result

4.1 $S(t)$ : $t_w$ and $T$ dependence

The $t$ dependence of $M_{ZFC}$ was measured under various conditions. Figure 1(a) shows the $T$ dependence of the ZFC and FC magnetization ($M_{ZFC}$ and $M_{FC}$) at $H = 5$ Oe, where $H$ is applied along a direction perpendicular to the c axis. The system was quenched from 50 to 1.9 K in the absence of $H$ before the measurement. The change of $T$ with the time $t$ during the measurement is shown in Fig. 1(b). The ZFC magnetization was measured with increasing $T$ from 1.9 to 3.75 K in the presence of $H (= 5$ Oe). The system was kept at $T = 3.75$ K for $2.27 \times 10^4$ sec. Subsequently the ZFC measurement was continued from 3.75 to 8 K. The system was annealed at 50 K for $1.2 \times 10^3$ sec. The FC magnetization was measured from 8 to 1.9 K. There is a remarkable increase of $M_{ZFC}$ during the one stop at $T = 3.75$ K. When the warming up of the system was restarted, the increase in $M_{ZFC}$ with $T$ is much weaker than that in $M_{ZFC}^{ref}$, as a reference without

Fig. 1. (a) $T$ dependence of $M_{ZFC}$ and $M_{FC}$ at $H = 5$ Oe for Cu$_{0.5}$Co$_{0.5}$Cl$_2$-FeCl$_3$ GBIC. The measurement was carried out after the ZFC aging protocol: annealing of the system at 50 K for $1.2 \times 10^3$ sec at $H = 0$ and quenching from 50 to 1.9 K. During the ZFC measurement the system was aging at 3.75 K for $2.27 \times 10^4$ sec. (b) The change of $T$ with $t$ during the measurement of $M_{ZFC}$ and $M_{FC}$. The discontinuity of $T$ with $t$ at $T = 4.4$ K is due to the system error occurring in the process of temperature stabilization. (c) The relaxation rate $S(t) = (1/H) dM_{ZFC}(t)/dt$ at 3.75 K. The time taken during the measurement of $M_{ZFC}$ from 1.9 to 3.75 K at $H = 5$ Oe was $t_0 = 5.4 \times 10^3$ sec. $t = 0$ is a time just after $T$ becomes 3.75 K.
the origin of $t$, the stop: $M_{ZFC}$ merges with $M_{ZFC}^\text{eff}$ well above $T_g$. Similar result of $M_{ZFC}$ vs $T$ was reported for a 3D Ising SG $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$. Bernardi et al. [5] have concluded that $M_{ZFC}$ is described by a scaling function of $R_T(t_w)$ and $L_T(t)$ [$M_{ZFC}(T, t) = G(L_T(t), R_T(t_w))]$, where $G$ is the scaling function. The $t$ dependence of $M_{ZFC}$ was monitored during the stop at $T = 3.75$ K, where the time taken during the measurement from 1.9 to 3.75 K was $5.4 \times 10^3$ sec, corresponding to a wait time $t_w$. Figure 2(c) shows the $t$ dependence of the relaxation rate $S(t)$, where the origin of $t$ ($t = 0$) is a time when $T$ reaches 3.75 K.

We have measured the $t$ dependence of $\chi_{ZFC} (= M_{ZFC}/H)$ at various fixed $T$, where $t_w = 2.0 \times 10^3$ sec and $H = 1$ Oe. The measurement was carried out after the ZFC aging protocol: annealing of the system at $T = 50$ K and $H = 0$ for $1.2 \times 10^3$ sec, quenching from 50 K to $T$, and isothermal aging at $T$ for $t_w$. The origin of $t$ ($t = 0$) is a time just after $H = 1$ Oe is applied at $T$.

**Fig. 2.** (a) and (b) $t$ dependence of $S$ for $3.0 \leq T \leq 4.7$ K. $H = 1$ Oe. The measurement of $\chi_{ZFC}$ vs $t$ was carried out after the ZFC aging protocol (annealing of the system at 50 K and $H = 0$ for $1.2 \times 10^3$ sec, quenching from 50 K to $T$, and isothermal aging for $t_w = 2.0 \times 10^3$ sec). $t = 0$ is the time just after $H = 1$ Oe is applied at $T$.

**Fig. 3.** $T$ dependence of $t_{cr}$ at which $S(t)$ has a peak value $S_{max}$. $t_w = 2.0 \times 10^3$ sec. $H = 1$ Oe. (b) $T$ dependence of the peak height $S_{max}$ (see Fig. 2).
Fig. 4. (a) $t$ dependence of $S$ for $0 \leq t_w \leq 1.5 \times 10^4$ sec. $T = 3.75$ K, $H = 5$ Oe. The ZFC aging protocol: annealing of the system at 50 K for $1.2 \times 10^3$ sec at $H = 0$, quenching from 50 to 3.75 K, and then isothermal aging at 3.75 K and $H = 0$ for a wait time $t_w$. The measurement was started at $t = 0$ when the field $H$ is turned on. (b) $t_{cr}$ vs $t_w$. The straight line denotes a relation given by $t_{cr} = (0.68 \pm 0.03)t_w$. (c) Scaling plot of $S/S_{\text{max}}$ vs $t/t_w$ for the limited $t_w$ and $t$ ($5.0 \times 10^3 \leq t_w \leq 1.5 \times 10^5$ sec, $0 \leq t \leq 6.0 \times 10^3$ sec).

Figure 4(a) shows the $t$ dependence of $S$ at various $t_w$, where $T = 3.75$ K and $H = 5$ Oe. The relaxation rate $S(t)$ shows a peak at $t = t_{cr}$, shifting to the long-$t$ side with increasing $t_w$. Similar behavior of $S(t)$ vs $t$ at various $t_w$ has been observed by Jönsson et al. for Ag (11 at.% Mn) [21], where $t_{cr} \approx t_w$. In Fig. 4(b) we show the characteristic time $t_{cr}$ as a function of $t_w$ for $0 \leq t_w \leq 3.0 \times 10^4$ sec, where a straight line denotes the relation described by $t_{cr} = (0.68 \pm 0.03)t_w$. Figure 4(c) shows the plot of $S(t)/S_{\text{max}}$ as a function of $t/t_w$, where only the data of $S$ vs $t$ with long $t_w$ ($5 \times 10^3 \leq t_w \leq 1.5 \times 10^4$ sec) for $0 < t < 6 \times 10^4$ sec are used. It seems that $S(t)/S_{\text{max}}$ is well described by a scaling function of $t/t_w$ [$S(t)/S_{\text{max}} = F(t/t_w)$] in the region of long $t_w$, although the data at $t_w = 1.5 \times 10^4$ sec slightly deviates from the other data. The scaling function $F(x)$ has a very broad peak centered at $x \approx 0.68$. This result suggests that the spin auto-correlation function $C(t_{w0}, t + t_w)$ is described by a scaling function of only $t/t_w$, since it is related to $S(t)$ by Eq. (6).

4.2 $S(t)$ for $0 \leq t_w \leq 750$ sec

We have measured the $t$ dependence of $\chi_{\text{ZFC}}$ at $T = 3.75$ K and $H = 5$ Oe as a wait time $t_w$ ($0 \leq t_w \leq 750$ sec) is varied as a parameter. The measurement was carried out after the ZFC aging protocol: annealing of the system at 50 K for $1.2 \times 10^3$ sec at $H = 0$, quenching from 50 to 3.75 K, and then isothermal aging at $T = 3.75$ K and $H = 0$ for a wait time $t_w$. The origin of $t$ ($t = 0$) is a time just after the field $H$ is turned on. As is described in Sec. 3 it takes $t_{w0} = 230 \pm 3$ sec until the temperature becomes stable at 3.75 K within the experimental uncertainty of $\pm 0.01$ K, after quenching the system from 50 to 3.75 K at $H = 0$. This time $t_{w0}$ is not included in the wait time $t_w$. If $t_{w0}$ is included in $t_w$, however, the effective wait time may be longer than $t_{w0}^{\text{eff}} (= t_{w0} + t_{w0})$. If this is the case, the measurement with $t_w = 0$ is not possible in a strict sense. In spite of such a situation, here we assume that $t_{w0}$ is not included in $t_w$. Figures 4(a) and (b) show the $t$ dependence of $S$ for $0 \leq t_w \leq 750$ sec, where $T = 3.75$ K and $H = 5$ Oe. For $t_w = 0$, $S(t)$ shows a shoulder around $t = 10^3$ sec and a broad peak at $t_{cr} = 6.45 \times 10^3$ sec. For $t_w = 100$ sec, a small peak of $S(t)$ is observed at $t_{cr} = 1.06 \times 10^3$ sec in addition to a possible broad peak around $t_{cr} = 1.0 \times 10^3$ sec. This small peak shifts to the short $t$-side with increasing $t_w$: $t_{cr} = 740$ sec for $t_w = 200$ sec. For $t_w = 750$ sec, a broad peak is observed at $t_{cr} = 2.8 \times 10^3$ sec. The values of $t_{cr}$ thus obtained are also plotted as a function of $t_w$ in Fig. 4(b). It should be noted that for very short $t_w$ ($0 \leq t \leq 200$ sec), there are at least two kinds of domains: domains with large size corresponding to long $t_{cr}$ ($\approx 6.5 \times 10^3$ sec) coexist with domains with small size corresponding to short $t_{cr}$ ($\approx 750$ sec) in the regular aging regime. Figure 5 shows the $t$ dependence of $S(t)$ at various $T$ ($3.4 \leq T \leq 4.5$ K) and $H = 1$ Oe for $t_w = 0$. At $T = 3.2$ K, $S(t)$ shows a shoulder around $t = 400$ sec and a very broad peak at $t_{cr} = 7.1 \times 10^3$ sec. This shoulder shifts to the long $t$-side with increasing
T and changes in to a broad peak. At $T = 3.6$ K, $S(t)$ shows a peak at $t_{cr} = 710$ sec. It tends to increase with further increasing $t$, suggesting that $t_{cr}$ is longer than $10^4$ sec. At $T = 3.8$ K, $S(t)$ shows a very broad peak centered at $t_{cr} = 3.8 \times 10^3$ sec.

Similar behavior has been reported by Rodriguez et al.\cite{25} in the time decay of the thermal remnant magnetization (TRM) of Cu$_{0.94}$Mn$_{0.06}$ ($T_g = 31.5$ K) with various wait time ($t_w = 0 - 10^4$ sec) and a series of rapid FC cooling protocol from 35 to 26 K at $H (=20$ Oe). The relaxation rate $S(t) = -(1/H)\text{d}M_{TRM}(t)/\text{d}ln t$ for $M_{TRM}(t)$ is equivalent to $S(t) = -(1/H)\text{d}M_{ZFC}(t)/\text{d}ln t$ for $M_{ZFC}(t)$.

They have shown that $S(t)$ at $t_w = 0$ exhibits a broad peak at an effective time $t^{eff}_{cr}$ ($= 19 - 406$ sec), which is strongly dependent on the FC cooling protocol. For the larger $t^{eff}_{cr}$, there is a significant contamination in $S(t)$ at $t_w \neq 0$ from the FC cooling protocol. Recently the long-time decay of $M_{TRM}(t)$ has been also examined by Kenning et al.\cite{27} for the same system with rapid FC cooling protocol and short wait time ($t_w = 7 - 110$ sec). A $t_w$-independent long-time decay overlaps with the $t_{cr}$-dependent short-time decay. For the short-time decay, the corresponding $S(t)$ exhibits a peak at $t_{cr}$ ($\approx t_w$). The long-time decay may be related to the initial state distribution developed during the FC cooling protocol.

4.3 $S(t)$ under the $H$-shift

We have measured the $t$ dependence of $\chi_{ZFC}$ at $T = 3.75$ K for various $H$, where $t_w = 1.0 \times 10^4$ and $3.0 \times 10^4$ sec. The measurements were carried out after the ZFC aging protocol: annealing of the system at $T = 50$ K and $H = 0$ for $1.2 \times 10^3$ sec, quenching from 50 to 3.75 K at $H = 0$, and isothermal aging at 3.75 K for $t_w$. The origin of $t$ ($t = 0$) is the time just after $H$ is turned on. The window time used in this measurement was 12 sec. Figures 7(a) and (b) show the $t$ dependence of $S$ for $t_w = 1.0 \times 10^4$ sec and $3.0 \times 10^4$ sec as $H$ is varied as a parameter, where $T = 3.75$ K. The relaxation rate $S(t)$ exhibits a peak at $t = t_{cr}$, corresponding to a characteristic time scale of crossover from the isothermal aging state under $H = 0$ to that under a finite $H$. Figure 8(a) shows the plot of $t_{cr}$ at $T = 3.75$ K as a function of $H$ for $t_w = 1.0 \times 10^4$ and $3.0 \times 10^4$ sec. The value of $t_{cr}$ for $t_w = 1.0 \times 10^4$ and $3.0 \times 10^4$ sec drastically decreases with increasing $H$. The value of $t_{cr}$ for $t_w = 3 \times 10^4$ sec is much larger than that for $t_w = 1 \times 10^4$ sec at low $H$ ($H < 50$ Oe). However, they are almost identical irrespective of $t_w$ for high $H$ ($H > 100$ Oe). We note that $\ln t_{cr}$ is linearly dependent on $H$ only for $H < H_0$; $H_0 = 50$ Oe for $t_w = 3 \times 10^4$ sec and 100 Oe for $t_w = 1.0 \times 10^4$ sec. In Fig. 8(b) we show the $H$ dependence of $S_{max}$. The peak value $S_{max}$ linearly decreases with increasing $H$ irrespective of $t_w$ and tends
to reduce to zero above 300 Oe. Similar behavior of \( t_{cr} \) vs \( H \) has been reported by Zotev et al. \[22\] in Cu (1.5 at.% Mn). The break of the linear dependence of \( \ln t_{cr} \) on \( H \) occurs at \( H_a \). The value of \( H_a \) decreases with increasing \( t_w \). Above \( H_a \), the data of \( \delta M = (M_{TRM} - M_{FC} - M_{ZFC}) \) vs \( H \) greatly deviates from zero, where \( M_{TRM} \) is the thermal remnant magnetization. This result suggests that the crossover from the quasi equilibrium to the nonequilibrium regime occurs at \( H_a \), leading to the violation of FDT.

According to Takayama \[8\], the \( H \) dependence of \( t_{cr} \) under the \( H \)-shift aging process is governed by three lengths, \( L_T(t_{cr}, H), R_T(t_{w}), \) and the crossover length \( L_H \) \[1\], given by
\[
L_H/L_0 \approx (H/T_H)^{-1/\delta},
\]
where \( \delta = (d/2 - \theta) \) and \( T_H \) is the magnetic field corresponding to a wall stiffness \( T \) (a typical energy setting the scale of free energy barriers between conformations). The scaling relation is predicted to exist between the normalized lengths \( y = L_T(t_{cr}, H = 0)/L_H \) and \( x = R_T(t_{w})/L_H \):
\[
y = x - c_H x^{1+\delta} \quad \text{with} \quad c_H = 0.15 \quad \text{for} \quad x < 1.
\]
For \( x \ll 1 \) corresponding to the case of low \( H \) and short \( t_w \), \( y = x \), implying that \( t_{cr} = t_w \). For \( x > 0.2 \) corresponding to large \( H \) and long \( t_w \), the curve \( y(x) \) deviates below the line \( y = x \), indicating that \( t_{cr} \) is shorter than \( t_w \). From this scaling relation, \( t_{cr} \) can be obtained as
\[
t_{cr} = t_w [1 - c_H (H/T_H)(t_w/t_0)^{\delta/z(T)}]^{1/z(T)}.
\]

The following features of \( t_{cr} \) vs \( H \) are derived from Eq. (5).

(i) The time \( t_{cr} \) is not simply proportional to \( t_w \). (ii) In the limit of \( H \approx 0 \), \( \ln t_{cr} \) is linearly dependent on \( H \):
\[
\ln(t_{cr}/t_w) = -\alpha_H H,
\]
with
\[
\alpha_H = z(T)(c_H/T_H)(t_w/t_0)^{\delta/z(T)}.
\]
The slope \( \alpha_H \) increases with increasing \( t_w \). (iii) The \( T \) dependence of \( t_{cr} \) comes from the exponent \( z(T) \). The slope

Fig. 7. \( t \) dependence of \( S \) for \( 5 \leq H \leq 300 \) Oe. \( T = 3.75 \) K. (a) \( t_w = 1.0 \times 10^4 \) sec. (b) \( t_w = 3.0 \times 10^5 \) sec. The time \( t = 0 \) is a time when \( H \) is turned on. The ZFC aging protocol before the measurement is similar to that used in Fig. 2.

Fig. 8. \( H \) dependence of (a) \( t_{cr} \) and (b) \( S_{max} \) for \( t_w = 1.0 \times 10^4 \) and \( 3.0 \times 10^4 \) sec, which is obtained from Fig. 7.
\( \alpha_H \) increases with increasing \( T \) below \( T_g \) mainly because of the factor \((t_w/t_0)^{-\alpha(\gamma(T))}\).

We find that our results of Fig. 9(a) is consistent with the above predictions. The slope \((\alpha_H = 0.0505 \pm 0.0003 /\text{Oe}) \) for \( t_w = 3.0 \times 10^4 \) sec is larger than that \((\alpha_H = 0.0262 \pm 0.0005 /\text{Oe}) \) for \( t_w = 1.0 \times 10^4 \) sec. If we assume that \( \delta = 1.37, 1/z(T) = bT/T_g, T_g = 3.92 \) K, \( b = 0.16 \), and \( t_0 = \tau^* = 5.29 \times 10^{-6} \) sec, \( T_H \) can be estimated as \( T_H = 2.15 \) kOe for \( t_w = 3.0 \times 10^4 \) sec and \( T_H = 3.30 \) kOe for \( t_w = 1.0 \times 10^4 \) sec. Our numerical calculation of \( \alpha_H \) for \( 3 \) K \( \leq T \leq T_g \) predicts that the slope \( \alpha_H \) increases with increasing \( T \) below \( T_g \): \( \alpha_H = 0.036 \) at \( T = 3.4 \) K and \( \alpha_H = 0.044 \) at \( T = 3.6 \) K for \( t_w = 3.0 \times 10^4 \) sec. Although the measurement on \( \alpha_H \) vs \( T \) has not been carried out in the present system, this prediction is consistent with the result reported by Zotev et al. \[22\] in Cu (1.5 at.\% Mn) as the increase of \( \alpha_H \) vs \( T \) with increasing \( T \) for \( 0.7 < T/T_g < 0.85 \).

**4.4 \( S(t) \) under the \( T \)-shift**

We have measured the \( t \) dependence of \( \chi_{ZFC} \) under the \( T \)-shift from the initial temperature \( T_i \) to the final temperature \( T_f \) \((= 3.75 \) K \( = 0.957T_g) \), where \( T_i = 3, 3.2, 3.4, 3.5, 3.6, \) and \( 3.9 \) K. The measurement was carried out after the ZFC aging protocol: quenching of the system from 50 K to \( T_i \), and isothermal aging at \( T = T_i \) and \( H = 0 \) for \( t_w \) \((= 3.0 \times 10^4 \) and \( 3.0 \times 10^3 \) sec). The origin of time \( (t = 0) \) is a time just after \( T \) was shifted from \( T_i \) to \( T_f = 3.75 \) K and subsequently \( H (= 5 \) Oe) was turned on. Figures 9(a) and (b) show the \( t \) dependence of \( S \) at \( T_f = 3.75 \) K at various initial temperature \( T_i \). The temperature difference is defined as \( \Delta T = T_f - T_i \): the positive \( T \)-shift for \( \Delta T > 0 \) and negative \( T \)-shift for \( \Delta T < 0 \). The relaxation rate \( S(t) \) exhibits a peak at \( t = t_{cr} \) irrespective of the sign of \( \Delta T \). The width of the peak in \( S(t) \) for the negative \( T \)-shift is much broader than that for the positive \( T \)-shift. In Fig. 9(c) we show \( t_{cr} \) as a function of \( \Delta T \) for the positive \( T \)-shift for \( t_w = 3.0 \times 10^4 \) and \( 3.0 \times 10^3 \) sec. The decrease of \( t_{cr} \) with increasing \( \Delta T \) is observed at low \( \Delta T \) for both \( t_w = 3.0 \times 10^4 \) and \( 3.0 \times 10^3 \) sec. The value of \( t_{cr} \) becomes independent of \( \Delta T \) at large \( \Delta T \). For the negative \( T \)-shift, the on the other hand, the value of \( t_{cr} \) for \( \Delta T = -0.15 \) K \( (T_i = 3.9 \) K) is larger than that for \( \Delta T = 0 \) K \( (T_i = 3.75 \) K) for both \( t_w = 3.0 \times 10^4 \) and \( 3.0 \times 10^3 \) sec. Similar behaviors of \( t_{cr} \) vs \( \Delta T \) for the positive \( T \)-shift have been reported by Granberg et al. [Cu (10 at. \% Mn)] \[13\], Djurberg et al. [Cu (13.5 at. \% Mn)] \[14\], and Jonsson et al. [Ag (11 at. \% Mn)] \[10\].

The temperature chaos scenario postulates that the SG equilibrium configurations at different temperatures at \( T_f \) and \( T_i \) are strongly correlated only up to the overlap-length \( L_{\Delta T} \), beyond which these correlations decay to zero. From the droplet theory, the overlap length \( L_{\Delta T} \) is described by \[12\]

\[
L_{\Delta T}/L_0 \approx (T^{1/2}/A)/T_T^{3/2} \sim 1/\zeta, \tag{9}
\]

where \( \zeta \) is the chaos exponent \((\zeta = d_s/2 - \theta)\), \( d_s \) is the fractal dimension of the surface of the droplet, and \( T_T \)
is the temperature corresponding to the wall stiffness $\gamma$. From this scenario a scaling relation is predicted to exist between the normalized lengths $y = (L_{T_f}(t_c)/L_{\Delta T})$ and $x = (|R_i(t_w)/L_{\Delta T}|$; $y = x - c_Tx^{1+z} \zeta$ with $c_T = 0.25$ and $\zeta = 0.385$ for $x < 0.15$ \cite{21,28}. The cumulative aging corresponds to the relation $y = x$ which is valid in the limit $x \approx 0$ (the small $|\Delta T|$ and short $t_w$). The large $|\Delta T|$ and long $t_w$ corresponds to large $x$. For $x > 0.05$, the curve $y$ deviates from the straight line $y = x$, corresponding to a rejuvenation due to the temperature chaos effect \cite{21,28}.

The value of $t_{cr}/t_0$ can be described by

$$t_{cr}/t_0 = (t_w/t_0)^{T_f/T_0} [1 - c_Tp(T_f)|\Delta T|/(t_w/t_0)^{z(T_f)}],$$

(10)

with $p(T_f) = T_f^{1/2}/T_0^{3/2}$. In the limit of $|\Delta T| \approx 0$, $t_{cr}$ is linear to $T$:

$$\ln(t_{cr}/t_0) = -\alpha_T|\Delta T|,$$

with

$$\alpha_T = z(T_f)c_Tp(T_f)(t_w/t_0)^{z(T_f)}.$$

In fact, the prediction that $\alpha_T$ increases with increasing $t_w$ is experimentally confirmed from Fig. \ref{fig:10}(c). The curve ($\ln(t_{cr}$ vs $|$\Delta T$|)$ is linearly dependent on $|$\Delta T$| > 0$ for $|$\Delta T$| < $\Delta T_0$ with the slope $\alpha_T$: $\alpha_T = 9.9 \pm 0.4$ and $\Delta T_0 = 0.3$ K for $t_w = 3.0 \times 10^4$ sec and $\alpha_T = 2.6$ and $\Delta T_0 \approx 0.15$ K for $t_w = 3.0 \times 10^3$ sec. If we assume that $\zeta = 0.385$, $c_T = 0.25$, $1/z(T_f) = 6T_f/T_0$, $b = 0.16$, and $t_0 = \tau^* = 5.29 \times 10^{-6}$ sec \cite{20} for $t_w = 3.0 \times 10^4$ sec, we have $p(T_f) = 1.62 \pm 0.07$ or $T_f = 1.12 \pm 0.03$ K for $t_w = 3.0 \times 10^4$ sec.

Numerical calculations of Eq. \ref{eq:10} are carried out as a function of $|$\Delta T$| for $t_w = t_w$(cal) = 2.04 \times 10^4$ sec, $T_f = T_0 - |\Delta T|$, $\zeta = 0.385$, $t_0 = \tau^* = 5.29 \times 10^{-6}$ sec, and $c_T = 0.25$. Note that we use $t_w$(cal) as $t_w$ instead of $t_w$(exp) ($= 3.0 \times 10^4$ sec) $t_w$(cal) = 0.68$t_w$(exp), which leads to the better agreement between our data and calculations at low $|$\Delta T$. In Figs. \ref{fig:10}(a) and (b) we show the results of $t_{cr}$ vs $|$\Delta T$|$ for the positive and negative $T$-shifts, respectively, where $p(T_f)$ is varied as a parameter from 0 to 2.0. For comparison, our data of $t_{cr}$ vs $|$\Delta T$|$ with $t_w = 3.0 \times 10^4$ sec for the positive $T$-shift are also plotted in Fig. \ref{fig:10}(a). For the positive $T$-shift, $t_{cr}$ decreases with increasing $|\Delta T|$, independent of $p(T_f)$ for $0 \leq p(T_f) \leq 2$. For the negative $T$-shift, $t_{cr}$ increases with increasing $|\Delta T|$ for $0 \leq p(T_f) < 0.7$. It increases with increasing $|\Delta T|$, showing a peak, and decreases with further increasing $|\Delta T|$ for $0.7 < p(T_f) < 0.92$ (see the inset of Fig. \ref{fig:10}(b)). On the other hand, it decreases with increasing $|\Delta T|$ for $0.92 < p(T_f) < 2$. This indicates that $t_{cr}$ decreases with increasing $|\Delta T|$ for both the positive and negative $T$-shifts for $0.92 < p(T_f) < 2$ (the symmetric $T$-chaos). We find that our data for both the positive and negative $T$-shifts agree well with the curve with $p(T_f) \approx 0.8$ in Fig. \ref{fig:10}(a), corresponding to $T_f = 1.80$ K. This value of $T_f$ is on the same order as that derived from the slope $c_T$.

For Ag (11 at. % Mn) \cite{21}, $t_{cr}$ decreases with increasing $|\Delta T|$ for the positive $T$-shift. For the negative $T$-shift, however, $t_{cr}$ shifts to the long-$t$ side at $|\Delta T| = 0.1$ K and then $t_{cr}$ decreases with further increasing $|\Delta T|$. Similar behavior has also been observed in Cu (13.5 at. % Mn) \cite{14}.

![Fig. 10.](image-url) Numerical calculations of $t_{cr}$ vs $|\Delta T|$ given by Eq. \ref{eq:10} for $T_f = 3.75$ K, $T_f = T_0 + |\Delta T|$, and $t_w = t_w$(cal) = 2.04 \times 10^4$ sec, where $T_0 = 3.92$ K, $b = 0.16$, $\zeta = 0.385$, $c_T = 0.25$, $t_0 = \tau^* = 5.29 \times 10^{-6}$ sec, and $0 \leq p(T_f) \leq 1$. (a) The positive $T$-shift ($|\Delta T| > 0$). For comparison, our data of $t_{cr}$ vs $|\Delta T|$ for $t_w = 3.0 \times 10^4$ sec are denoted by solid triangles. The value of $t_w$ (= $t_w$(cal) = 2.04 \times 10^4$ sec) used in the calculation is different from that used in the experiment ($t_w$(exp) = 3.0 \times 10^4 sec), where $t_w$(cal) = 0.68$t_w$(exp) (see Fig. \ref{fig:11}(b)). (b) The negative $T$-shift ($|\Delta T| < 0$). Although the maximum of $|\Delta T|$ is $T_g - T_f = 0.17$ K in the present system, the value of $t_{cr}$ for the negative $T$-shift is independent of $T_g$ and is applicable to the other systems with different $T_g$.

4.5 Memory effect in $\chi'$ and $\chi''$

The memory effect in the SG system is defined as follows. When the system is cooled down to a low temperature
Fig. 11. $T$ dependence of (a) $\Delta \chi'$ and (b) $\Delta \chi''$: single memory experiments (I and II). $f = 0.05$ Hz, $h = 0.1$ Oe. The measurement (I) (denoted as gradual) was carried out after the ZFC aging protocol (I): gradual decrease of $T$ from 20 to 3.75 K, isothermal aging at $T_1 = 3.75$ K for 6.3 hours, and further gradual decrease from 3.75 to 1 K. The measurement (II) (denoted as quenched) was carried out after the ZFC aging protocol (II): quenching of the system from 20 to 3.75 K at $H = 0$, isothermal aging at 3.75 K for 10 hours, and further quenching from 3.75 to 1 K. Both $\chi'$ and $\chi''$ were simultaneously measured with increasing $T$ (aging ZFC curves). $\Delta \chi'$ and $\Delta \chi''$ are the deviations of the aging ZFC curve from the reference ZFC curve measured with increasing $T$ after the standard ZFC aging protocol: quenching from 20 to 1 K at $H = 0$. Above 4.3 K, $\Delta \chi'$ is independent of the detail of the ZFC aging protocol. (ii) The absorption $\Delta \chi''$ exhibits a negative local minimum at 3.75 K for both cases of the rapid and gradual cooling. Above 4.1 K, $\Delta \chi''$ is independent of the detail of the ZFC aging protocol. In summary, the equilibration at 3.75 K gives rise to a dip in $\Delta \chi'$ and $\Delta \chi''$, suggesting that the memory of spin configurations which are imprinted during the cooling process, is recalled during the heating. The dip of $\Delta \chi'$ and $\Delta \chi''$ at 3.75 K for the rapid cooling is much narrower than that for the gradual cooling. The reason is that in the case of the gradual cooling the spin configurations at $T$ not equal to 3.75 K are also imprinted during the cooling process. Similar but more pronounced single memory effect has been reported by Jonsson et al. for Ag (11 at. % Mn) [10].

Figures 12(a) and (b) show the $T$ dependence of $\Delta \chi'$ and $\Delta \chi''$ for the double memory experiment, where $T_1 = 3.75$ K, $T_2 = 3.0$ K, $f = 0.05$ Hz, and $h = 0.1$ Oe. The measurement was carried out after the ZFC aging protocol (only in the case of rapid cooling): quenching of the system from 20 to 3.75 K at $H = 0$ at the rate 0.13 K/sec, isothermal aging at $T_1 = 3.75$ K for $t_w1 = 3.6 \times 10^4$ sec, quenching from 3.75 to 3 K at the rate 0.025 K/sec, isothermal aging at $T_2 = 3.0$ K for $t_w2 = 3.6 \times 10^4$ sec, and quenching from 3.0 to 1 K at the rate 0.04 K/sec. Both $\chi''_{mem}$ and $\chi'''_{mem}$ were simultaneously measured with increasing $T$ (aging ZFC curves). $\Delta \chi'$ and $\Delta \chi''$ are the deviations of the aging ZFC curve from the reference ZFC curve.

below $T_g$, a memory of the spin configurations which is imprinted in the specific cooling sequence, can be recalled when the system is re-heated at a constant heating rate [16]. In a single memory experiment, the memory is imprinted at $T_1$ for $t_w1$ during the ZFC aging protocol. In a double memory experiment, the memory is imprinted at $T_1$ for $t_w1$, and at $T_2$ ($< T_1$) for $t_w2$ during the ZFC aging protocol. The dispersion and absorption thus recalled with increasing $T$ are defined as $\chi'_{mem}(\omega, T)$ and $\chi''_{mem}(\omega, T)$, respectively. For comparison, the dispersion and absorption as references [$\chi'_{ref}(\omega, T)$ and $\chi''_{ref}(\omega, T)$], are also obtained with increasing $T$ after the system is quenched from a high temperature above $T_g$ to the lowest temperature. Such AC susceptibility data are called as the ZFC reference susceptibilities, where no memory is imprinted. For clarity we define the difference between the aging ZFC and reference ZFC susceptibilities as $\Delta \chi'(\omega, T) = \chi'_{mem}(\omega, T) - \chi'_{ref}(\omega, T)$ and as $\Delta \chi''(\omega, T) = \chi''_{mem}(\omega, T) - \chi''_{ref}(\omega, T).$ Figures 11(a) and (b) show the $T$ dependence of $\Delta \chi'$ and $\Delta \chi''$ for the single memory experiment, where $T_1 = 3.75$ K, $f = 0.05$ Hz, and $h = 0.1$ Oe. Two kinds of measurements were carried out, depending on cooling rate during the ZFC aging protocol: (i) the gradual cooling (gradual decrease of $T$ from 20 to 3.75 K at the cooling rate $5.6 \times 10^{-4}$ K/sec, isothermal aging at $T_1 = 3.75$ K for $t_w1 = 2.27 \times 10^4$ sec, and further gradual decrease from 3.75 to 1 K at the rate $2.6 \times 10^{-4}$ K/sec), and (ii) the rapid cooling (quenching of the system from 20 to 3.75 K at $H = 0$ at the rate 0.13 K/sec, isothermal aging at $T_1 = 3.75$ K for $t_w1 = 3.6 \times 10^4$ sec, and further quenching from 3.75 to 1 K at the rate 0.06 K/sec). Both $\chi'_{mem}$ and $\chi''_{mem}$ were simultaneously measured with increasing $T$ (aging ZFC curves) from 1.9 to 8 K at the rate $1.7 \times 10^{-4}$ K/sec. The reference ZFC curves ($\chi'_{ref}$ and $\chi''_{ref}$) were measured with increasing $T$ from 1.9 to 8 K at the rate $1.7 \times 10^{-4}$ K/sec after the standard ZFC aging protocol: quenching from 20 to 1 K at $H = 0$ at the rate 0.15 K/sec. The results are as follows. (i) For the case of gradual cooling, $\Delta \chi'$ exhibits negative local minima at 3.4 and 3.75 K, while for the case of rapid cooling, $\Delta \chi'$ exhibits a negative local minimum at 3.75 K. Above 4.3 K, $\Delta \chi'$ is independent of the detail of the ZFC aging protocol. (ii) The absorption $\Delta \chi''$ exhibits a negative local minimum at 3.75 K for both cases of the rapid and gradual cooling. Above 4.1 K, $\Delta \chi''$ is independent of the detail of the ZFC aging protocol. In summary, the equilibration at 3.75 K gives rise to a dip in $\Delta \chi'$ and $\Delta \chi''$, suggesting that the memory of spin configurations which are imprinted during the cooling process, is recalled during the heating. The dip of $\Delta \chi'$ and $\Delta \chi''$ at 3.75 K for the rapid cooling is much narrower than that for the gradual cooling. The reason is that in the case of the gradual cooling the spin configurations at $T$ not equal to 3.75 K are also imprinted during the cooling process. Similar but more pronounced single memory effect has been reported by Jonsson et al. for Ag (11 at. % Mn) [10].
for the single memory effect. It seems that the spin configurations imprinted at \( T = T_1 \) is partially reinitialized by the spin configurations imprinted at \( T_2 = 3.0 \) K during the cooling process may be partially reinitialized by the spin configurations imprinted at \( T_2 = 3.0 \) K for the double memory effect has been reported by M. Suzuki, I. S. Suzuki: Aging, rejuvenation, and memory effects in Cu_0.5Co_0.5Cl_2-FeCl_3 GBIC.

Both \( \chi' \) and \( \chi'' \) were simultaneously measured with increasing \( T \) (aging ZFC curves). \( \Delta \chi' \) and \( \Delta \chi'' \) are the deviations of the aging ZFC curve from the reference ZFC curve measured with increasing \( T \) after the standard ZFC aging protocol: quenching from 20 to 3.75 K at \( H = 0 \). Similar (but more pronounced) double memory effect has been reported by Jonsson et al. for Ag (11 at. % Mn) [10]. The condition for the appearance of the dips at \( T_1 \) and \( T_2 \) (= \( T_1 - \Delta T \)) is that the overlap distance \( L_{\Delta T} \) given by Eq. (10) with \( T = T_2 \) is larger than the size \( R_{T_1}(t_{w1}) \). The spin configurations imprinted at \( T_1 \) are partially reinitialized by the spin configurations imprinted at \( T_2 = 3.0 \) K. Similar (but more pronounced) double memory effect has been reported by Jonsson et al. for Ag (11 at. % Mn) [10]. The condition for the appearance of the dips at \( T_1 \) and \( T_2 \) (= \( T_1 - \Delta T \)) is that the overlap distance \( L_{\Delta T} \) given by Eq. (10) with \( T = T_2 \) is larger than the size \( R_{T_1}(t_{w1}) \).

![Fig. 12](image)

**Fig. 12.** \( T \) dependence of (a) \( \Delta \chi' \) and (b) \( \Delta \chi'' \), double memory experiment. \( f = 0.05 \) Hz, \( h = 0.1 \) Oe, \( H = 0 \). The measurement was carried out after the ZFC aging protocol: quenching of the system from 20 to 3.75 K at \( H = 0 \), isothermal aging at 3.75 K for 10 hours, quenching from 3.75 to 3.0 K, isothermal aging at 3.0 K for 10 hours, and quenching from 3.0 to 1.9 K. Both \( \chi' \) and \( \chi'' \) were simultaneously measured with increasing \( T \) (aging ZFC curves). \( \Delta \chi' \) and \( \Delta \chi'' \) are the deviations of the aging ZFC curve from the reference ZFC curve measured with increasing \( T \) after the standard ZFC aging protocol: quenching from 20 to 1.9 K at \( H = 0 \). For the double memory effect are smaller than those for the single memory effect. It seems that the spin configurations which are imprinted at \( T_1 = 3.75 \) K during the cooling process may be partially reinitialized by the spin configurations imprinted at \( T_2 = 3.0 \) K. Similar (but more pronounced) double memory effect has been reported by Jonsson et al. for Ag (11 at. % Mn) [10]. The condition for the appearance of the dips at \( T_1 \) and \( T_2 \) (= \( T_1 - \Delta T \)) is that the overlap distance \( L_{\Delta T} \) given by Eq. (10) with \( T = T_2 \) is larger than the size \( R_{T_1}(t_{w1}) \). The spin configurations imprinted at \( T_1 \) is partially reinitialized by the spin configurations imprinted at \( T_2 = 3.0 \) K when \( \Delta T \) is larger than the threshold temperature \( (\Delta T)_t \) given by

\[
(\Delta T)_t = \left( \frac{T_0^{3/2}}{T_1^{1/2}} \right) \left( \frac{t_{w1}}{t_0} \right)^{-\zeta/(T_1^*)}
\]

When \( t_{w1} = 3.6 \times 10^4 \) sec, \( t_0 = \tau^* = 5.29 \times 10^{-6} \) sec, and \( \zeta = 0.385 \), \( (\Delta T)_t \) is estimated as \( (\Delta T)_t = 0.14 T_0^{-1/2} \) using the value of \( T_0^* \) (\( = 1.12 - 1.8 \) K) obtained in Sec. 4.4. The value of \( (\Delta T)_t \) is estimated as \( (\Delta T)_t = 0.16 - 0.33 \) K, which is smaller than \( \Delta T = 0.75 \) K.

**4.6 Rejuvenation effect in \( \chi' \) and \( \chi'' \) under the \( T \) - and \( H \)-shifts**

The rejuvenation and memory effects in \( \chi' \) and \( \chi'' \) under the \( T \)-shift are observed in our system. Figures (a) and (b) show the \( t \) dependence of \( \chi'(\omega, t) \) and \( \chi''(\omega, t) \) at \( T_1 = 3.55 \) K under the \( T \)-shift between \( T_1 = 3.75 \) K and
$T_2 = 3.55 \text{ K}$, where $f = 0.05 \text{ Hz}$ and $h = 0.1 \text{ Oe}$. Here the data at $T_1 = 3.75 \text{ K}$ are not shown (see the data of $\chi''$ at $T_1 = 3.75 \text{ K}$ in the previous paper [26]). First our system was quenched from 10 K to $T_1 = 3.75 \text{ K}$ at $H = 0$. The origin of $t$ ($t = 0$) is a time when $T$ becomes $T_1$. The relaxation of $\chi'$ and $\chi''$ was measured as a function of $t$ during a period $t_{w1} = (\approx 8.2 \times 10^3 \text{ sec})$. The temperature was then changed to $T_2$ (the negative $T$-shift). The relaxation of $\chi'$ and $\chi''$ was measured as a function of $t$ for a period $t_{w2} = (\approx 8.2 \times 10^3 \text{ sec})$ at $T_2$. The system was again heated back to $T_1$ (the positive $T$-shift). These processes were repeated subsequently. Just after every negative $T$-shift, both $\chi'$ and $\chi''$ do not lie on the reference curves of $\chi'$ and $\chi''$ at $T_2$ obtained when the system is quenched to $T_2$ directly from 10 K at $t = 0$. The values of $\chi'$ and $\chi''$ are larger than the reference curves, indicating the partial reinitialization (rejuvenation) in $\chi'$ and $\chi''$ after the negative $T$-shift. Just after every positive $T$-shift, however, both $\chi'$ and $\chi''$ lie on to the reference curves of $\chi'$ and $\chi''$ at $T_1$ obtained when the system is quenched to $T_1$ directly from 10 K at $t = 0$, indicating the memory effect in $\chi'$ and $\chi''$ after the positive $T$-shift. Note that the reference curve coincides with a curve where the lowest points for each relaxation in $\chi'$ and $\chi''$ are connected as a function of $t$. The strong rejuvenation effect for the negative $T$-shift is also predicted from numerical study by Takayama and Hukushima [1] using the MC simulation on the 3D Ising EA SG model. Here we note that the threshold temperature differences under the positive and negative $T$-shift between $T_1$ and $T_2$, $(\Delta T)_+$ and $(\Delta T)_-$, are given by

$$(\Delta T)_+ = \left(\frac{T_2^{3/2}}{T_1^{3/2}}\right)(tw_2/t_0)^{-\zeta/z(T_2)}$$

and

$$(\Delta T)_- = \left(\frac{T_2^{3/2}}{T_1^{3/2}}\right)(tw_1/t_0)^{-\zeta/z(T_1)}$$

(12)

respectively. When $T_1 = 3.75 \text{ K}$, $T_2 = 3.55 \text{ K}$, $t_{w1} = t_{w2} = 8.2 \times 10^3 \text{ sec}$, $t_0 = \tau^* = 5.29 \times 10^{-6} \text{ sec}$, and $\zeta = 0.385$, we have $(\Delta T)_+ = 0.1487^{3/2}$ and $(\Delta T)_- = 0.1637^{3/2}$.

The rejuvenation effect in $\chi'$ and $\chi''$ under the $H$-shift is also observed in our system. Figures 14(a) and (b) show the $t$ dependence of $\chi'$ and $\chi''$ at $T = 3.75 \text{ K}$ under the $H$-shift, where $f = 0.05 \text{ Hz}$ and $h = 0.1 \text{ Oe}$. After the ZFC aging protocol (quenching from a high temperature above $T_g$ to a temperature $T = 3.75 \text{ K}$), $\chi'$ and $\chi''$ at $H = 0$ were measured for a period $t_{w1} = (\approx 1.3 \times 10^4 \text{ sec})$, where the origin of $t$ ($t = 0$) is a time when $T$ becomes 3.75 K. The field is changed from 0 to $H = (\approx 50 \text{ Oe})$ at $t = t_{w1}$. After this $H$-shift, $\chi'$ and $\chi''$ were measured for a period $t_{w2} = (\approx 1.3 \times 10^4 \text{ sec})$. Subsequently, the field was turned off from $H = 0$ to 50 Oe and the measurements were carried out at $H = 0$ for a period $t_{w1}$. This process was repeated. We find that both $\chi'$ and $\chi''$ undergo drastic jumps under the $H$-shift (from 0 to 50 Oe) to values higher than the reference curves at $H = 50 \text{ Oe}$. They also undergo drastic jump under the $H$-shift (from 50 to 0 Oe) to values higher than the reference curves at $H = 0$. These results suggest that a partial reinitialization (rejuvenation effect) of $\chi'$ and $\chi''$ occurs for the $H$-shifts (0 to 50 Oe and 50 to 0 Oe). Note that the point at $T = 3.75 \text{ K}$ and $H \approx 50 \text{ Oe}$ is located on the de Almeida-Thouless (AT) line in the $(H,T)$ phase diagram [26]. On this AT line, the $T$-derivative $\delta H/\delta T$ shows a local minimum, where $\delta = \chi_{FC} - \chi_{ZFC}$.

When the overlap distance $L_H$ given by Eq. 7 is smaller than the size $R_T(t_w)$, the spin configuration imprinted at $H = 0$ is partially reinitialized at $H > H_1$, where the threshold magnetic field is defined as

$$H_1 = Y_H(t_w/t_0)^{-\delta/z(T)}$$

(13)

When $t_w = 3.6 \times 10^4 \text{ sec}$, $t_0 = \tau^* = 5.29 \times 10^{-6} \text{ sec}$, $\delta = 1.37$, and $T = 3.75 \text{ K}$, $H_1$ is estimated as $H_1 = 8.67 \times 10^{-3} \text{T}_{H_1}$. Using the value of $\text{T}_{H_1} = 2 - 3 \text{kOe}$ obtained in Sec. 4, the value of $H_1$ is estimated as $17 - 25 \text{ Oe}$, which is lower than 50 Oe.
5 Discussion and conclusion

The non-equilibrium nature of the spin dynamics in 3D Ising SG Cu$_{0.5}$Co$_{0.5}$Cl$_2$-FeCl$_3$ GBIC has been studied from the $t$ dependence of $\chi_{ZFC}$, $\chi'$, and $\chi''$ after specific ZFC protocols including the $T$-shift ($\Delta T$) and $H$-shifts. The relaxation rate $S(t)$ shows a peak at $t_{cr}$ ($\approx t_w$), corresponding to a crossover from quasi-equilibrium dynamics to non-equilibrium dynamics. The value of $t_{cr}$ strongly depends on the wait time $t_w$, $T$, $H$, and $\Delta T$. The rejuvenation effects are observed in $\chi'$ and $\chi''$ under the negative $T$-shift and both the positive and negative $H$-shifts. The spin configurations imprinted under the ZFC aging protocols are recalled on heating the system.

We find that the observed change of $t_{cr}$ under the $T$- and $H$-shifts is well explained in terms of the scaling relations where the overlap lengths $L_{\Delta T}$ and $L_H$ play a significant role. Under the $T$-shift from $T = T_i$ to $T_f$ ($= T_i + \Delta T$), the size of domains are unaffected for sufficiently small $|\Delta T|$, where $L_{\Delta T}$ is larger than $R_{T_i}(t_w)$. Then the relaxation rate $S(t)$ shows a peak at $t_{cr}$ where $L_{T_f}(t)$ is equal to $R_{T_f}(t_w)$. In contrast, the size of domains are affected for sufficiently large $|\Delta T|$. The overlap length $L_{\Delta T}$ becomes lower than $R_{T_i}(t_w)$. Then $S(t)$ has a peak at $t_{cr}$, where $L_{T_f}(t)$ is equal to $L_{\Delta T}$. Under the $H$-shift, the size of domains are unaffected during the $H$ shift for sufficiently small $H$, where the overlap length $L_H$ is larger than $R_{T}(t_w)$. The value of $t_{cr}$ is dependent on $t_w$. In contrast, the size of domains are affected during the $H$ shift for sufficiently large $H$, where $L_H$ becomes lower than $R_{T}(t_w)$. Then $S(t)$ has a peak at $t_{cr}$, where $L_{T}(t) \approx L_H$.

We discuss the scaling relation of the $T$ dependent relaxation rate $S(T,t)$. As pointed out in Sec. 4.2, $\chi_{ZFC}(T,t)$ below $T_g$ may be described by a scaling function

$$\chi_{ZFC}(T,t) = G(x)$$

with $x = L_{T}(t)/R_{T}(t_w)$. From the definition, $S(T,t)$ is derived as

$$S(T,t) = d\chi_{ZFC}(T,t)/d\ln t = (1/z(T))H(x)$$

with $H(x) = x(dG(x)/dx)$. It follows that $S(t)$ is described by a scaling function $H(x)$ except for the factor $1/z(T)$.

When the scaling function $H(x)$ has a peak at $x = a$ (constant), then $S(t)$ has a peak at $t = t_{cr}$, where $x = L_{T}(t)/R_{T}(t_w) = a$. Then $t_{cr}$ is simply described as

$$\ln(t_{cr}/t_w) = (T_g/bT_g)\ln a$$

(16)

When $a$ is larger than 1, $\ln(t_{cr}/t_w)$ increases with decreasing $T$. In fact, the least squares fit of the data of $\ln(t_{cr}/t_w)$ vs $t$ with $t_w = 2.0 \times 10^5$ sec in Fig. 4(a) ($H = 1$ Oe) to Eq. 16 yields $T_g/bT_g \ln a = 37.1 \pm 1.9$. Since $T_g = 3.92$ K and $b = 0.16$, we have $a = 4.5 \pm 0.4$. It is predicted from Eq. 16 that $S_{max}$ increases with increasing $T$ for $T < T_g$ since $S_{max}$ is linearly dependent on the factor $1/z(T)$ ($= bT_g/T_g$). Experimentally, as shown in Fig. 3(b) with $t_w = 2.0 \times 10^5$ sec, the peak height $S_{max}$ strongly depends on $T$. The peak height $S_{max}$ exhibits a broad peak around 3.6 - 3.8 K, just below $T_g$. The linear increase of $S_{max}$ with increasing $T$ below $T_g$ is considered to be due to the factor $1/z(T)$. The decrease of $S_{max}$ with increasing $T$ above $T_g$, however, cannot be explained in terms of the above model because the scaling relation is valid only for $T < T_g$. In Sec. 4.3 we show that $S(t)/S_{max}$ at $T = 3.75$ K and $H = 5$ Oe obeys a $t/t_w$-scaling law for long $t_w$ ($5.0 \times 10^3 \leq t \leq 1.5 \times 10^4$ sec) and $0 \leq t \leq 6.0 \times 10^3$ sec (see Fig. 3(c)): $S(t)/S_{max}$ is well described by a scaling function $F(t/t_w)$ which has a peak at $t_{cr}/t_w \approx 0.68$. Using Eq. 16, the value of $a$ is estimated as $a = 1.11$, indicating that the constant $a$ at $H = 5$ Oe is different from that at $H = 1$ Oe.

Finally we consider the cause of the complicated behavior of $S(t)$ at $T = 3.75$ K for $0 \leq t_w \leq 750$ sec. One of the cause is the way how $T$ approaches 3.75 K during the ZFC aging protocol. The temperature drops rapidly to $3.50$ K and slowly approaches $3.75 \pm 0.01$ K from the below (usually) within 230 sec. The experiment of $S(t)$ under the $T$ shift (see Sec. 4.3) suggests that the initial undercool is not so important because of the temperature difference larger than the threshold temperature difference ($\Delta T$). However, the subsequent approach of $T$ to $3.75$ K for a wait time $t_{w0} \leq 230$ sec may play a significant role in the aging behavior after $t = 0$ at $T_m = 3.75$ K. The threshold temperature difference ($\Delta T$)$_t$ is estimated as

$$(\Delta T)_t = (T_t^{3/2}/[(T_m - (\Delta T)_t)^{1/2}]) (t_w0/t_0)^{-\zeta/z(T_m - (\Delta T)_t)}$$

(17)

from the condition that $L_{\Delta T} = R_{T - \Delta T}(t_{w0})$. When $t_0 = 230$ sec, $\zeta \approx 0.385$, and $t_{w0} = 230$ sec, ($\Delta T$)$_t$ is estimated as ($\Delta T$)$_t = 0.24$ K for $T_T = 1.12$ K and 0.56 K for $T_T = 1.80$ K. This suggests that the spin configuration at $T_m = 3.75$ K after $t = 0$ is affected by that imprinted at $T_m - \Delta T$ at the wait time $t_{w0}$. Note that the value of $t_{cr}$ at $T_m - \Delta T$ is larger than that at $T_m$ for the same $t_w$ (see Fig. 3(a)). The appearance of two peaks in $S$ vs $t$ at $0 \leq t_w \leq 200$ sec may be associated with two domains generated for the wait time $t_{w0}$ at $T_m - \Delta T$ and for $t_{w0}$ at $T_m$.

In conclusion, we have undertaken an extensive study on the $t$ dependence of the relaxation rate $S(t)$ mainly below $T_g$ under the various conditions including the $T$- and $H$-shifts. The $t$ dependence of $S$ is well explained in terms of the scaling relation. The peak of $S(t)$ occurs when the mean SG domain size $L_T(t)$ coincides with $R_{T}(t_w)$, where $t_w$ is the wait time. Our results indicate that the aging, memory, and rejuvenation phenomena observed in our system are very similar to those in conventional spin glass systems.

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