Techniques for the Studies on the Aging dynamics in Spin Glasses

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Abstract:
Here we present a brief review on the method for studying the aging dynamics of spin glass phase using SQUID magnetometer. Typical examples of our recent results are also shown.

The abbreviations used here are as follows.

FC: field-cooled
ZFC: zero-field cooled
TRM: thermoremnant magnetization
IRM: ithothermal remnant magnetization
t_w: wait time
t_s: stop time
t_a: age of the system
T_g: spin freezing temperature
$S(t)$: relaxation rate
Stretched exponential decay

I. DC magnetization Measurements

A. Time (t) dependence of $M_{ZFC}(t)$ and $M_{TRM}(t)$

(a) $M_{ZFC}(t, t_w)$ after the ZFC aging protocol

The system is cooled from the high temperature well above the spin freezing temperature T_g to a temperature T_m in the absence of magnetic field H. The system is aged at T_m for a wait time t_w at $H = 0$. Then the magnetic field is turned on at $t = 0$ (the age $t_a = t_w$). The ZFC magnetization $M_{ZFC}(t)$ is measured as a function of t. The relaxation rate $S_{ZFC}(t)$ is derived from the relation

$$S_{ZFC}(t) = \frac{1}{H}\frac{dM_{ZFC}(t)}{d\ln t}. \quad (1)$$

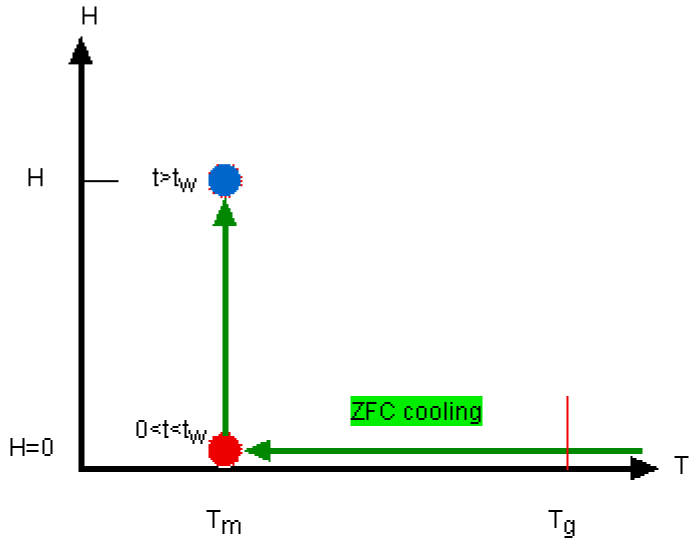

Diagram 1

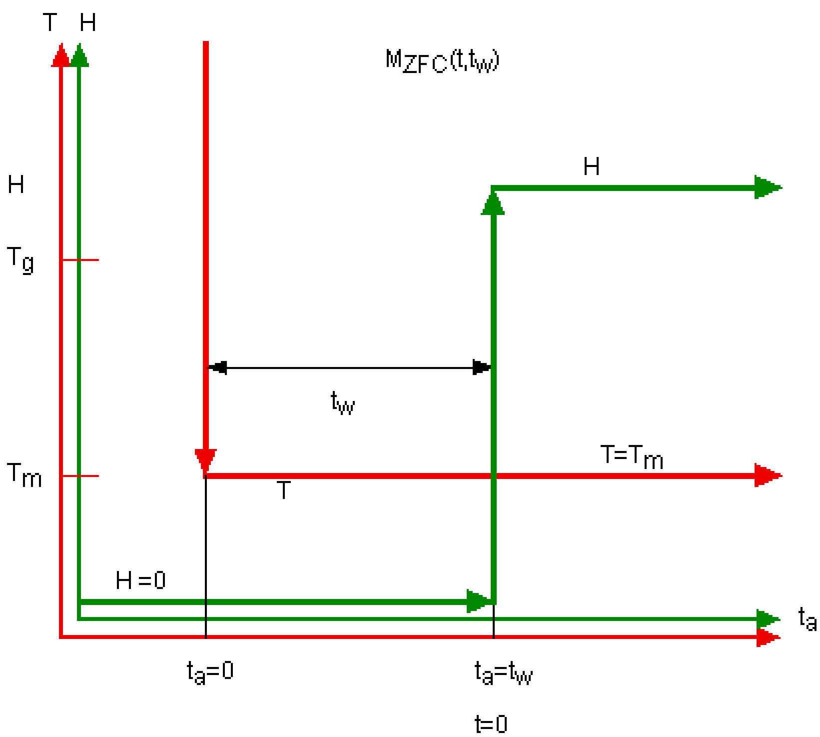

Diagram 2

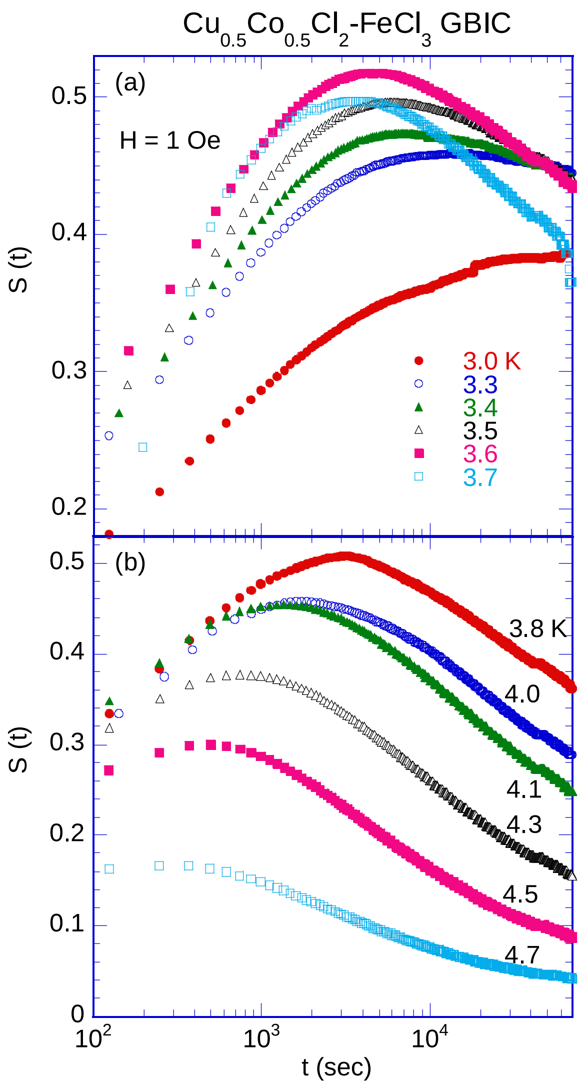

Figure A (a) and (b) t dependence of S for $3.0 \le T \le 4.7$ K. H = 1 Oe. The measurement of χ_{ZFC} vs. t was carried out after the ZFC aging protocol (annealing of the system at 50 K and H = 0 for 1.2×10^3 s, quenching from 50 K to T, and isothermal aging for $t_w = 2.0 \times 10^3$ s). t = 0 is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

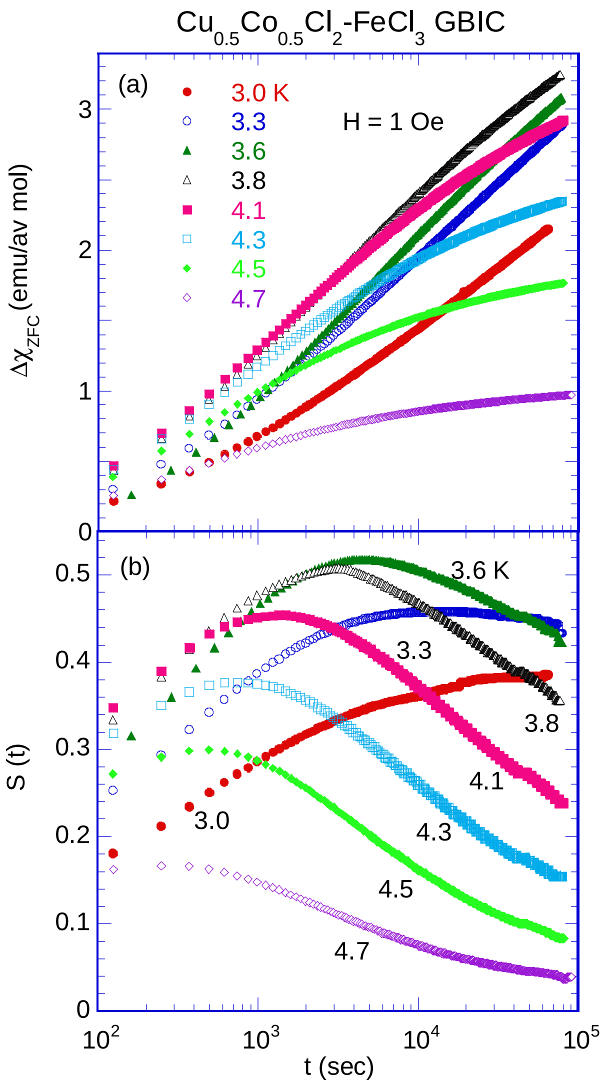

Figure B t dependence of (a) $\chi_{ZFC}(t)$ and (b) $S(t)$ at various H ($20 \leq H \leq 300$ Oe). $t_w = 1.0 \times 10^4$ sec. $T = 3.50$ K [Ref. 3].

(b) $M_{TRM}(t, t_w)$ after the FC aging protocol

The system is cooled from the high temperature well above the spin freezing temperature T_g to a temperature T_m in the presence of magnetic field H. The system is aged at T_m for a wait time t_w in the presence of H. Then the magnetic field is turned off at $t = 0$ (or the age $t_a = t_w$). The TRM magnetization $M_{TRM}(t)$ is measured as a function of t. The relaxation rate $S_{TRM}(t)$ is derived from the relation

$$S_{TRM}(t) = \frac{1}{H}\frac{dM_{TRM}(t)}{d\ln t}. \qquad (2)$$

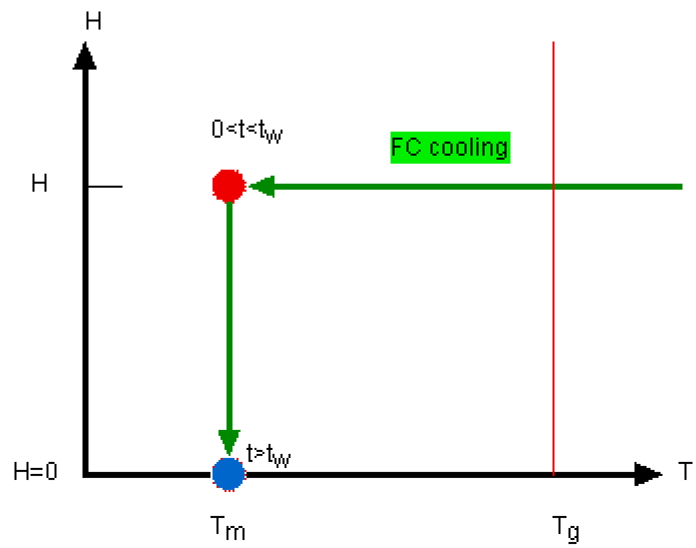

Diagram 3

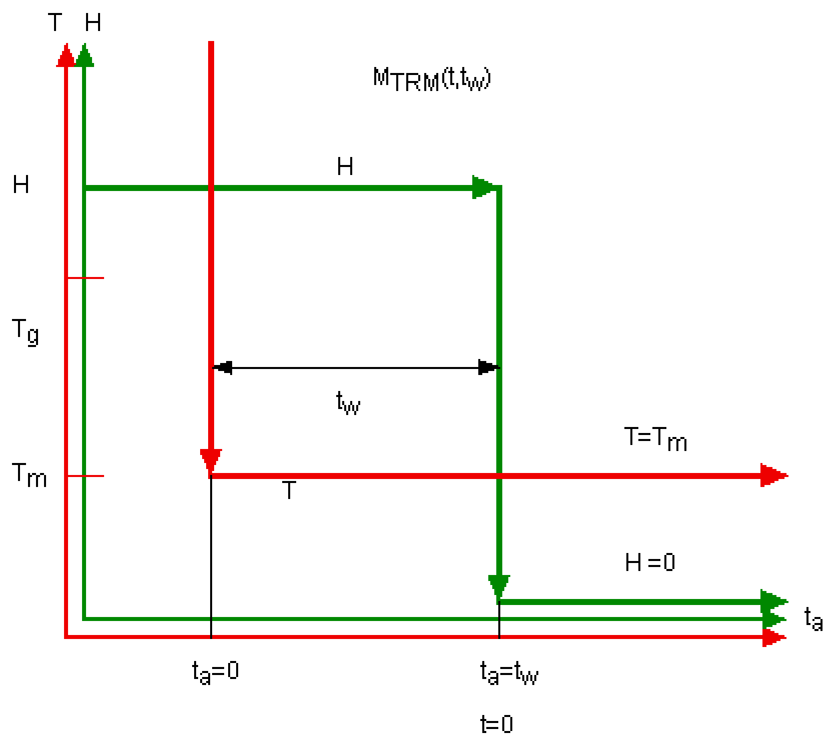

Diagram 4

B. T dependence of M_{ZFC}, M_{ZFC}, M_{TRM}, M_{IRM}

(a) M_{FC} vs T measurement

The magnetization of the system is measured with decreasing temperature while the system is slowly cooled from the high temperatures well above T_g in the presence of the magnetic field H.

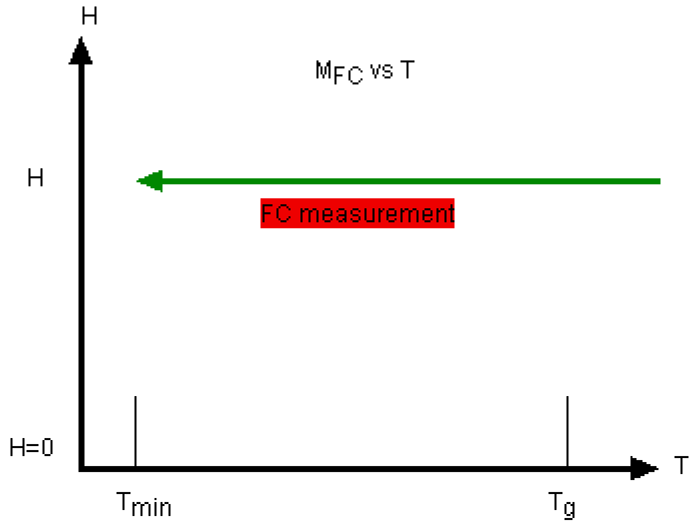

Diagram 5

(b) M_{ZFC} vs T measurement

The system is cooled from the high temperature well above T_g to a temperature T_{min} in the absence of H (= 0). The magnetic field is turned on at $T = T_{min}$. The ZFC magnetization M_{ZFC} is measured as a function of temperature with increasing T from $T = T_{min}$ at H.

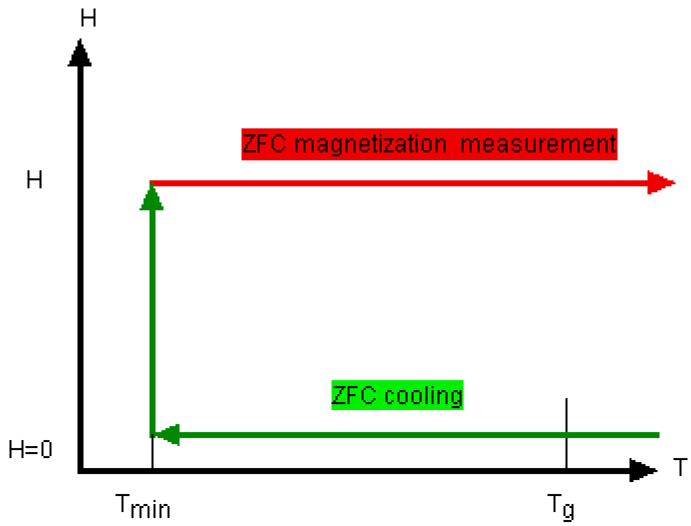

Diagram 6

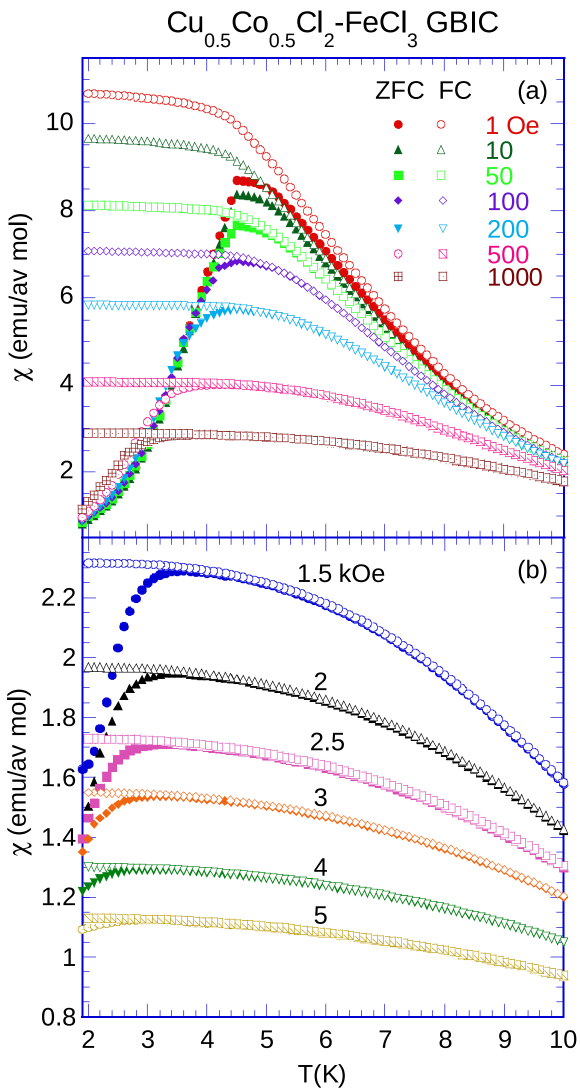

Figure C (a) and (b) *T* dependence of χ_{ZFC} and χ_{FC} at various *H* for $Cu_{0.5}Co_{0.5}Cl_2$-$FeCl_3$ GBIC. *H* is applied along *c* plane (graphene basal plane) perpendicular to the *c* axis [Ref. 1].

(c) M_{TRM} vs *T* measurement

The system is cooled from the high temperature well above the spin freezing temperature T_g to a temperature T_{min} in the presence of magnetic field *H*. The magnetic field is turned off at $T = T_{min}$. The TRM magnetization M_{TRM} is measured as a function of temperature with increasing *T* from $T = T_{min}$ at $H = 0$.

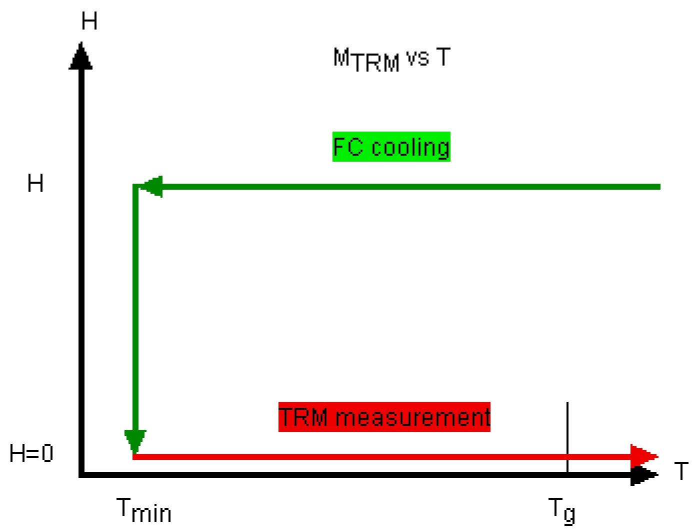

Diagram 7

(d) M_{IRM} vs T measurement

The system is cooled from the high temperature well above the spin freezing temperature T_g to a temperature T_{min} in the absence of magnetic field H. The system is aged at T_{min} for a wait time t_w at $H = 0$. Then the magnetic field is turned on at H and subsequently it is turned off. The IRM magnetization $M_{ZFC}(t)$ is measured as a function of temperature with increasing T from $T = T_m$ at $H = 0$.

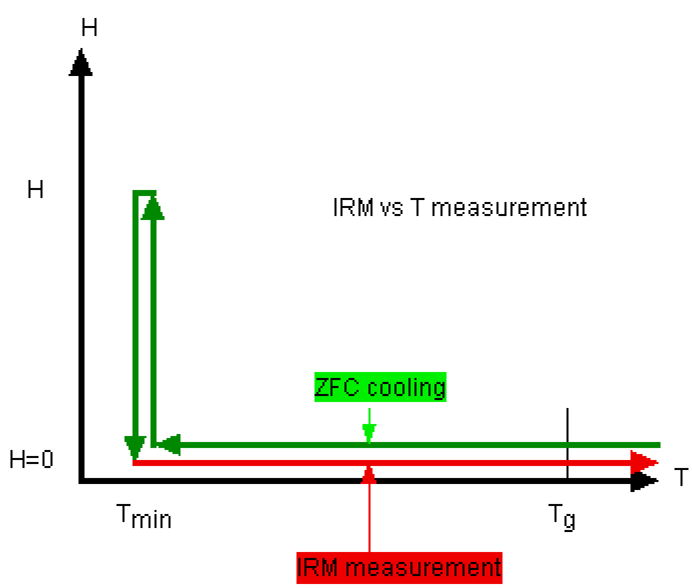

Diagram 8

C.   Time dependence of M_{ZFC} and M_{TRM} under the T shift

The system is cooled from the high temperature well above the spin freezing temperature T_g to a temperature T_i (the initial temperature) in the absence of magnetic field H. The system is aged at T_i for a wait time t_w at $H = 0$. Then temperature is shifted to the final temperature T_f (= $T_i \pm \Delta T$) at H = 0. Immediately after the magnetic field is turned on at t = 0 (the age t_a = t_w), the ZFC magnetization $M_{ZFC}(t)$ is measured as a function of t at $T = T_f$.

(a) M_{ZFC}(t) under the positive T shift

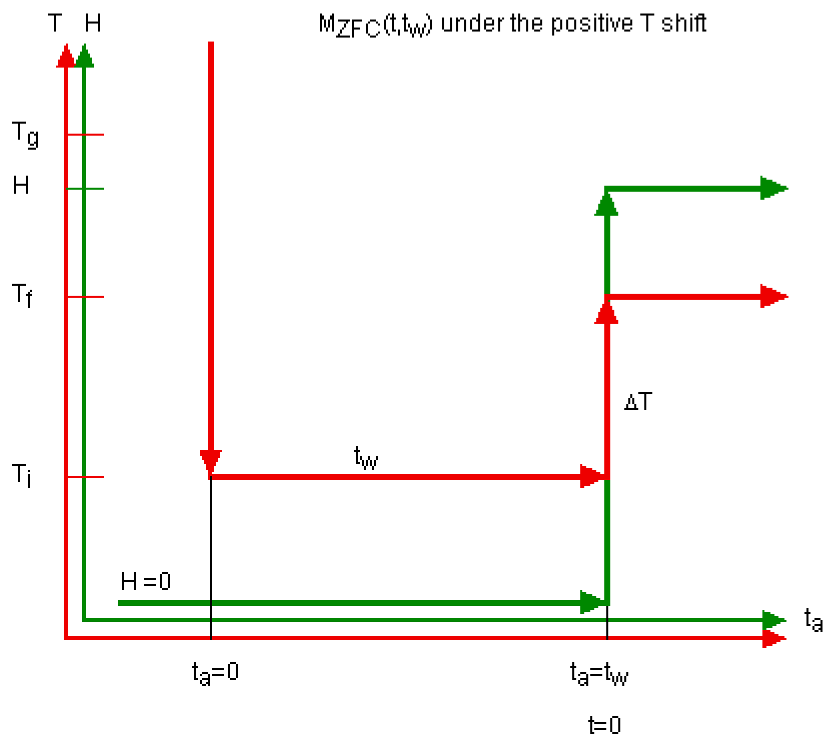

Diagram 9

(b) M_{ZFC}(t) under the negative T shift

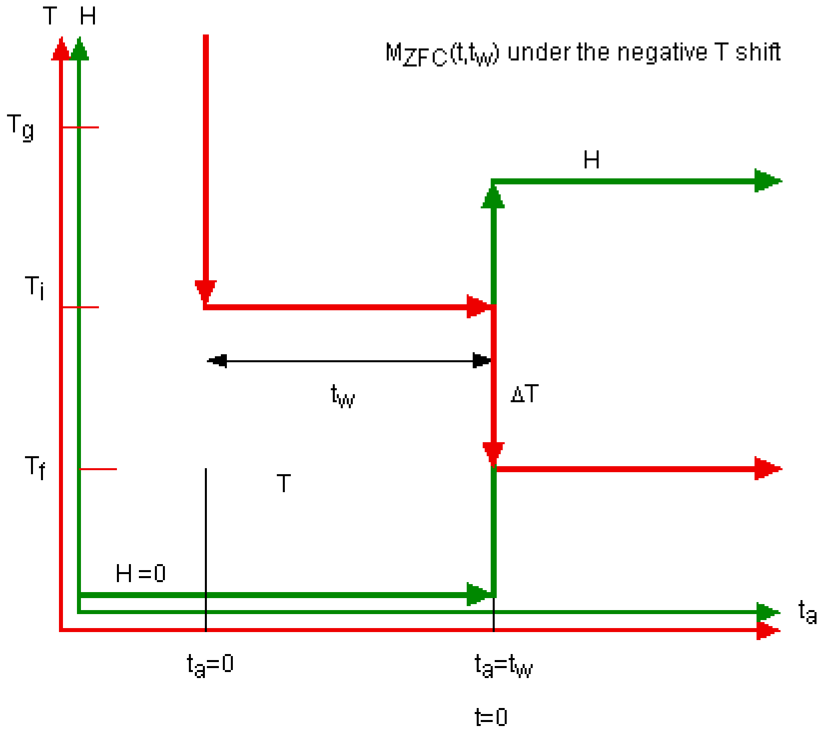
T
H
M_ZFC(t,t_w) under the negative T shift
Tg
H
Ti
tw
ΔT
Tf
T
H =0
ta
ta=0
ta=tw
t=0

Diagram 10

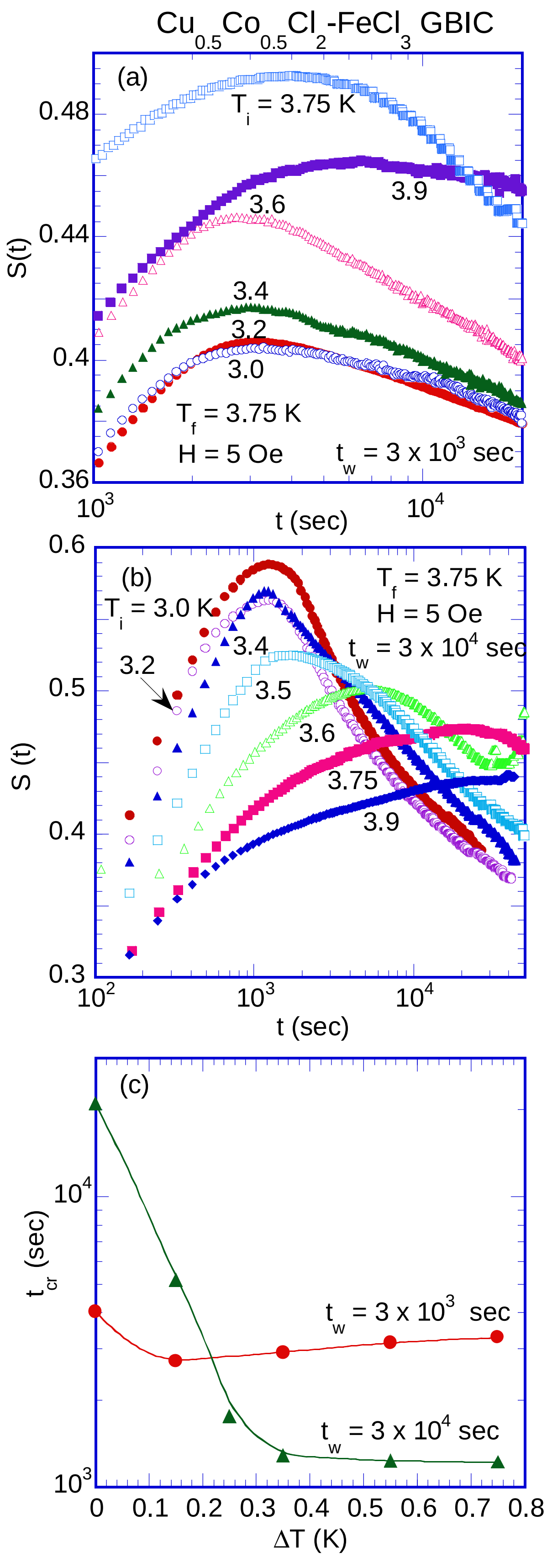

Figure D t dependence of S at T_f = 3.75 K under the T shift from T_i to T_f, where T_i = 3.0 - 3.9 K. (a) t_w = 3.0 × 10^3 s. (b) tw = 3.0 × 10^4 s. The ZFC aging protocol is as follows: quenching of the system from 50 K to T_i, and isothermal aging at T = T_i and H = 0 for t_w. The measurement was started at t = 0, just after T was shifted from T_i to T_f = 3.75 K and subsequently H (= 5 Oe) was turned on. (c) t_{cr} vs. ΔT for the case of positive T-shift (t_w = 3.0 × 10^3 and 3.0 × 10^4 s), where T_i = T_f - ΔT (ΔT > 0) and T_f = 3.75 K. The solid lines are guides to the eye [Ref. 2].

D. Genuine TRM and ZFC magnetization measurement

(a) Genuine TRM measurement

The system is cooled from high temperature well above T_g to a stop temperature T_s ($<T_g$) in the presence of the magnetic field (H). The system is aged at $T = T_s$ for a wait time ts. at H. Then the system is cooled from T_s to a minimum temperature T_{min} at H. The magnetic field is turned off at T_{min}. The magnetization $M_{TRM}(T, T_s, t_s)$ is measured with increasing temperature in the absence of H.

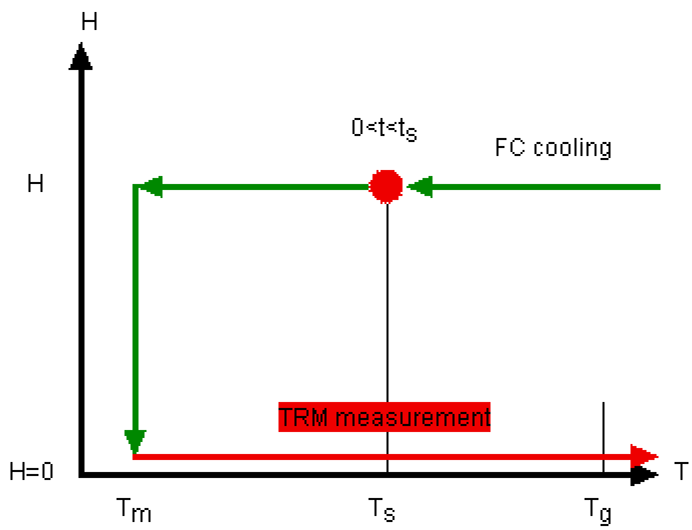

Diagram 11

The reference curve is measured as follows. The system is cooled from high temperature well above T_g to T_{min} in the presence of H. The magnetic field is turned off at T_{min}. The magnetization $M_{TRM}(T, \text{ref})$ is measured with increasing temperature in the absence of H.

The genuine TRM magnetization is defined as the difference between $M_{TRM}(T, T_s, t_s)$ and $M_{TRM}^{ref}(T, \text{ref})$.

$$\Delta M_{TRM}(T, T_s, t_s) = M_{TRM}(T; T_s, t_s) - M_{TRM}^{ref}(T). \qquad (3)$$

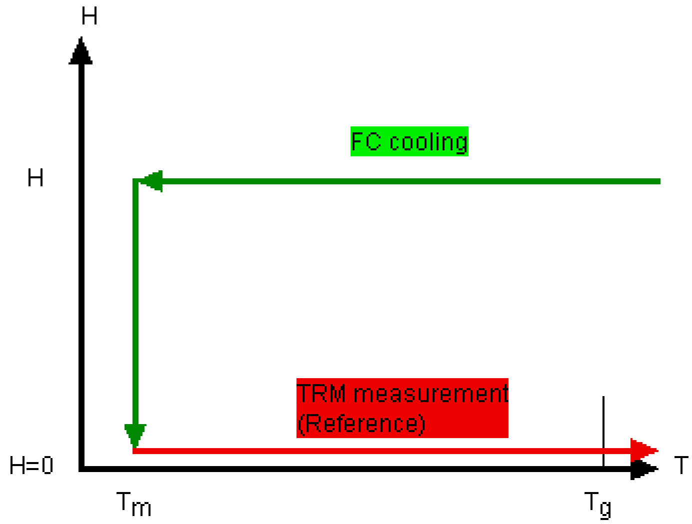

Diagram 12

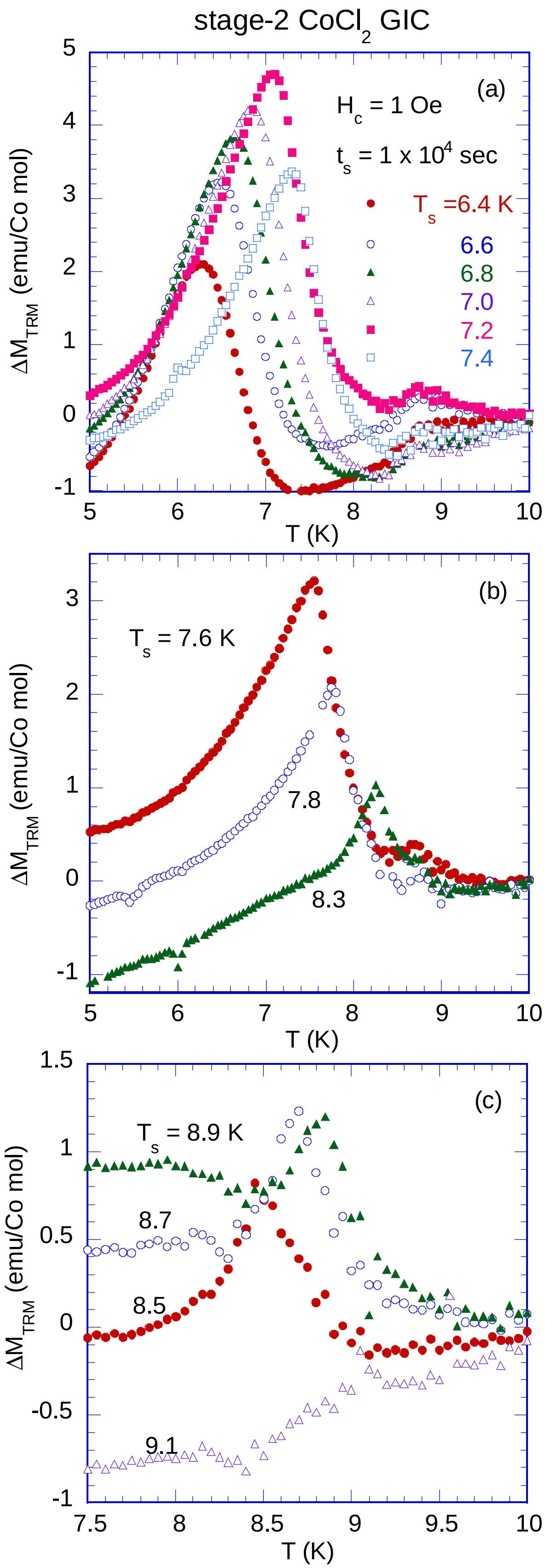

Figure E T dependence of $\Delta M_{\mathrm{TRM}}(T;T_s,t_s)$ [= $M_{\mathrm{TRM}}(T;T_s,t_s)$ - $M_{\mathrm{TRM}}^{\mathrm{ref}}(T)$], $t_s = 1.0 \times 10^4$ sec. $6.4 \leq T_s \leq 9.1$ K. $H_c = 1$ Oe. (a) $6.6 \leq T \leq 7.4$ K. (b) $7.6 \leq T \leq 8.3$ K. (c) $8.5 \leq T \leq 9.1$ K [Ref. 4].

(b) Genuine ZFC measurement

The system is cooled from high temperature well above T_g to a stop temperature T_s ($<T_g$) in the absence of the magnetic field ($H = 0$). The system is aged at $T = T_s$ for a wait time ts. at H =0. Then the system is again cooled from T_s to a minimum temperature T_{min} at $H = 0$ The magnetic field is turned on at T_{min}. The magnetization $M_{ZFC}(T, T_s, t_s)$ is measured with increasing temperature in the presence of H.

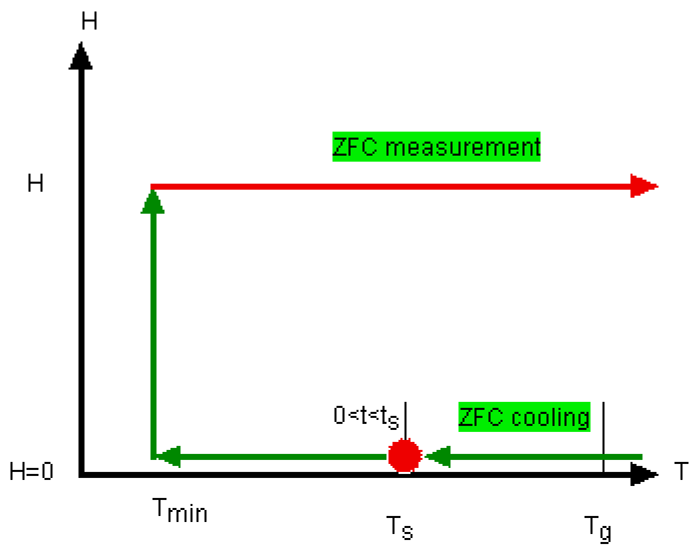

Diagram 13

The reference curve is measured as follows. The system is cooled from high temperature well above T_g to T_{min} in the absence of H. The magnetic field is turned on at T_{min}. The magnetization $M_{ZFC}(T, \text{ref})$ is measured with increasing temperature in the presence of H.

The genuine ZFC magnetization is defined as the difference between $M_{ZFC}(T, T_s, t_s)$ and $M_{ZFC}^{ref}(T, \text{ref})$.

$$\Delta M_{ZFC}(T, T_s, t_s) = M_{ZFC}(T; T_s, t_s) - M_{ZFC}^{ref}(T). \quad (4)$$

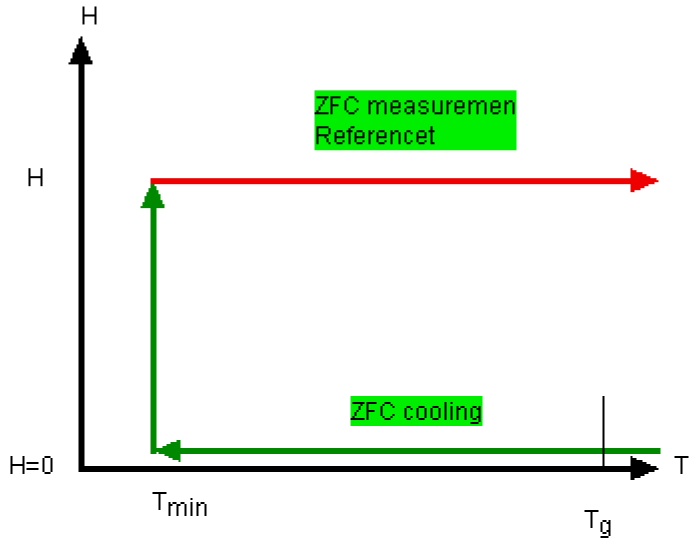

Diagram 14

E. Memory effect proposed by Sun et al. (University of Illinois, 2003)

We present a peculiar memory effect observed in our system using a unique FC cooling protocol. First the system is cooled through the FC cooling protocol from high temperature well above T_g to intermittent stop temperatures T_s in the presence of H_c. When the system is cooled down to T_s the field was cut off (H = 0) and aged at T_s for t_s (typically 3.0x10^4 sec). In this case, the magnetization $M_{FC}^{IS}(T\downarrow)$ decreases with time due to the relaxation. After the wait time t_s at each stop temperature T_s, the field (H_c) is applied again and the FC cooling process is resumed. Such a FC cooling process leads to a step-like behavior of $M_{FC}^{IS}(T\downarrow)$ curve. The value of $M_{FC}^{IS}(T\downarrow)$ after resuming below the lowest stop temperature behaves almost in parallel to that of the FC magnetization without the intermittent stops (as reference curve). After reaching the minimum temperature (typically 1.9 K), the magnetization $M_{FC}(T\uparrow)$ was measured in the presence of H (= H_c) as the temperature is increased at the constant rate (typically 0.05 K/min). The magnetization $M_{FC}(T\uparrow)$ thus measured exhibits broad peaks near T_s. The spin configuration imprinted at the intermittent stop at T_s for a wait time t_s at H = 0 during the FC cooling process strongly affects the T dependence of $M_{FC}(T\uparrow)$ when the temperature is increased, exhibiting a peculiar memory effect.

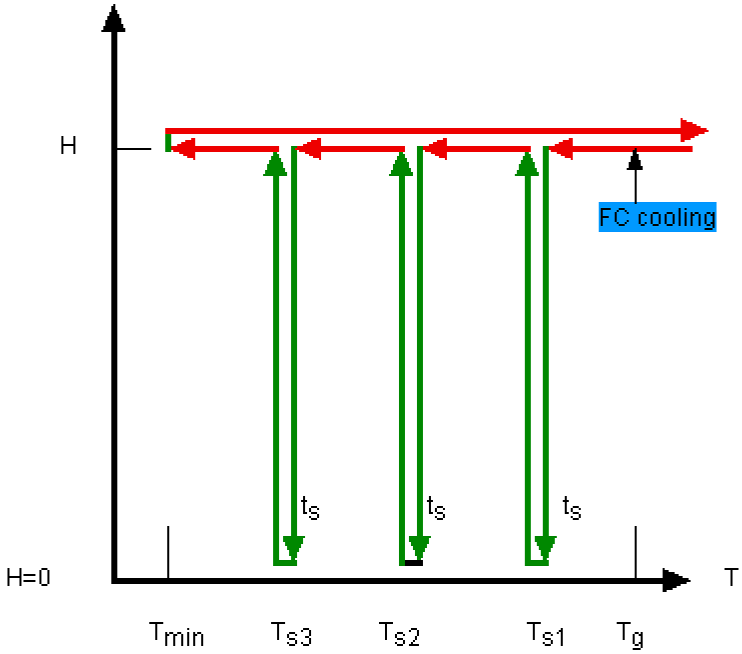

Diagram 15

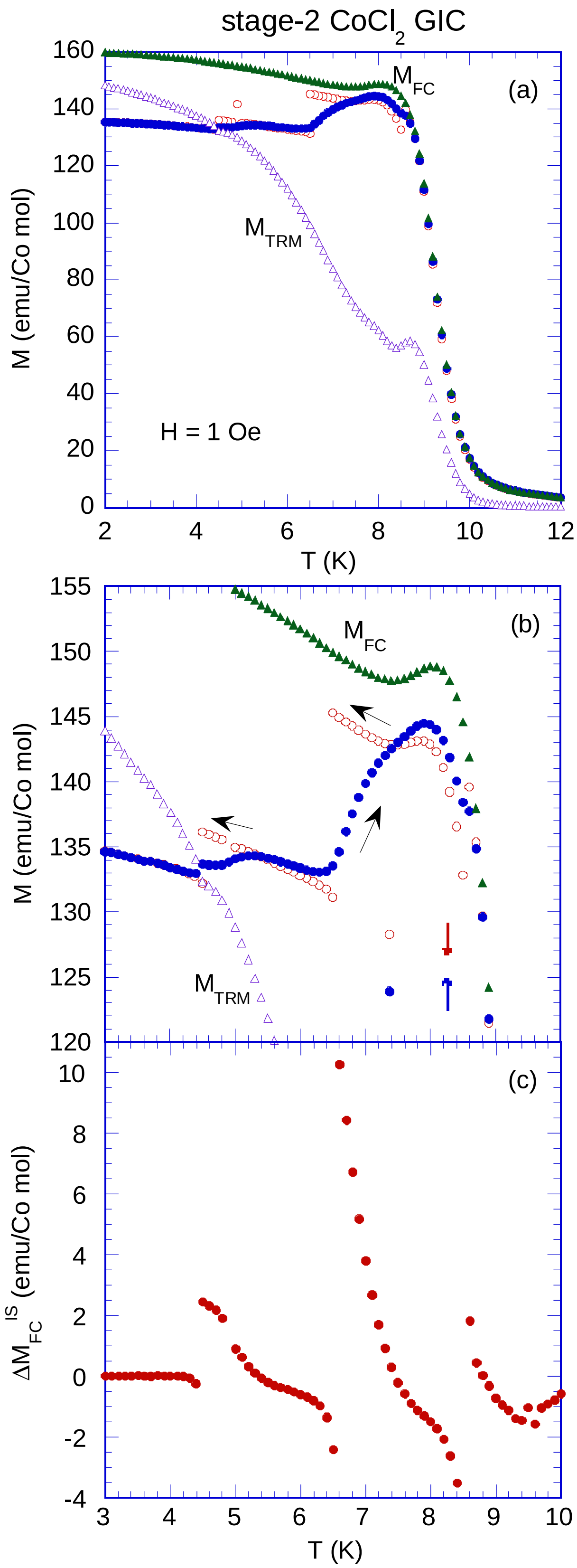

Figure F (a) and (b) T dependent of $M_{FC}^{IS}(T\downarrow)$ and $M_{FC}^{IS}(T\uparrow)$ observed in the following FC cooling protocol. The system was quenched from 50 to 12 K in the presence of H (= 1 Oe). $M_{FC}^{IS}(T\downarrow)$ was measured with decreasing T from 12 to 1.9 K but with intermittent stops at T = 8.5, 6.5, 4.5 K for a wait time t_w = 3.0 × 10^4 sec. The field is cut off during each stop. $M_{FC}^{IS}(T\uparrow)$ was measured at H = 1 Oe with increasing T after the above cooling process. The T dependence of M_{FC}^{ref} and M_{TRM}^{ref} are also shown as reference curves. These are measured after the FC cooling protocol without intermittent stop (reference curves). (c) T dependence of the difference ΔM_{FC}^{IS} [$= M_{FC}^{IS}(T\downarrow) - M_{FC}^{IS}(T\uparrow)$] [Ref. 4].

F. Memory effect proposed by Matsuura

(a) The TRM case

Before the TRM magnetization measurement, a field cooling (FC) protocol was carried out, consisting of (a) annealing of the system at high temperatures well above T_g for typically 1200 sec in the presence of H , (b). quenching of the system from this temperature to the initial temperature T_i and (c) aging the system at $T = T_i$ at H for a wait time t_w = 100 sec. Just after the magnetic field was turned off, the TRM magnetization was measured with increasing T from T_i to T_1 (the U-turn temperature) and subsequently with decreasing T from T_1 to T_i. (the cooling process). In turn, it was measured with increasing T from T_i to T_2 (the heating process) and subsequently with decreasing T from T_2 to T_i (the cooling process). This process was repeated for the U-turn temperatures T_r (r = 3, 4, 5, …), where $T_g > T_r > T_i$. The schematic diagram of these processes for the TRM measurement is shown in **Fig.(a)**.

(b) The ZFC case

Before the ZFC magnetization measurement, a zero-field cooling (ZFC) protocol was carried out. It consists of the following process, (a) annealing of the system at high temperatures well above T_g for tyoically 1200 sec in the absence of H, (b) quenching of the system from this temperature to the initial temperature T_i ($<T_g$), and (c) aging at T_i and $H = 0$ for a wait time t_w = 100 sec. Just after the magnetic field (H = 1 Oe) is applied to the system, the ZFC magnetization M_{ZFC} was measured using the same procedure of heating and cooling: $T_i \rightarrow T_1 \rightarrow T_i \rightarrow T_2 \rightarrow T_i \rightarrow T_3 \rightarrow T_i \rightarrow$ and so on, where T_r (r = 1, 2, …) is the U-turn temperature and $T_r > T_i$. The schematic diagram of these processes is shown in **Fig. (b)**.

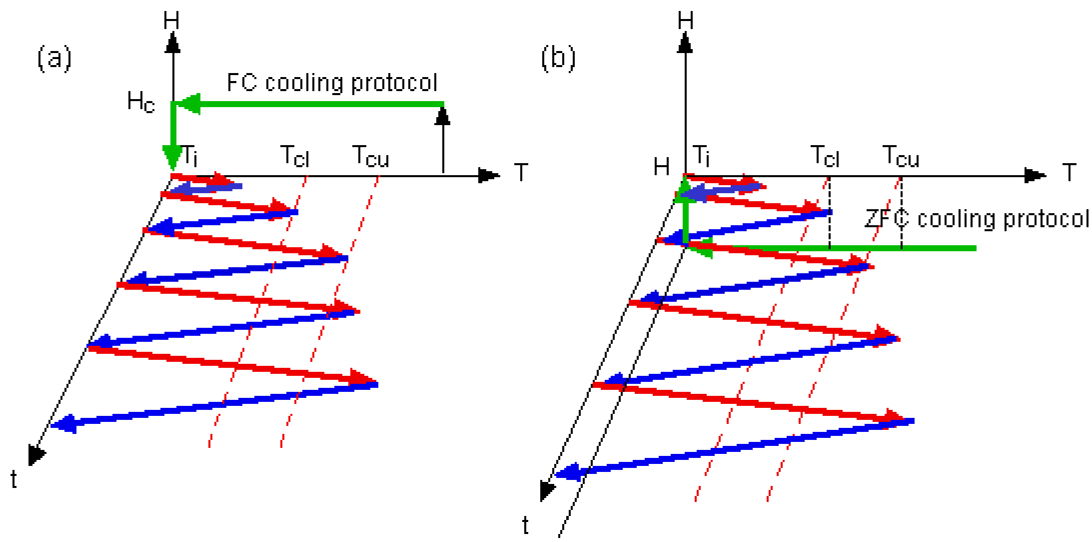

Diagram 16

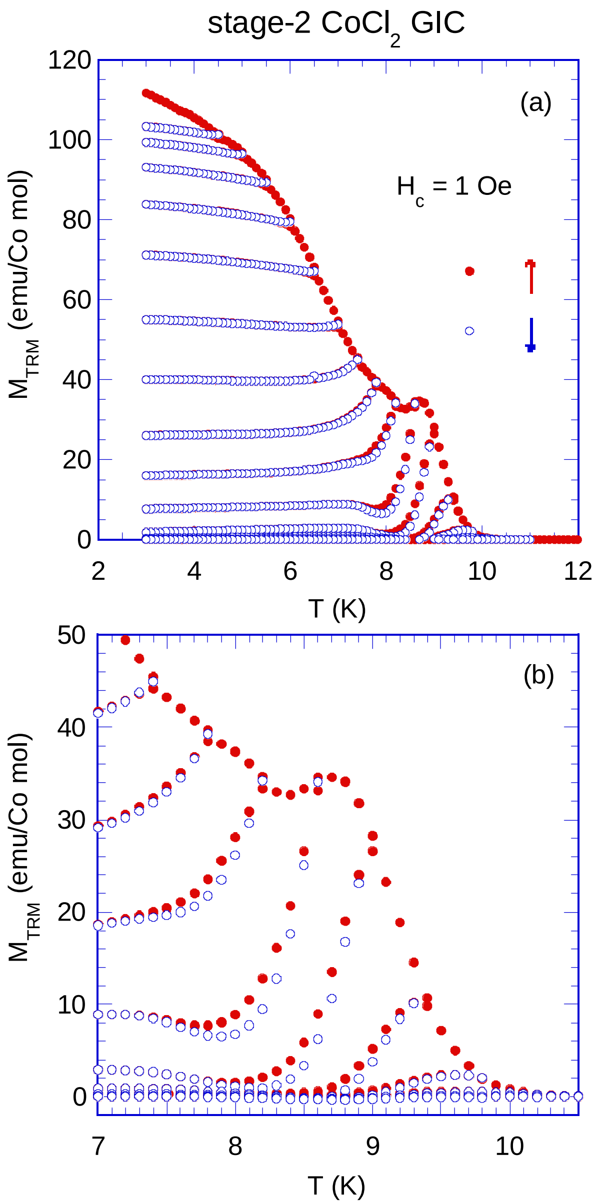

Figure G (a) and (b) T dependence of M_{TRM} measured at H = 0 in a series of heating (closed circles) and cooling (open circles) process after the FC cooling of the system from 50 to T_i = 3 K in the presence of H_c (= 1 Oe). Note the data of M_{TRM} shown here is a corrected one by subtraction of the original data from the M_{FC} part under the remanent magnetic field (≈ 5 mOe) [Ref. 4].

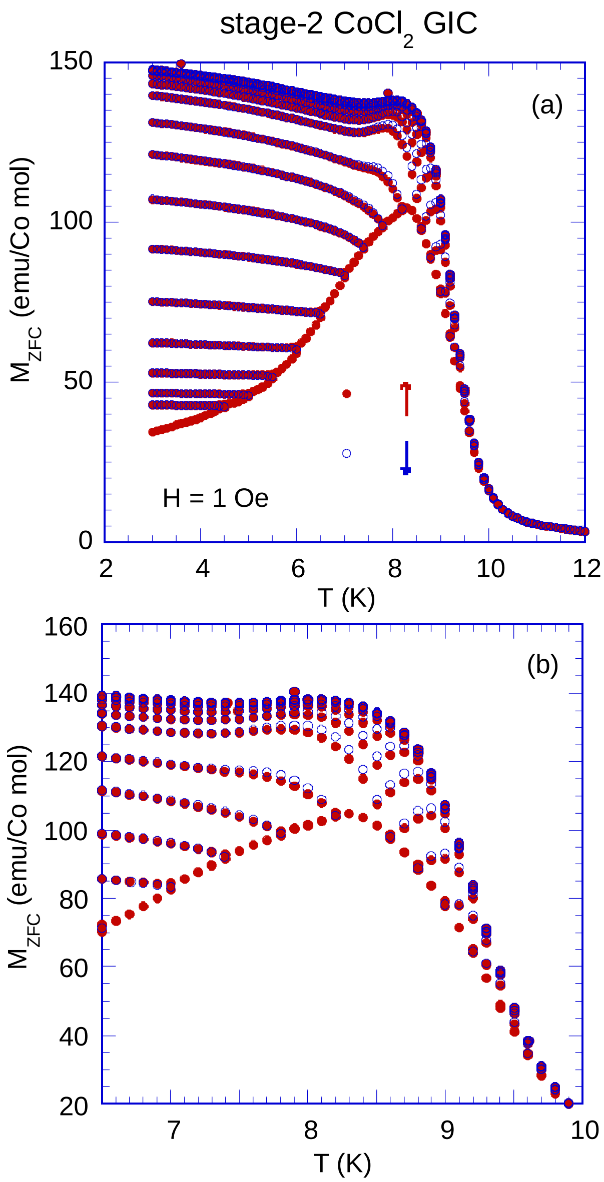

Figure H (a) and (b) *T* dependence of M_{ZFC} measured at H = 1 Oe in a series of heating (closed circles) and cooling (open circles) processes, after the ZFC cooling of the system from 50 to 3 K in the absence of H [Ref. 4].

II AC magnetic susceptibility

A. *t* dependence of χ' and χ'' at various frequencies

The system was quenched from high temperature well above Tg to T ($<T_g$) at time (age) t_a = 0. Both χ' and χ'' are measured simultaneously as a function of time t at constant T. Each point consists in the successive measurements of various frequencies. The absorption χ'' decreases with increasing t and is well described by a single scaling function given by

$$\chi''(\omega, t) = \chi''_0(\omega) + A_0''(\omega t)^{-b''}. \quad (5)$$

and corresponds to the asymptotic *f*-dependent value so that χ'(ω, *t*) tends to

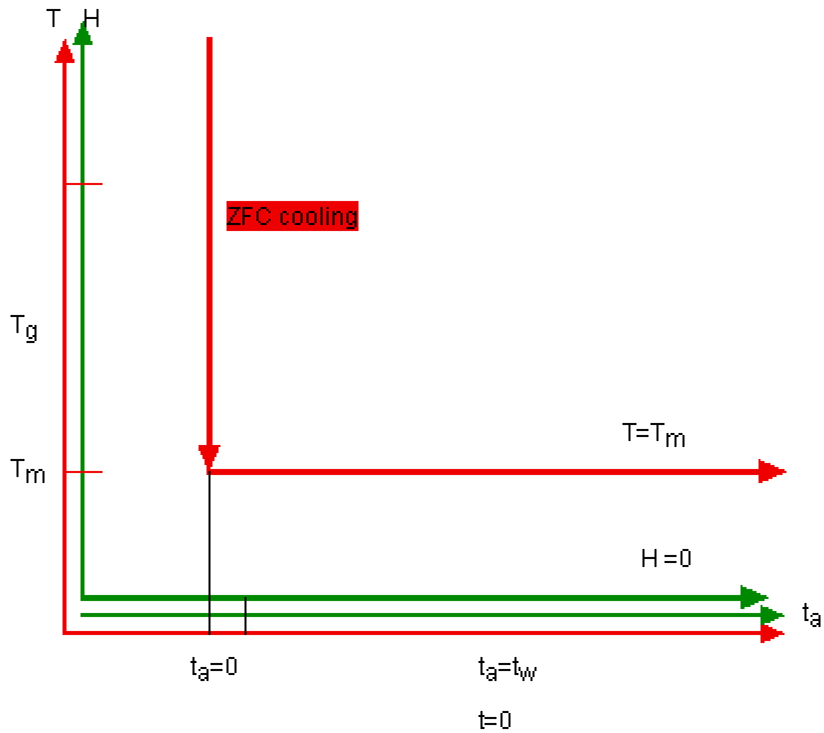

Diagram 17

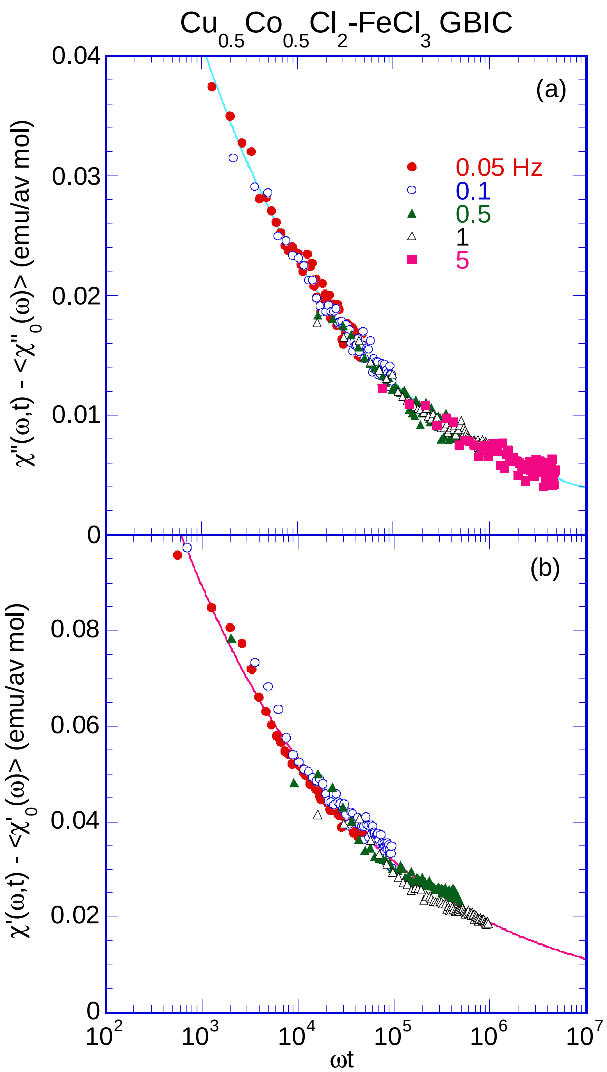

Figure I Scaling plot of (a) $\Delta\chi''(\omega,t)$ [= $\chi''(\omega,t)$ − $\langle\chi''_0(\omega)\rangle$] and (b) $\Delta\chi'(\omega,t)$ [= $\chi'(\omega,t)$ − $\langle\chi'_0(\omega)\rangle$] as a function of ωt for the data at f = 0.05, 0.1, 0.5, 1, and 5 Hz. The definition of stationary susceptibility $\langle\chi''_0(\omega)\rangle$ and $\langle\chi'_0(\omega)\rangle$ is given in the text. The solid lines are the data to the power-law form. Each curve is vertically shifted by $\langle\chi''_0(\omega)\rangle$ or $\langle\chi'_0(\omega)\rangle$ [Ref. 1].

B. Single and double Memory effects

When the system is cooled down to a low temperature 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. In a single memory experiment, the memory is imprinted at T_1 for t_s during the ZFC aging protocol. In a double memory experiment, the memory is imprinted at T_1 for t_{s1} and at T_2 ($<T_1$) for t_{s2} 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. 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. One can define the difference between the aging ZFC and reference ZFC susceptibilities as

$$\Delta\chi'(\omega, T) = \chi'_{mem}(\omega, T) - \chi'_{ref}(\omega, T), \tag{6}$$

$$\Delta\chi''(\omega, T) = \chi''_{mem}(\omega, T) - \chi''_{ref}(\omega, T), \tag{7}$$

(a) Single memory effect

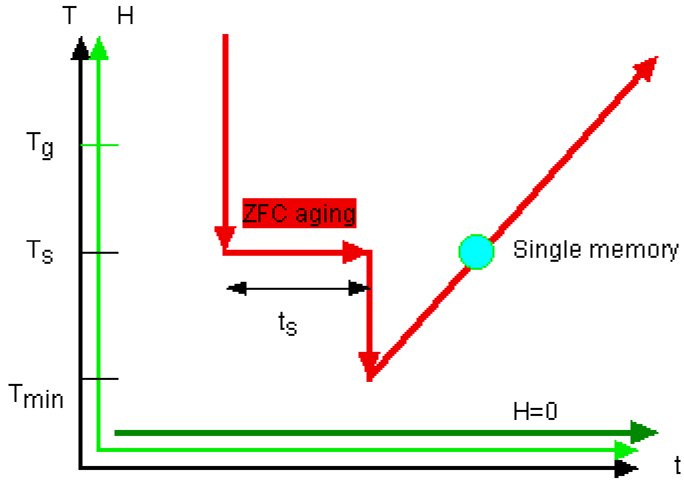

Diagram 18

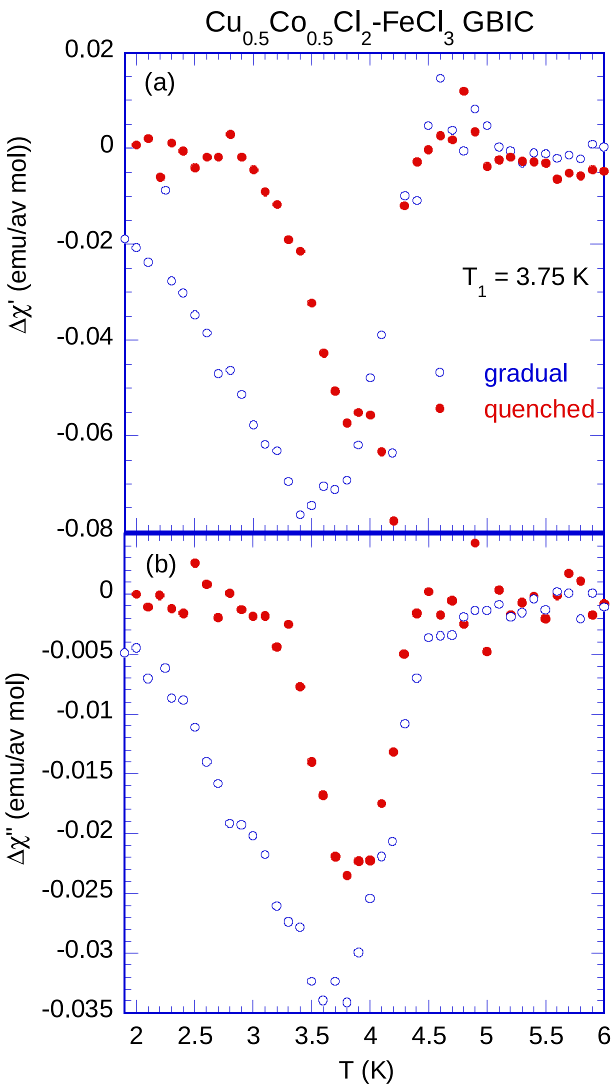

Figure J T dependence of (a) $\Delta\chi'$ and (b) $\Delta\chi''$: single memory experiments (I and II). f = 0.05 Hz. h = 0.1 Oe. The measurement (I) (denoted as gradual) was carried out after the ZFC aging protocol (I): gradual decrease of T from 20 to 3.75 K, isothermal aging at T_1 = 3.75 K for 6.3 hours, and further gradual decrease from 3.75 to 1.9 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 1.9 K. Both χ' and χ'' 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 [Ref. 2].

(b) Double memory effect

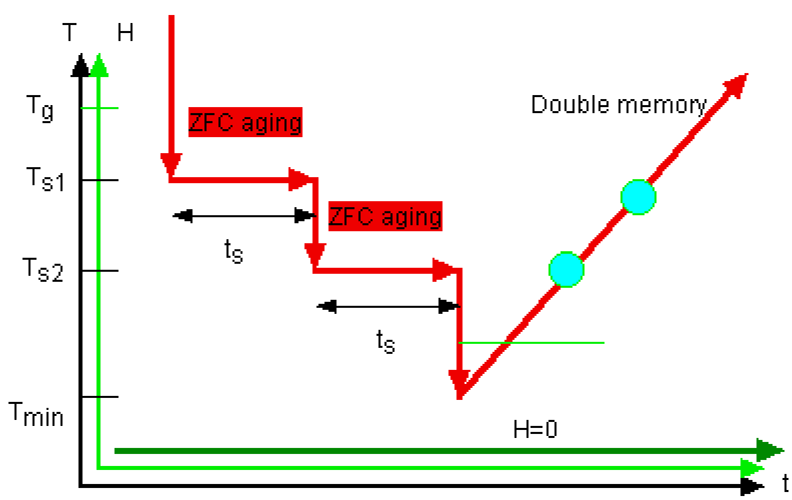

Diagram 19

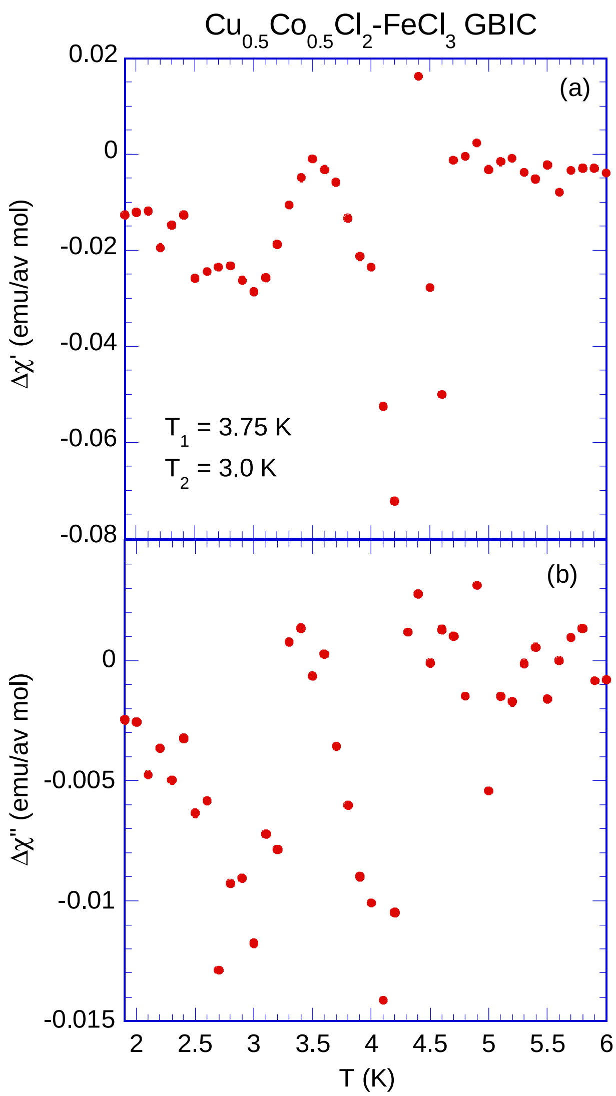

Figure K T dependence of (a) $\Delta\chi'$ and (b) $\Delta\chi''$: double memory experiment. f = 0.05 Hz. h = 0.1 Oe. H = 0. The measurement was carried out after the ZFC aging protocol: quenching of the system from 30 to 3.75 K at H = 0, isothermal aging at 3.75 K for 10 hours, quenching from 3.0 to 1.9 K. Both χ' and χ'' 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 [Ref. 2].

C. The rejuvenation and memory effects in $\chi'(\omega,t,H)$ and $\chi''(\omega,t,H)$ under the *T* shift

The system is quenched from high temperature well above T_g to T_1 at H = 0. The origin of t (t = 0) is a time when T becomes T_1. The relaxation of χ' and χ'' is measured as a function of t during a period t_{s1}. The temperature was then changed to T_2 (the negative T-shift). The relaxation of χ' and χ'' is measured as a function of t for a period t_{s2} at T_2. The system was again heated back to T_1 (the positive T-shift). These processes are repeated subsequently. Just after every negative T-shift, both χ' and χ'' do not lie on the reference curves of χ' and χ'' at T_2 obtained when the system is quenched to T_2 directly from 10 K at t = 0.

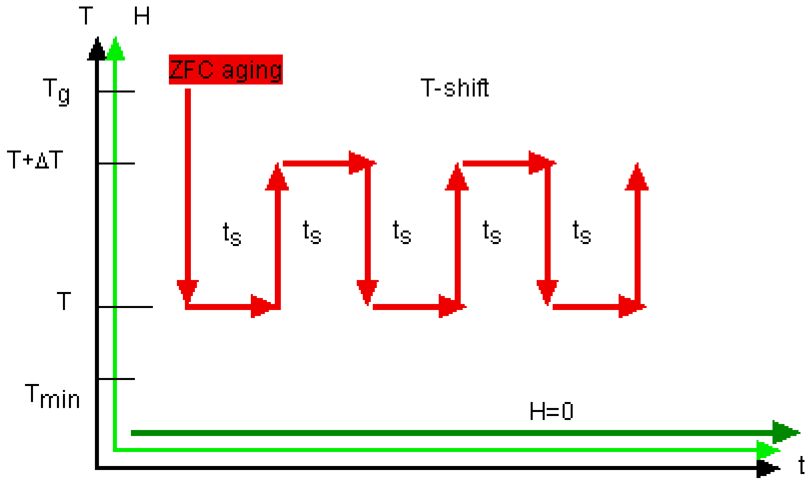

Diagram 20

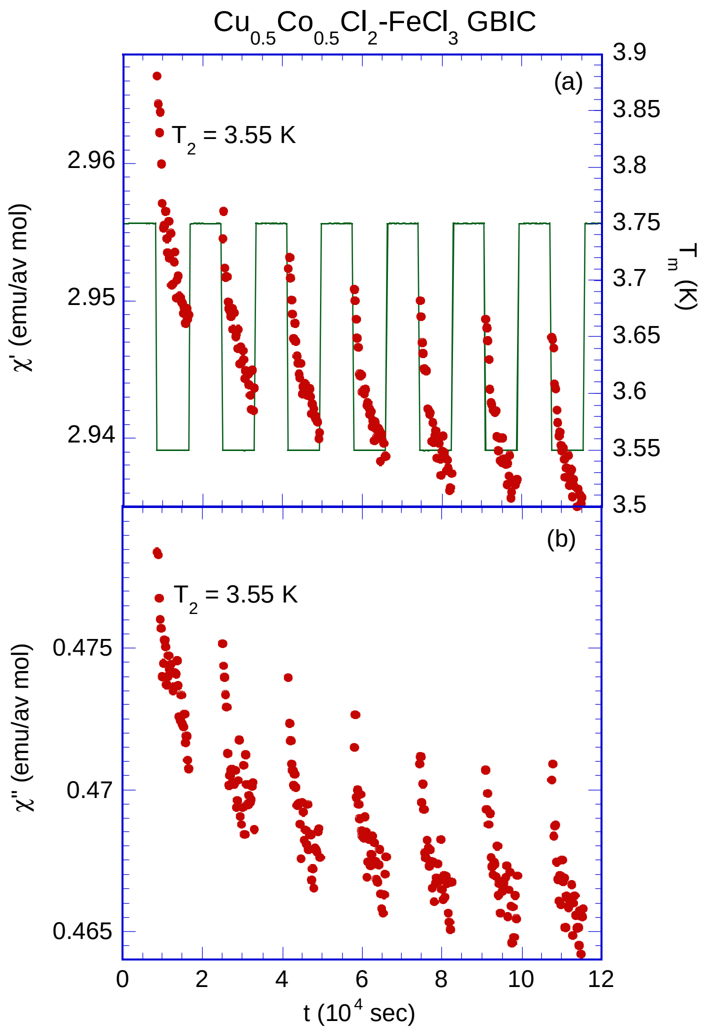

Figure L Relaxation of $\chi'(\omega,t)$ and $\chi''(\omega,t)$ at T_2 = 3.55 K during a temperature cycle between T_1 = 3.75 K and T_2 = 3.55 K. The change of T with t is also shown. t = 0 is a time just after the standard ZFC aging protocol: quenching of the system from 50 to 3.75 K at H = 0 [Ref. 2].

D. The rejuvenation and memory effects in $\chi'(\omega,t,H)$ and $\chi''(\omega,t,H)$ under the *H* shift

After the ZFC aging protocol (quenching from a high temperature above T_g to a temperature *T*), χ' and χ'' at $H = 0$ are measured for a period t_{s1}, where the origin of *t* ($t = 0$) is a time when *T* becomes T_m. The field is changed from 0 to *H* at $t = t_{s1}$, After this *H*-shift, χ' and χ'' were measured for a period t_{s2}. Subsequently, the field is turned off from *H* to 0 and the measurements were carried out at $H = 0$ for a period t_{s1}. This process is repeated.

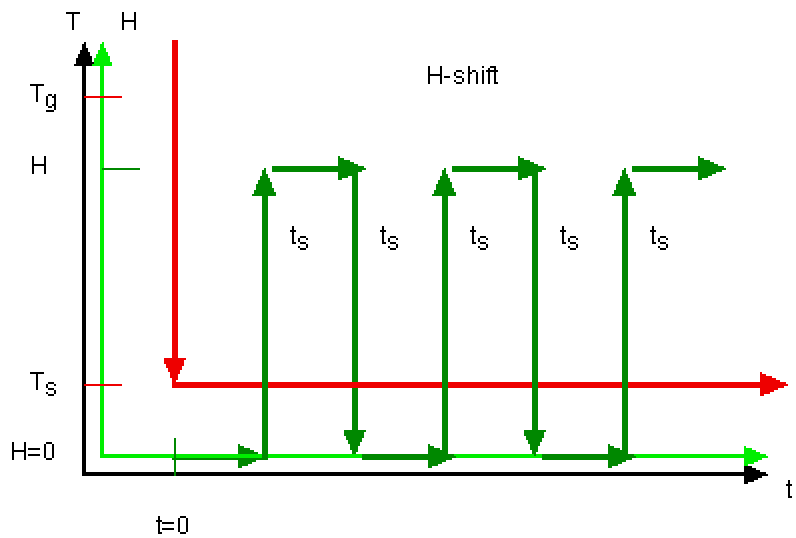

Diagram 21

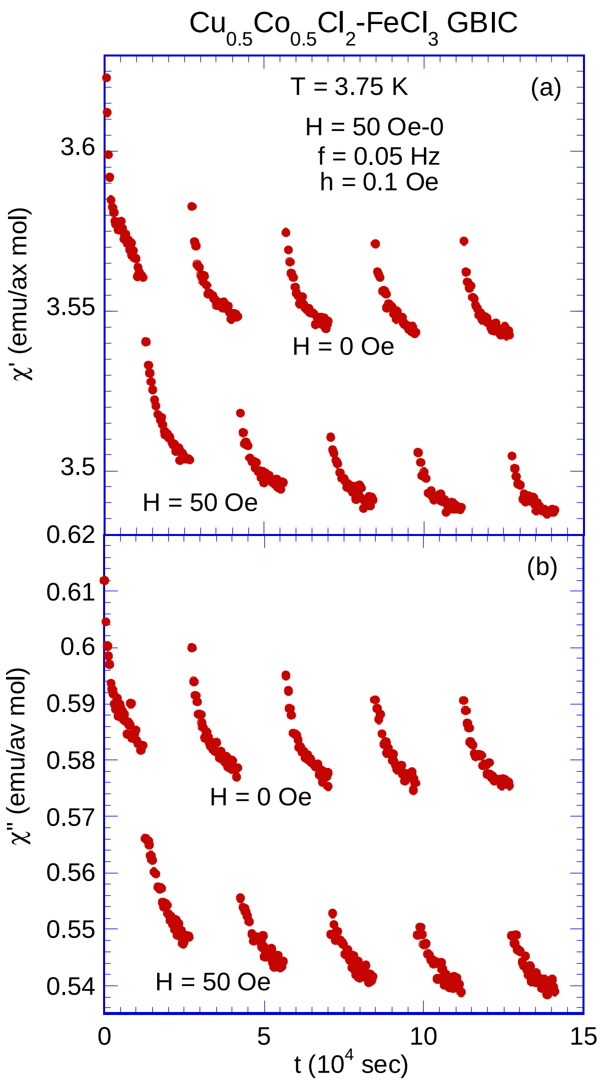

Figure M Relaxation of $\chi'(\omega,t)$ and $\chi''(\omega,t)$ during a magnetic-field cycle between H = 0 and 50 Oe. T = 3.75 K. t = 0 is a time just after the standard ZFC aging protocol: quenching of the system from 50 to 3.75 K at H = 0 [Ref. 2].

III. Fundamental Properties

A. π/2 rule

Now we consider the t dependence of S_{ZFC}(t) when t_w is very short. We note that $S_{ZFC}(t)$ is related to the absorption $\chi''(\omega)$ of the AC magnetic susceptibility by

$$S_{ZFC}(t) = d\chi_{ZFC}(t)/d\ln t = (2/\pi)\chi''(\omega), \qquad (9)$$

where $t = 2\pi/\omega = 1/f$, $\omega(=2\pi f)$ and f is the angular frequency and frequency of the AC magnetic susceptibility, respectively. In the AC measurement one is able to explore the beginning of the aging regime, a so-called quasistationaly regime which represents a much smaller time window than in the ZFC measurement.

B. Stretched exponential relaxation

Here we present a simple review on the stretched exponential relaxation of the zero-field cooled magnetization of SG phase after the ZFC aging protocol. Theoretically and experimentally it has been accepted that the time variation of $\chi_{ZFC}(t)$ may be described by a product of a power-law form and a stretched exponential function

$$\chi_{ZFC}(t) = M_{ZFC}(t)/H = \chi_0 - At^{-m} \exp[-(t/\tau)^{1-n}], \qquad (10)$$

where the exponent m may be positive and is very close to zero, n is between 0 and 1, τ is a characteristic relaxation time, and χ_0 and A are constants. In general, these parameters are dependent on t_w. This form of $\chi_{ZFC}(t)$ incorporates both the nonequilibrium aging effect through the stretched exponential factor $[\exp[-(t/\tau)^{1-n}]$ in the crossover region ($t \approx t_w$ and $t>t_w$) between the quasi equilibrium state and nonequilibrium state, and an equilibrium relaxation response at $t<<t_w$ through a pure power-law relaxation (t^{-m}). A stretched exponential only enters the functional form of the SG relaxation to characterize the aging phenomenon and is not associated with the equilibrium relaxation. Note that Ogielski fits his data by a stretched exponential multiplied by a power function. Similar law is also observed in the relaxation rate of various systems. The relaxation for $T>T_g$ is considerably slower. For $0.6<T/T_g<1$, Ogielski fits it by a power law with a different temperature dependence of exponent m. When $t<<\tau$, $\chi_{ZFC}(t)$ is well described by a power law form given by At^{-m}. However, in the regime of $t \approx \tau$, the stretched exponential relaxation is a very good approximation in spite of finite m that is very small.

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