Slow Dynamics for Spin-Glass-Like Phase of a Ferromagnetic Fine Particle System

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(Received 7 December 1998)

Effects of temperature changes on aging phenomena are studied for the spin-glass-like phase of a ferromagnetic fine particle system. The results obtained during the short experimental periods are, in appearance, asymmetric about temperature changes. However, the extension of the periods shows that the effects of temperature changes are symmetric. These results can be explained by the droplet model well. [S0031-9007(99)09194-2]

PACS numbers: 75.50.Lk, 75.50.Tt

Ferromagnetic fine particle systems have been intensively investigated as a disordered system with random anisotropy and competing dipolar interactions [1,2]. There has been a controversy of whether there is a phase similar to the spin-glass phase or not. Recently, some results indicating the existence of a phase transition have been reported by using frozen magnetic fluids containing uniform particles [3–5]. These papers have shown a critical slowing down of the relaxation and a divergence of the nonlinear susceptibility at finite temperature T_g [3,4]. Below T_g, a plateau of temperature dependence has been observed for equilibrium susceptibility [5]. Therefore, it is now widely accepted that there is a cooperative freezing at T_g as seen in spin glass. However, little attention has been given to the nature of the phase observed for ferromagnetic fine particle systems below T_g.

For the spin-glass phase, the correct understanding of its nature is still open to question. Two models have dominated the discussion. One is the droplet model [6–8], and the other is the hierarchical model [9]. Studies on the effects of the temperature changes for the aging phenomena have been considered to be a key to the examination of these models in spin glass [7–9]. In the droplet model, the system has two pure equilibrium spin configurations which are related by global spin reversal. In the configuration, the relative orientations of spins at distances longer than a characteristic length (overlap length, $L_{\Delta T}$) are quite sensitive to small temperature changes. Such sensitivity has been called "chaos" [10]. For this reason, we should observe the domain growth restarts from the size $L_{\Delta T}$ not only after cooling but also after heating, when the grown domain is larger than $L_{\Delta T}$. On the other hand, in the hierarchical model, a multivalley structure is hierarchically organized on the free energy surface, and the valleys merge with increasing temperature. For this reason, the hierarchical model without the concept of the chaos predicts that the aging is fully initialized only by raising the temperature, while the results of the aging are held during temporary cooling. Here, we must mention that some researchers have striven to construct a new model based on the idea that the two models are mutually supplemented [11], because both the models described above cannot explain some features observed by the recent experiments [11,12]. Therefore, although much still remains to be done, it is clear that the analyses of the effects of temperature changes have thrown new light on the understanding of the nature of the spin-glass phase.

In ferromagnetic fine particle systems, many researchers have observed a slowing down of the response to the static magnetic field with time spent at a constant temperature [2,4]. However, there is no experiment for the aging phenomena with temperature changes. In this study, to clarify the nature of the phase observed in fine particle systems, we measured the relaxation of the ac susceptibility with temperature changes. The results are compared with the hierarchical model and the droplet model described above.

The sample is the frozen iron-nitride magnetic fluid that is named d1 in the previous papers [5]. Electron microscopy shows that the magnetic fluid contains spherical particles whose diameters d are 6 ± 1 nm. The volume fraction of the particles is about 2.5%. From this value, the particle number density is estimated at about $1/(2.8d)^3$. By the absorbed layer of surfactant molecules, the particles are dispersed in the carrier liquid of kerosene without agglomeration. For this reason, ferromagnetic particles in the sample, which is solidified in the zero field, are considered to be randomly fixed in solidified kerosene. In the previous paper [5], we have reported that the sample d1 has a spin-glass-like phase below $T_g \approx 70$ K. We note here that the magnetic moment fluctuates between directions that are parallel and antiparallel to the easy axis of each particle, as seen in Ising systems, since the anisotropy energy is estimated at 0.1 eV that is much higher than the thermal energy below T_g.

ac susceptibility $\chi' + i\chi''$ was measured by using a SQUID magnetometer in a magnetic shield of Permalloy, where the residual field was reduced well below 0.03 Oe. The procedure involving the temporary temperature changes is shown in the inset of Fig. 1(a). After the sample was quenched from 150 K ($>T_g$) to $T_m = 47$ K ($\approx 0.7T_g$) in the zero field, we measured ac susceptibility as a function of time t. The applied field has a frequency of 800 mHz and an amplitude of 1.0 Oe. After period t_1 spent at T_m, a temporary temperature change to $T_m + \Delta T$ was performed during a period t_2. Since the kerosene is an insulator with low thermal conductivity, the real temperature of fine particles has a delay of about 1 min from the nominal temperature measured by a thermometer near the sample. For this reason, we used the periods longer than 1×10^3 s (1 ks).

The solid curves in Fig. 1 show the time variation of the out-of-phase component χ'' without a temporary temperature change at $T_m = 47$ K. It is found that χ'' decays with t after the quench. In other words, there is a relaxation that varies the response of the ac field, as discussed for spin glass [8,9]. The solid circles in Fig. 1(a) show the relaxation curve of χ'' with a temporary heating during $t_2 = 1.2$ ks for $\Delta T = +7$ K ($\approx +0.1T_g$). We find that after temporary heating χ'' starts from a level much higher than that reached before heating. In some analyses for spin glass [8,9], such behavior is considered to be caused by the initialization of the relaxation due to temporary heating. On the other hand, Fig. 1(b) shows the relaxation curve of χ'' with a temporary cooling during $t_2 = 15.7$ ks for $\Delta T = -7$ K ($\approx -0.1T_g$). It should be noted that χ'' comes back to the level it reached before cooling, when the temperature returns to T_m after temporary cooling. The open circles in Fig. 1(b) show the same χ'' against $t - t_2$ which is the total time spent at $T_m = 47$ K. It is found that the relaxation curve during period t_3 is on the continuation of the curve during t_1. In other words, the results of the relaxation before temporary cooling are retrieved when the temperature returns. Therefore, the experimental results observed during the short periods are consistent with the predictions of the hierarchical model discussed for spin glass [9].

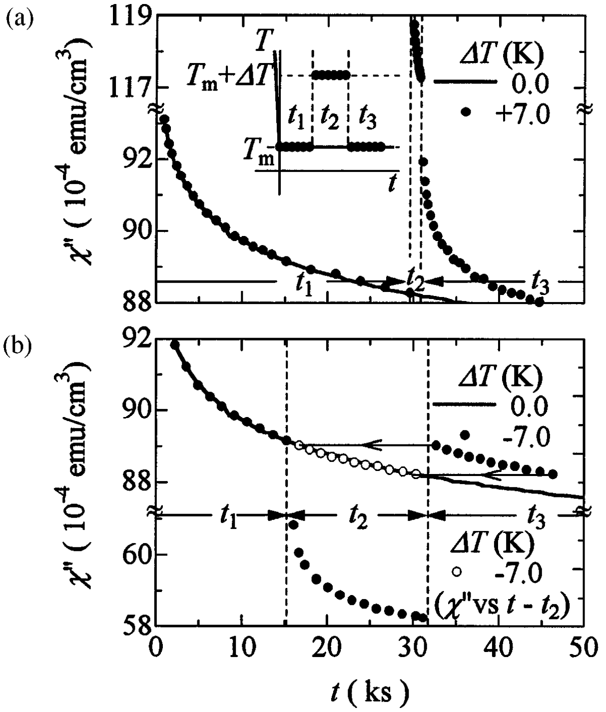

FIG. 1. Relaxation curves of the out-of-phase component χ'' of the ac susceptibility (a) with temporary heating at $T_m + \Delta T = 47 + 7$ K and (b) with temporary cooling at $47 - 7$ K. The open circles are plotted against $t - t_2$. The solid lines show the isothermal curves at $T_m = 47$ K. The procedure is shown in the inset.

Recently, Vincent *et al.* have reported some new results for a typical spin glass $CdCr_{1.7}In_{0.3}S_4$ that has been discussed by the hierarchical model [12]. A novel relaxation of χ'' was observed before the retrieval after temporary cooling, and it was termed "a transient faster relaxation." Because such faster relaxations after temporary cooling have been already observed for a Cu-Mn spin glass and discussed by the droplet model [8], this phenomenon has attracted considerable attention. It is interesting to check for the existence of the transient relaxation in this system. Figure 2 shows the relaxation of χ'' when the period of temporary cooling for $\Delta T = -7$ K is extended to 305.9 ks. We find a faster relaxation at the beginning of t_3. It starts from a level higher than that reached before cooling. However, except for a singular period at the beginning of t_3, the curve during t_3 becomes a continuation of the curve during t_1 on the plot for the total time spent at $T_m = 47$ K, although an effective extra time $t_{\mathrm{eff}} = 2 \pm 1$ ks has to be added to the total time $t - t_2$. For these reasons, we shall call the faster relaxation in the singular period "transient relaxation" in this Letter. The observed result also shows that the relaxation at $T_m - \Delta T = 47 - 7$ K during $t_2 = 305.9$ ks has an effect that is equivalent to the effect of the relaxation at $T_m = 47$ K during $t_{\mathrm{eff}} = 2 \pm 1$ ks. In other words, the equilibration of the magnetic moment configuration during the aging at $T_m - \Delta T$ is similar to that at T_m. However, it is clear that the equilibration at $T_m - \Delta T$ is not identical

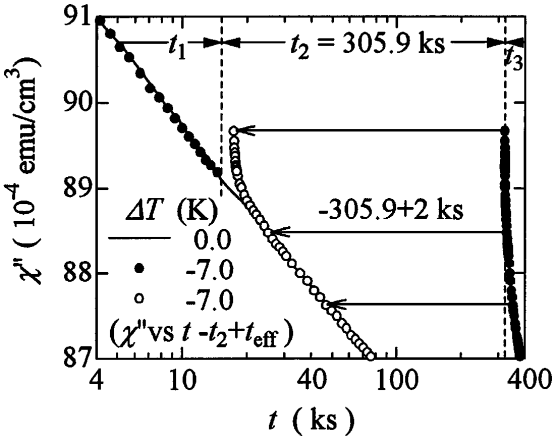

FIG. 2. Relaxation curve of χ'' with a long period of t_2 of temporary cooling at $T_m + \Delta T = 47 - 7$ K. The open circles are plotted against $t - t_2 + t_{\mathrm{eff}}, t_{\mathrm{eff}} = 2 \pm 1$ ks. The solid line shows the isothermal curve at $T_m = 47$ K.

with that at T_m, because a transient relaxation exists before the relaxation curve returns to the isothermal curve.

These results drive us to the question of whether the faster relaxation observed after temporary heating for $\Delta T = +7$ K arises from the full initialization of the relaxation before it. Figure 3 shows the relaxation curve measured for a long time after temporary heating during the period t_2 of 1.2 ks. We should notice that the curve after temporary heating crosses the isothermal curve at $T = 47$ K, and that it approaches the isothermal curve again from under it with time. It is obvious that the curve after temporary heating cannot be lower than the isothermal curve, if the faster relaxation after temporary heating restarts from the state that is the same with the initial state after the first quench. The inset of Fig. 3 shows the relaxation curves against the time spent after temporary heating, $t - t_1 - t_2$. We can confirm that the curves of $\chi''(t - t_1 - t_2)$ are different from the isothermal curve of $\chi''(t)$ after the quench. We also find that the curves depend on t_1. These results indicate that the faster relaxation is not caused by the full initialization of the relaxation during t_1. Here, in order to clarify the reason of the crossing of the curves in Fig. 3, we shall consider the effective time as discussed above. If the curve of $\chi''(t)$ during t_3 in Fig. 3 shifts 17 ks toward a longer time, it is just on the isothermal curve of $\chi''(t)$ in the time range after $t \sim 70$ ks. In other words, the relaxation during temporary heating affects the relaxation at T_m as $t_{\text{eff}} = 1.2 + 17$ ks, when the time passes sufficiently. Therefore, the faster relaxations after temporary heating are also transient relaxations.

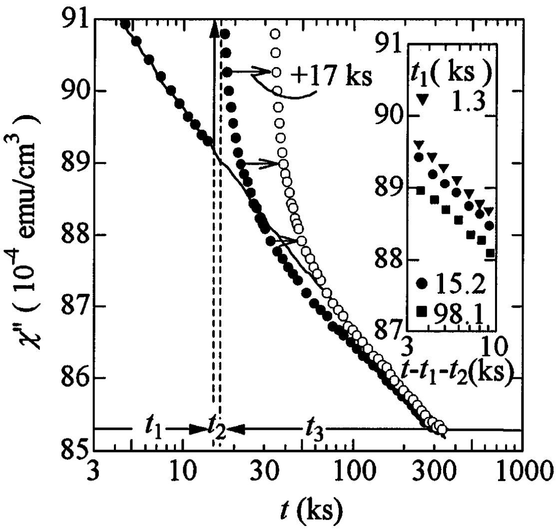

FIG. 3. Relaxation curve of χ'' with temporary heating at $T_m + \Delta T = 47 + 7$ K during $t_2 = 1.2$ ks. It is shown in a wide time range. The open circles are plotted against $t + 17$ ks. The solid line shows the isothermal curve at $T_m = 47$ K. The inset shows the relaxation curves against the time spent after temporary heating, $t - t_1 - t_2$, during periods t_3. The different symbols show the curves when the system was aged for different periods t_1 before temporary heating during $t_2 = 1.2$ ks.

As shown above, the equilibrations of the magnetic moment configuration at different temperatures are not the same, but similar to each other. This qualitative property is symmetrical about temperature changes, although t_{eff}/t_2 monotonously increases with ΔT from $\Delta T \approx -0.1T_g$ to $\Delta T \approx +0.1T_g$. In order to explain such results by using the asymmetric variation of the free energy surface with temperature changes, we must introduce the concept of the chaos, which has symmetric effects about differences of temperature, into the hierarchical model [12]. Although this seems valid, its complication may confuse us. Therefore, at the present stage, we will leave the phase space picture and discuss the observed results in real space.

In the droplet model [6–8], the grown domains at a temperature continue to grow at another temperature, if they are smaller than $L_{\Delta T}$ when the temperature changes. On the other hand, if they are larger than $L_{\Delta T}$, the domain growth restarts from the state with domains of size $L_{\Delta T}$. Because the grown domains must have various sizes, these two phenomena coexist for various ΔT in a wide time range. It is reasonable to interpret the two phenomena as the origins of the two observed features of this system: the effective time and the transient relaxation.

Here, we note the reason why no transient relaxation can be observed after the short temporary cooling. A possible interpretation by the droplet model is that the restarted domain growth at $T_m - \Delta T$ proceeds little during it and the domains cannot become larger than $L_{\Delta T}$, because the thermal activation process becomes slower when the temperature is lowered. Now, we notice that, if the droplet model can be applied, the transient relaxation should be enhanced with the time spent before a temperature change. The reason is the fraction of grown domains larger than $L_{\Delta T}$ increases with the time. For this reason, we are interested in the variation of the transient relaxation with the length of periods before temperature changes. Because the relaxation curves during t_3 depend on t_1 as shown above, the domains that grew during t_1 at T_m remain when the temperature returns to T_m. Therefore, to compare the equilibrations at different temperatures exactly, the temperature change should be only once. The procedure is shown in the inset of Fig. 4. After the sample was quenched to $T_m + \Delta T$, we measured ac susceptibility by applying the same ac field as stated above. After various periods t_1', the temperature was shifted to T_m. Since too long of a period is required for the observation of the transient relaxation for $|\Delta T| = 7$ K, we decreased $|\Delta T|$ to 2 K $\approx 0.03T_g$.

Figure 4 shows the relaxation curves of χ'' during t_1' at $T_m + \Delta T = 49$ K, and during t_2' at $T_m = 47$ K. It is found that the relaxation curves are just on the isothermal curve at 47 K after the transient relaxations, if the effective times t_{eff} at 47 K corresponding to t_1' at 49 K are assumed to be about 3 times the length in t_1'.

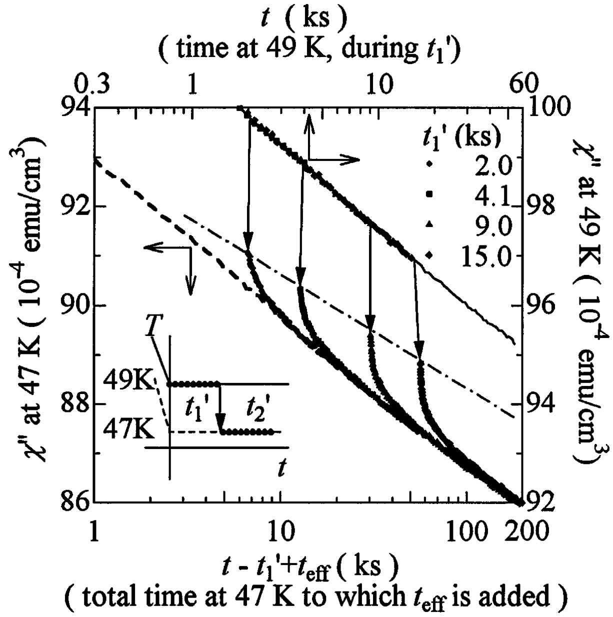

FIG. 4. Relaxation curves of χ'' with temperature shifts from 49 to 47 K. The relaxation curves at 47 K are plotted against the total time at 47 K that includes effective extra time $t_{\rm eff}$. The solid line and broken line show the isothermal curves at 49 K and 47 K, respectively. The procedure is shown in the inset.

In this figure, we should notice that the magnitude of the transient relaxation increases with t_1'. The period of the transient relaxation also increases intensively with t_1', although it is not conspicuous because the time is plotted logarithmically. These results are consistent with the prediction by the droplet model. On the other hand, we should not overlook the fact that the ratio $t_{\rm eff}/t_1'$ does not decrease with t_1'. The following explanation can be considered. The droplet model predicts the larger $L_{\Delta T}$ for the smaller $|\Delta T|$ [6–8]. If $|\Delta T|$ of $0.03 T_g$ is assumed to be small, the fraction of the domains which becomes larger than $L_{\Delta T}$ should be small in comparison with the rest. On this condition, the variation of $t_{\rm eff}/t_1'$ is not appreciable, because the similarity that appears as $t_{\rm eff}$ is held by the rest.

The ac susceptibility was measured for a frozen iron-nitride magnetic fluid. For the first time, the effects of the temperature changes on the aging phenomena are discussed for ferromagnetic fine particles with dipolar interactions and random anisotropy. We observe transient relaxations after the temperature changes from $T_m \pm |\Delta T|$ to T_m. After those, the relaxation curves are on the isothermal relaxation curve at T_m, if effective times at T_m are assumed for the periods at $T_m \pm |\Delta T|$. The droplet model for spin glass can well explain these symmetric results about temperature change.

We acknowledge helpful discussions and technical assistance by Dr. T. J. Sato and Dr. T. Taniyama. We wish to thank Dr. T. Jonsson, Professor P. Nordblad, Professor P. Svedlindh, Dr. T. Komori, and Professor H. Takayama for valuable advice.

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