A reversion of magnetization decay in spin glasses

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Abstract. Reversion of thermoremanent magnetization decay is observed in a canonical spin glass; that is, the system is remagnetized despite the absence of a magnetic field. This unexpected phenomenon occurs only when the system is heated or cooled back to the temperature $T_0$ at which it was previously subjected to a magnetic field for a significant length of time. Therefore, this reversion can be interpreted as a restoration of the spin state existing before at $T_0$. This new information will be a key to resolving a long-standing controversy regarding the nature of spin glasses.

Spin glasses have been theoretically studied as a prototype for other glassy systems. However, little is known about their nature [1]–[12]. Two contrasting pictures are well known. The first is the mean-field picture [1], according to which numerous stable spin configurations coexist at a given temperature. The second is the droplet picture [3, 4], according to which only one equilibrium spin configuration (and its spin-reversed counterpart) exists at each temperature. However, this configuration is extremely sensitive to temperature. Although these predicted equilibrium states are essentially different, experimental verification of these predictions is impractical because the process of relaxation is extremely slow [13]–[21]. For instance, the out-of-phase component of ac susceptibility $\chi''$, the energy dissipation, continues to gradually decrease throughout isothermal experiments over a period of hours [15, 16].

Intriguingly, such slow relaxations, also known as aging phenomena, are remarkably affected by thermal perturbations; that is, if the system is temporarily cooled subsequent to isothermal aging, $\chi''$ becomes larger. In aging terminology, a more dissipative system is younger; thus, this effect is termed rejuvenation [7, 20]. On the other hand, after temporary cooling, $\chi''$ easily returns to the small value attained before cooling, as if the stability of the aged system was retrieved. This is recognized as the memory effect [7, 16]. Because the coexistence of rejuvenation and the memory effect is extraordinary, these findings were expected to provide

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Figure 1. Rough sketches of the effects of relatively large thermal perturbations on the two major scenarios regarding the nature of spin glasses. (a) The evolution of domains in actual space in the ghost domain scenario. The colours represent the projections of the spin configuration onto the equilibrium spin configuration \( \{S_i(T, J)\} \) and its global spin inversion \( \{-S_i(T, J)\} \) at temperatures \( T_0 \) and \( T_0 \pm \Delta T \). In this panel, the temperature difference \( |\Delta T| \) is assumed to be so large that the so-called overlap length \( L_{\Delta T} \) is disregarded. As a result, \( \{S_i(T_0 \pm \Delta T, J)\} \) is completely different from \( \{S_i(T_0, J)\} \). (b) The landscape of the free-energy surface in the spin configuration space for the hierarchical scenario, where the thick blue and red arrows denote evolution during aging with temporary cooling and temporary heating, respectively. The upper panel shows the thermal histories.

In the ghost domain scenario of the droplet picture \([8]\), an equilibrium state with a set of bonds \( J_{ij} \) at temperature \( T \) is given by its stable spin configuration \( \{S_i(T, J)\} \), where \( S_i \) denotes the spin at site \( i \). The other possible stable configuration in the same environment \( (T, J_{ij}) \) is the only global spin inversion \( \{-S_i(T, J)\} \). Because these configurations are paired like ferromagnets, isothermal aging at \( T_0 \) simply proceeds with the gradual growth of domains corresponding to either \( \{S_i(T_0, J)\} \) or \( \{-S_i(T_0, J)\} \), as shown in subpanels (a)–(c) in figure 1(a). Thus, the system becomes less dissipative with the growth of domains. However, during the growth at \( T_0 \), the spin

\[ S_i = S_i(T_0, J) \quad \text{and its counterpart} \quad -S_i(T_0, J) \]

\[ S_i = -S_i(T_0, J) \quad \text{and} \quad -S_i(T_0, J) \]

\[ \text{Ghost domain scenario} \]

\[ \text{Only one stable spin configuration} \quad \{S_i(T_0, J)\} \quad \text{and its counterpart} \quad -S_i(T_0, J) \]

\[ \text{No correlation between} \quad \{S_i(T_0, J)\} \quad \text{and} \quad \{-S_i(T_0, J)\} \]

\[ \text{Hierarchical scenario} \]

\[ \text{Numerous stable spin configurations at each} \quad T \]

\[ \text{Hierarchically organized multi-valley structure} \]
configuration \{S_i(t)\} is completely random with respect to the spin configuration \{S_i(T_0 - \Delta T, J)\} that is stable at a sufficiently different temperature \(T_0 - \Delta T\) (see subpanels \((\zeta)\)–\((\theta)\)), because \(S_i(T_0 - \Delta T, J)\) is entirely uncorrelated with \(S_i(T_0, J)\) on the condition that \(\Delta T\) is large enough for the overlap length \(L_\Delta T\) to be just 1 in the unit lattice. Therefore, the domains corresponding to \(S_i(T_0 - \Delta T, J)\) (or \(-S_i(T_0 - \Delta T, J)\)) nucleate in every part and start to grow independent of the original domain structures, when the temperature is lowered to \(T_0 - \Delta T\) halfway through aging at \(T_0\) (see subpanels \((\theta)\)–\((i)\)). In this case, the system again becomes highly dissipative as it is initially quenched from a paramagnetic state. On the other hand, if \(\{S_i(t)\}\) during the temporary cooling is projected onto \(S_i(T_0, J)\), we can find ghost-like noisy patterns with the same spatial structure as the original one, as illustrated in subpanel \((\delta)\). The reason is that breakdowns of the destabilized original domains are as slow as the domain growth. Thus, when the temperature returns to \(T_0\), \(\{S_i(t)\}\) is not random with respect to \(\{S_i(T_0, J)\}\) (and \(-S_i(T_0, J)\)) but rather resembles one of them within each ‘ghost domain’. As a result, the original domain structure is easily restored after reheating and the system regains the previous stability, as shown in subpanel \((\varepsilon)\).

It is worth noting that such a restoration based on the noisy ghost domains would be almost improbable if the other numerous spin configurations were also stable at \(T_0\). We may visually interpret that the last Z-shaped pattern in \((\varepsilon)\) cannot be recovered if the magenta, purple and violet domains exist at \(T_0\) in addition to the red and blue ones. Thus, the ghost domain scenario is consistent with the extraordinary features of the observed aging phenomena.

On the other hand, in the mean-field picture, aging has been discussed as an exploration of the spin-configuration space with numerous valleys on the complex free-energy surface, the lowest of which represent the equilibrium pure states. A hierarchical scenario \([2, 15]\) has been inspired by the organization of equilibrium pure states in the Parisi solution \([1]\). Briefly, this scenario assumes that all the valleys are hierarchically subdivided into smaller valleys with lowering the temperature, as sketched in figure 1(b). Consequently, the start of exploration of the subdivided valleys makes the system dissipative on cooling, whereas a merger of these subdivided valleys on reheating leads to a return to the original valley in the spin-configuration space and causes the memory effect. Subsequently, these remarkable features were also analytically reproduced within the mean-field picture \([6]\). As discussed here, the outcome obtained by temporary cooling can be explained by both the ghost domain and hierarchical scenarios.

At this stage, we should note that both rejuvenation and restoration can occur after temporary heating as well as after temporary cooling in the ghost domain scenario. On the other hand, in the hierarchical scenario, the system becomes dissipative at the beginning of the temporary heating period, at which the exploration of the spin-configuration space restarts in a fresh landscape created by a merger of the valleys, as sketched in figure 1(b). When the temperature is lowered again at the end of the heating period, the system is left in one of the redivided valleys in the region explored during temporary heating. In this case, a spontaneous return to the original valley is apparently improbable in the spin-configuration space, because there are many redivided valleys other than the original. Therefore, a crucial difference between the two scenarios is the restoration of the original spin configuration after temporary heating. In addition to the conventional approach using the system age estimated from its dissipativity at that time, it is necessary to adopt a new experimental approach to ascertain whether the spin configuration is actually restored to the original. This is because the recovery of dissipation capability \(\chi''\) to the extent attained before thermal perturbations can be attributed not only to such a restoration but also to a simple reactivation of components that remained
Figure 2. Typical variations in $M_{\text{TRM}}$ for the Cu$_{97}$Mn$_3$ alloy. After quenching in a magnetic field, the system was maintained at $T_0 = 18.0$ K for an isothermal waiting period $t_w$ of 50 ks; then, the magnetic field was removed at $T_1 = T_0$ and $t = 0$. Halfway through the decay, the sample was temporarily cooled to $T_1 - |\Delta T|$, as shown in the inset. The solid curve represents the isothermal reference curve; shaded areas denote cooling periods. The open circles for $\Delta T = -0.1$ K indicate that the relaxation curve shifted leftwards by 3.4 ks.

dormant (frozen) during the perturbations. On the other hand, magnetization $M = \Sigma S_i$ reflects the global evolution of the spin configuration $\{S_i(t)\}$. Therefore, $M$ would reattain its original value in the case of restoration, whereas the relaxation of $M$ would simply restart from the value in the dormant state in the case of reactivation. For this reason, we carefully studied the relaxation of $M$ for a canonical spin glass made of a CuMn alloy.

A CuMn alloy was prepared with an Mn content of 3.4 at.% to avoid undesirable phenomena such as the ordered phase of Cu$_5$Mn$_2$ [22], chemical short-range order [23] or mictomagnetism (cluster glass) [24] that are observed for concentrations higher than 10 at.%. In this alloy, typical spin-glass behaviour was observed below the transition temperature $T_{SG} = 20.6$ K [25]. First, the sample was quenched from 40 K (paramagnetic state) to the desired temperature $T_0$ in a magnetic field of strength $H = 79.6$ A m$^{-1}$. The sample was then maintained at $T_0$ for a period $t_w$. Then, the field was removed at $T_1$ and the measurement of thermoremanent magnetization $M_{\text{TRM}}$ started ($t = 0$). Finally, the samples were temporarily cooled/heated in the period from $t_1 > 0$ to $t_2 > t_1$.

Figure 2 shows typical variations in thermoremanent magnetization $M_{\text{TRM}}$. Under isothermal conditions, $M_{\text{TRM}}$ gradually decreases after the removal of the magnetic field at $T_1$ (= $T_0 = 18.0$ K). This decrease slows further when the temperature is temporarily lowered to $T_1 - |\Delta T| = 17.9$ K halfway through the decay at $t_1 = 1$ ks. On subsequent reheating at $t_2 = 10$ ks, the $M_{\text{TRM}}$ decrease is accelerated. When the relaxation curve obtained after reheating is shifted leftwards by 3.4 ks, it can be superimposed on the isothermal aging curve (represented by open red circles on the solid black curve); that is, a period of 9 ks at $T_1 - |\Delta T|$ is equivalent to a period of 5.6 ks at $T_1$. The only difference between the aging phenomena at these temperatures is the excitation rate. In other words, $\Delta T = -0.1$ K is too small for
observing the rejuvenation and memory effects. Such phenomena are termed accumulative aging [20].

On the contrary, non-intuitive features are seen on further cooling (e.g. \( \Delta T = -0.2, -0.4, \) and \(-1.8 \) K). The first unanticipated phenomenon is a temporary acceleration of the decrease in \( M_{TRM} \) at \( t_1 \) despite lowered temperatures. Since a significant enhancement in the decay rate is equivalent to a rise in \( \chi'' \), this phenomenon can be interpreted as rejuvenation, which has been mentioned above. Even more surprising is the fact that, despite the absence of a magnetic field, \( M_{TRM} \) increases steeply and reattains almost its original value immediately after the subsequent reheating at \( t_2 \). Paramagnetic impurities, even if present, would make no contribution to \( M_{TRM} \) at zero magnetic field. Therefore, this steep increase in \( M_{TRM} \) is supposed to be an intrinsic property of the CuMn alloy.

Our concern here is whether or not similar increases in \( M_{TRM} \) occur when the system is temporarily heated, apart from overall decreases that are generally expected for \( M_{TRM} \) at zero magnetic field. Figure 3 shows the decay curves of \( M_{TRM} \) when the temperature is temporarily raised from \( T_1 = 18.0 \) K to \( T_1 + |\Delta T| = 18.2 \) K for periods from \( t_1 = 0.55 \) ks to \( t_2 = 0.75 \) ks. In the case of figure 3(a), the system was maintained at \( T_0 = 18.0 \) K for a long period before the removal of the magnetic field. In this case, the decay rate is found to have increased approximately 50-fold at the onset of temporary heating at \( t_1 \). This rapid acceleration may also be affected by rejuvenation, because it is difficult to explain the acceleration as a simple thermal activation resulting from a 1% increase in thermal energy. More importantly, an evident increase in \( M_{TRM} \) is also observed at the end of the heating period, as was observed at the end of the cooling period with \( |\Delta T| \geq 0.2 \) K in figure 2. Because the increase in \( M_{TRM} \) occurs not only on reheating but also on recooling, it cannot be attributed to any thermally reversible component.

In other words, the demagnetization process of the spin configuration is certainly reversed at the end of these thermal perturbations. Remarkably, a more magnetized configuration becomes more stable at zero magnetic field.

Note that reversion of magnetization decay does not always occur at the end of thermal perturbations, as shown in figure 3. In the case where the system is maintained at \( T_0 = 18.2 \) K prior to the removal of the magnetic field, we can find a temporary increase in \( M_{TRM} \) at the beginning of the heating period, in addition to an overall decrease in \( M_{TRM} \) at zero magnetic field (see figure 3(b)). Despite the same thermal perturbation as that shown in figure 3(a), the reversion occurs when the temperature is raised to \( T_0 = 18.2 \) K. On the other hand, even if the spin configuration is previously equilibrated in the magnetic field for a long period at \( T_0 = 17.8 \) K, no increase in \( M_{TRM} \) occurs at either the beginning or the end of the thermal perturbations between 18 and 18.2 K (see figure 3(c)). To sum up, the demagnetization process of the spin configuration in zero magnetic field is reversed only when the system is returned to the original temperature at which a magnetized configuration previously formed in a magnetic field. Hence, it is reasonable to say that the spin configuration preformed at a certain temperature regains its stability on return to that particular temperature. Consequently, the relaxation is reversed and the preformed configuration is restored spontaneously. As discussed at the beginning of this paper, the hierarchical scenario cannot be easily modified to reproduce the restoration observed after temporary heating.

Let us now carefully examine the phenomena. In figure 2, the decrease in \( M_{TRM} \) is approximately 30% of its initial value during the cooling period at \( \Delta T = -1.8 \) K. When the system is assumed to have been homogeneously magnetized in its initial state, it can be said that one-sixth of the spin configuration was rearranged during temporary cooling. In the
Figure 3. Variations in $M_{TRM}$ for the Cu$_{97}$Mn$_3$ alloy with temporary heating. After quenching in a magnetic field, the system was maintained at $T_0$ (=18.0, 18.2 or 17.8 K) for 250 ks; then, the field was removed at $T_1 = 18.0$ K and $t = 0$. Halfway through the decay, the sample was temporarily heated to $T_1 + |\Delta T| = 18.2$ K, as shown in the inset. The solid curves represent the reference curves without temporary heating; shaded areas denote heating periods. Unusual increases at zero magnetic field (indicated by arrows) are evident when the system is heated or cooled back to temperature $T_0$, while overall decreases, which are generally expected for $M_{TRM}$ at zero magnetic field, are found in the other parts of the relaxation curves.

spin-configuration space, the transfer distance during cooling, $d_{12} = (4N)^{-1} \cdot \Sigma [S_i(t_1) - S_i(t_2)]^2$, is given as $q_{EA}/6$, where $q_{EA}$ is the self-overlap or the so-called Edwards–Anderson order parameter. Similarly, $d_{12}$ during heating, which is shown in figure 3(a), is roughly estimated to be $q_{EA}/12$. Although the accuracy of these estimates may be disputable, it is certain that $d_{12}$ is considerably longer.

The question then arises as to how the system can explore long distances in the spin-configuration space with a multi-valley structure during thermal perturbations and how it can spontaneously return to the original valley after such long-distance exploration. One of the
possible explanations for the former phenomenon is that the spin configuration \( \{ S_i(t_1) \} \), which is relatively stable at \( T_0 \), is destabilized at \( T_0 \pm \Delta T \). Consequently, the system is released from the original valley. Then, it promptly explores the region with a typical distance \( d_{12} \) in the spin-configuration space. The required condition is that the free-energy surface at \( T_0 \pm \Delta T \) has no deeper valleys in the region around the position where the original valley existed before at \( T_0 \). In other words, any stable spin configurations at \( T_0 \pm \Delta T \) are fairly different from that preformed at \( T_0 \); the extent of the difference is indicated by \( d_{12} \). Next, we emphasize the fact that the spontaneous return of the system to the original valley after the perturbations would have been impossible if the system had fallen into the basin of attraction of another deep valley at \( T_0 \) on the return path with a distance \( d_{12} \). In brief, the overall free-energy landscape at \( T_0 \) has a funnel-like structure in the explored region with a typical distance \( d_{12} \). Thus, the information obtained on the free-energy surface seems to be consistent with that indicated by the droplet picture; that is, only a single pair of spin configurations is stable at each temperature, while another pair replaces it when the temperature changes.

However, in addition to the two major scenarios, various others have also been added to the long-standing controversy regarding the nature of spin glasses. Typically, their basis is the conventional view that glassy states represent a kind of ‘frozen’ state. For example, successive freezing has been assumed for components such as excitation \([9]\), fluctuation \([10]\) or the local field \([12]\) in some scenarios for spin glasses. If the original state of the component is preserved (frozen) during temporary cooling, its reactivation on reheating is indeed observed as the memory effect concerning the dissipativity. Similarly, the memory effect would also be seen even after temporary heating if the heating period is short enough for the original state to be preserved. It is worth noting that this kind of memory effect is not accompanied by any restoration. Hence, these scenarios should at least be modified to eliminate such serious inconsistency with the present finding. As exemplified here, the information obtained in this study would be a key to validate a variety of scenarios proposed for spin glasses.

In summary, our observation of the restoration of the original spin configuration indicated by the reversion of magnetic relaxation cannot be easily explained by the hierarchical scenario, but it is possible in the alternative ghost domain scenario of the droplet picture. The overall structure of the free-energy surface in the currently explored region with significant size, estimated from analyses of intensive rearrangement and subsequent restoration, seems to be consistent with that indicated by the droplet picture, although further study is required to clarify whether or not this argument can be expanded into the entire spin-configuration space. Additionally, we should remember that the results obtained here apparently disagree with the conventional view that glassy systems are simply frozen. Thus, the present information not only enables us to verify the validity of the two major scenarios but also helps resolve the long-standing controversy regarding the nature of spin glasses. At this stage, we cannot ascertain whether the solution is the present ghost domain scenario, the further advanced ‘hierarchical droplet’ scenario \([16]\) or a completely different scenario.

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