Comparative review of aging properties in spin glasses and other disordered materials

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(Received August 10, 2018)

Aging phenomena have been studied in very different materials like polymers, supercooled liquids or disordered orientational crystals. We recall here the main features of aging in spin glasses, and use this example of magnetic systems as a guideline for the description of the others. A particular attention is put on the sensitivity of aging to the cooling rate and to temperature variations. This allows us to point out differences between temperature specific processes, yielding “rejuvenation and memory effects” as known in spin glasses, and domain growth processes, giving cumulative contributions at different temperatures. The relevance of wall depinning processes to rejuvenation and memory effects is discussed at the light of recent results on disordered ferromagnets.

KEYWORDS: aging, glassy dynamics, disordered systems, out-of-equilibrium phenomena

§1. Introduction

In materials science, aging is most commonly viewed in terms of a steady degradation of the properties of the considered material and is mostly a consequence of the action of some external factor (chemical reaction, irradiation effects, mechanical fatigue ...). However many materials and especially disordered materials experience a spontaneous long time evolution, without any external agent, whose direct consequences can be as important as for the usual chemical aging. This is well known in structural glasses and amorphous polymers and is referred to as physical aging.1) Much work has been done on these systems, since it was first recognised that their quenched state was unstable and evolved as a function of time (the aging time2,3)). As quoted by L.C.E. Struik1) “the phenomenon of physical aging is very important from a practical point of view. Several properties of glassy polymers, e.g. their small strain mechanical properties, undergo marked changes and strongly depend on the aging time. In the testing of such plastics, the aging time is just as important as other parameters such as temperature, stress level, humidity, etc. Furthermore, a knowledge of the aging behaviour of a material is indispensable to the prediction of its long-term behaviour from
short term tests". This sets the real problem in the field of out-of-equilibrium phenomena and has become a great challenge for modern statistical physics.

In recent years, much interest has been focused on the aging properties of various disordered systems: orientational glasses, supercooled liquids, charge density waves, and vortices in superconductors. Many efforts have also been devoted to disordered antiferromagnets in relation with the random field Ising model, or to disordered ferromagnets and the problem of domain growth in presence of randomly distributed pinning centers. Among all these examples, spin glasses are of special interest. They are the closest realisation of simple theoretical models of randomly interacting objects. There has been tremendous theoretical and experimental work on the slow and out-of-equilibrium dynamics of spin glasses. The question arises on how all these results compare with those of other types of glassy systems. Lately there has been quite an effort to unify experimental procedures and to generalise theoretical approaches. This makes a meaningful if not complete comparison possible. It is the aim of this paper to review some important features of aging phenomena and discuss their relevance in different types of systems. Two questions are on stake: Is there any universality in the aging phenomena? How can the comparison help define a better understanding and a more accurate description of the aging phenomena?

§2. Aging time dependence and scaling properties

Fig. 1. Response of a PVC sample submitted to a small stress after aging during $t_a$, as a function of time (from ref. 1).

Experimental evidence of aging appears, for instance, through the time dependence of the response functions at a given temperature after an initial quench from above the freezing temperature.
As the state of the system evolves with the aging time \( t_a \), which is by definition the total time spent at the working temperature after the initial quench, the dynamical response functions also change with \( t_a \). Hence, in the response to a small excitation applied after waiting a time \( t_w \) from the quench, two time scales are involved: \( t_a \), the aging time, and \( t \), the observation time, i.e. the time following the application of the excitation. The aging time \( t_a \) is then equal to \( t_w + t \).

Fig. 1, taken from,\(^1\) gives a generic example of the aging behavior in the mechanical properties of amorphous polymers. In these experiments, a PVC sample is quenched from 90°C (above the freezing temperature \( T_g \)) down to the working temperature \( T = 20°C \). A small mechanical stress is applied after a certain waiting time \( t_w \) (here denoted as \( t_e \)) and the strain is measured as a function of time \( t \). The plot shows the very slow relaxations and their strong \( t_w \) dependence up to the largest possible values of the waiting time. Similar properties are observed for the thermoremanent magnetisation (TRM) in spin glasses.\(^6–8\)

Fig. 2 reports results obtained on a silver compound containing 2.6% manganese impurities (Ag:Mn_{2.6\%}, \( T_g = 10.4K \)). In this case, the sample was quenched down to \( T = 0.87T_g \) in a small magnetic field which is removed after the waiting time \( t_w \). The figure displays the magnetisation as a function of \( t/t_w \), \( t \) being the time after the removal of the field.\(^8\)

Ac measurements also give very clear evidence for the aging phenomena. For instance, the ac shear modulus of polymers has been investigated,\(^9\) and ac measurements of the dielectric susceptibility are currently used in the investigation of amorphous polymers,\(^10\) supercooled liquids\(^11\) or orientational glasses.\(^12\) Fig. 3 gives, in the spin glass case, an example of the time dependence of the magnetic ac susceptibility. The observed behavior of the out-of-phase component \( \chi''(\omega, t) \) at frequency \( \omega \) corresponds to \( \chi''(\omega, t) = \chi''_0(\omega) + B(\omega t)^{-\beta} \), where \( \chi''_0(\omega) = A\omega^\alpha \) is the infinite time “equilibrium” value of the susceptibility, \( \beta \sim 0.2 – 0.4 \), and \( \alpha \) increases from 0.05 at low \( T \) to 0.2 close to \( T_g \).

Within the time scales used in these ac measurements, the aging part of the magnetic susceptibility of spin glasses is seen to scale as \( \omega t \). This should imply a \( t/t_w \) dependence for the aging part of the thermoremanent magnetisation as long as the excitation field is small enough to be in the linear regime for the response function. In the plot of fig. 2, a clear and systematic departure from a good \( t/t_w \) scaling can be seen, for two reasons: the equilibrium relaxation (corresponding to \( \chi''_0(\omega) \) in the ac experiment) has not been subtracted, and also \( t/t_w \) is not the correct scaling variable (a more detailed analysis is given in\(^8\)). This latter observation has early been made by Struik in amorphous polymers.\(^1\) It was noted that the relaxation curves of the compliance could only be superposed in the short time limit, by a logarithmic shift of the time scale with a factor \( \mu = 0.9 \), hence a scaling of the type \( t/t_w^\mu \). This suggested a logarithmic shift of all the response times, i.e. a scaling for their distribution as \( g(\tau, t_a) = G[\tau/(t_a^\mu \tau^\alpha)^{1-\mu}] \), where \( t_a = t_w + t \) is the aging time and \( \tau_a \) some time normalisation factor. With this assumption any infinitesimal change of
magnetisation can be written as: \( \frac{dm}{m} = \frac{dt}{\tau} = \frac{dt}{(t_a \tau_s^{-\mu})} \). Changing the time variable \( t \) into an effective time \( \lambda \) defined by \( d\lambda = dt.(t_w/t_a)^\mu \), \( \frac{dm}{m} \) becomes: \( \frac{dm}{m} = d\lambda/(t_w \tau_s^{-\mu}) \).

This entails the following scaling form for the aging part of the relaxation function: \( m(t, t_w) = M[\lambda/(t_w \tau_s^{-\mu})] \). The effective time \( \lambda \) is equal to \( t \) for \( t \ll t_w \) \( (t_a \approx t_w) \). For larger values of \( t \), it accounts for the change of aging time during the relaxation, which for long \( t \) is indeed not measured at a constant age of the system. Such a scaling for the relaxation function works remarkably well.
for the amorphous polymers. It works as well for all known spin glasses. An example is given in fig.4, where the whole set of data corresponding to $t_w$'s ranging from 300s to 30000s collapse on the same master curve.\textsuperscript{7,8)}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure4.pdf}
\caption{Aging part of the TRM (same data as in Fig.2), obtained after subtracting to the TRM a stationary contribution which is equivalent for the magnetisation to the “equilibrium” value $\chi''_0$ for the susceptibility. 5 curves, obtained for $t_w=300, 1000, 3000, 10000$ and 30000 s are superimposed as a function of the scaling variable defined in the text.}
\end{figure}

In other systems than spin glasses, the experimental results lead to a whole diversity of conclusions. It was found that the ac susceptibility of disordered ferromagnets followed an $\omega t$ scaling as in spin glasses,\textsuperscript{13)} but the ac dielectric susceptibility of supercooled glycerol\textsuperscript{11)} and of the orientational glass $K_{1-x}Li_xTaO_3$ with $x = 0.025$ (KLT)\textsuperscript{12)} did not. In the case of the orientational glass, the dielectric constant is frequency independent and only depends on the aging time after the quench.

\section*{§3. Cooling rate effects}

In most experimental situations the system is not instantly quenched, the cooling time is not quite negligible and may have an important effect. This is clearly the case for the KLT orientational glass,\textsuperscript{12)} which indeed shows a huge dependence on the cooling rate. The level of the in-phase dielectric susceptibility $\epsilon'$ is quite different for the various cooling rates explored. The difference is much larger than the amplitude of the relaxation itself, leading to an apparent equilibrium value at “infinite” $t_a$ which depends on the cooling rate. The smaller the cooling rate, the lower the
value for $\epsilon'$, and the smaller the amplitude of the relaxation. In this system, relaxations at higher temperatures strongly affect the low temperature value of the response function.

This is also the case in disordered ferromagnets. Due to "impurities" (or defects) acting as pinning centers, domain walls propagate very slowly through the network and the equilibrium distribution of domains can only be reached in very long times. The dynamics is fastest at the highest temperature compatible with the ferromagnetic groundstate, i.e. close to the transition temperature. This is seen in Fig. 5 which shows the behavior of the magnetic susceptibility of a ferromagnetic thiospinel sample around the ferromagnetic transition. This system ($CdCr_{1.9}In_{0.1}S_4^{14}$) is a $Cr^{3+}$ 95% version of the spin-glass thiospinel of Fig. 3 ($Cr^{3+}$ 85%). The out-of-phase susceptibility displays a marked hysteresis between successive cooling and heating procedures. The width of the hysteresis decreases as the temperature sweeping rate is decreased. The downward shift of the out-of-phase susceptibility $\chi''$ is related to the decrease of dissipative processes as the overall surface of the walls decreases. The effect on the in-phase susceptibility $\chi'$ is very small since this quantity is a volume response.

![Graph showing magnetic susceptibility behavior](image)

Fig. 5. $CdCr_{1.9}In_{0.1}S_4$ disordered ferromagnet: hysteresis behavior of ($\chi''$) in the vicinity of the ferromagnetic transition, upon cooling and re-heating at the different rates of 0.17K/min (full symbols) or 0.05K/min (open symbols). No hysteresis is visible on $\chi'$ (inset).

Similar cooling rate effects are present in supercooled liquids and in amorphous polymers. For instance, supercooled glycerol has a much lower value of the dielectric constant if aged at an intermediate temperature, compared to its value following a quench.\(^{11}\) In the case of amorphous polymers, cooling rate effects are found in temperature cycling experiments.\(^{10}\)
In this regard, spin glasses are quite singular, cooling rate effects are indeed very small compared to overall aging dynamics.\textsuperscript{15,16} Fig. 5 shows the influence of a very long stay (t=900 min) right below the transition ($T/T_g = 0.96$) on the relaxation at lower temperature ($T/T_g = 0.72$) in the CdCr$_{1.7}$In$_{0.3}$S$_4$ thiospinel spin glass.\textsuperscript{18} The TRM decay, as well as the $\chi''$ relaxation (inset), are completely insensitive to the 900 min stay at 0.96 $T_g$.

It is clear that the previous thermal history before reaching the working temperature had no relevant effect. \textit{Aging at higher temperature did not bring the system closer to its equilibrium at lower temperatures.} This conclusion is strongly supported by the temperature cycling experiments.

Fig. 6. CdCr$_{1.7}$In$_{0.3}$S$_4$ thiospinel spin glass ($T_g = 16.7 K$): influence of a 900 min aging time at 16K (0.96 $T_g$) on aging at 12K (0.72 $T_g$). The main part of the figure displays TRM relaxations. Solid lines are standard measurements for $t_w = 15, 30, 900$ min. Open circles: 900 min stay at 16K, and waiting time of 30 min at 12K. The inset shows the result of the same procedure in a 0.01 Hz $\chi''$ measurement. The stars are obtained for a direct quench, while the circles correspond to the intermediate waiting at 16K. The data are seen to be independent of the experimental procedure (from\textsuperscript{18}).

§4. Temperature cycling experiments

Fig. 7 gives an example of the spin glass behavior during a \textit{negative temperature cycling procedure}.\textsuperscript{8,17} The first remarkable point is the upward jump of $\chi''$ and the subsequent strong relaxation after the temperature decrease from 12K to 10K, even though the relaxation at 12K was already rather close to its equilibrium value. The observed relaxation at 10K is the same as the relaxation
obtained after a direct quench from above $T_g$ to 10K. *The aging processes appear to be reinitialised during a downward temperature change* (rejuvenation effect). The second remarkable point is that, as T comes back to 12K, $\chi''$ retrieves the value it had reached after the initial aging at 12K and resumes its relaxation. The inset of the figure shows that both relaxations at 12K are in exact continuation of each other. The temperature decrease and the relaxation at the lower temperature did not affect the state of the system at 12K. *The system kept the whole memory of its previous state* (memory effect).

![Diagram](image)

**Fig. 7.** $\text{CdCr}_{1.7}In_{0.3}S_4$ spin glass ($T_g = 16.7K$): effect of a negative temperature cycling $12K - > 10K - > 12K$ on the time dependence of $\chi''$ ($f=0.01\text{Hz}$). The inset shows the relaxation measured during $t_3$ plotted in continuation of the initial relaxation during $t_1$ (the solid line is a relaxation at $0.72T_g$ without temperature cycling).

These rejuvenation and memory effects can be nicely visualized using an alternative temperature cycling procedure. In this procedure (Fig.8), the susceptibility is continuously recorded as a function of temperature. The solid line in Fig.8 is a reference curve, obtained in a continuous temperature sweep at 0.1K/min. In another run, the continuous cooling is stopped at 12K for an aging stage of 7h (open diamonds), during which $\chi''$ relaxes downwards. The rejuvenation effect appears upon resuming the cooling from 12K: the $\chi''$ data increase back up to the reference curve within a rather small temperature variation. Yet, this reference curve is not an equilibrium curve; when, at 9K, the cooling is stopped once more (during 40h), $\chi''$ relaxes strongly downwards, and again merges with the reference curve when cooling continues down to 5K. When the sample is now continuously re-heated, $\chi''$ displays two sharp dips centered at 9 and 12K, matching the values
Fig. 8. Rejuvenation and memory effects on the ac out-of-phase susceptibility of the $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$ spin glass. The measuring frequency is 0.04Hz with an ac field of 0.3 Oe and a temperature sweeping rate of 0.1K/min. While cooling down (open diamonds), two stops are made: one at 12K during 7h, the other one at 9K during 40h. The reference curve (solid line) is measured during continuous re-heating after continuous cooling. The inset shows the same result on the Cu:Mn spin glass.\textsuperscript{16, 19}

reached after the aging period. Aging at both temperatures of 9 and 12K has remained imprinted in the system during the whole run at lower temperatures. As soon as the temperature becomes higher than that at which aging occurred, the memory effect disappears, and any further cooling procedure simply follows the normal reference curve. The inset of Fig.8 shows that the behavior observed for the $\text{Cr}^{3+}$ 85% thiospinel insulating compound is found with exactly the same characteristic features in the metallic spin glass Cu:Mn.\textsuperscript{19}

Similar experiments have been performed\textsuperscript{13} on the $\text{Cr}^{3+}$ 95% thiospinel sample of Fig.5 ($\text{CdCr}_{1.9}\text{In}_{0.1}\text{S}_4$\textsuperscript{14}), which shows a ferromagnetic transition at $T_c = 70K$, followed at $T_g = 10K$ by the reentrance of a spin-glass phase. At all temperatures below $T_c$, ferromagnetism has been characterized by neutron diffraction measurements.\textsuperscript{20} Important irreversibilities are evidenced below $T_c$ by a large split between the zero-field cooled and field-cooled curves, indicative of slow dynamics. Fig.9 presents the out-of-phase susceptibility $\chi''$ of this sample close to $T_c$ as a function of temperature, using a procedure equivalent to that of Fig.8. The curve with open diamonds corresponds to a run in which the temperature was decreased down to $T_1 = 67K$ where the system was aged for a time of 3.5h. This aging leads to a noticeable decrease of $\chi''$. The temperature was then further decreased, down to temperatures $T_d$ ranging between 64 and 30K. The curve never comes back exactly to the reference curve, but there is clearly some rejuvenation, since $\chi''$ first increases before
decreasing again as the temperature is lowered from $T_1$. In the subsequent reheating procedure, no dip can be observed in the case of the lowest $T_d = 30K$ (open circles). Thus no memory effect exists in this case. However, if the lowest temperature $T_d$ reached in the run comes closer to $T_1$, a partial memory can be detected. The amplitude of the memory effect increases as $T_d$ becomes closer and closer to $T_1$. In the reentrant spin-glass phase below 10K, the same rejuvenation and (full) memory effects as in usual spin glasses have been found.\(^{13}\)

Very recently, the non-diluted thiospinel $CdCr_2S_4$ has also been studied. Below the ferromagnetic transition at $T_c = 85K$, important irreversibilities are again found. At low temperature (5K), surprisingly, $\chi''$ shows a peak which is similar to that observed at the onset of the reentrant phase in the diluted thiospinels. However, the $\chi''$ relaxations in this region are very weak compared to the spin-glass case. In these thiospinel compounds, the first-neighbour interactions are ferromagnetic, while the next nearest-neighbour interactions are antiferromagnetic. It is therefore not excluded\(^{21}\) that the $\chi''$ peak signs up the reentrance of a strongly disordered (but almost frozen) low-temperature state, even in this non-diluted compound. Slightly below $T_c = 85K$, the same experimental procedure as in Fig.9 has been applied. In this region close to the ferromagnetic transition, aging relaxations are important, and the same rejuvenation and (partial) memory effects are found as in the diluted compound of Fig.9.

Similar experimental procedures have been applied to other systems. In most cases,\(^{10-12}\) no strong rejuvenation effects are found when going to lower temperatures. Aging at different temperatures is essentially cumulative, and is mainly important in the vicinity of the glass transition.\(^ {10}\) During a negative temperature cycling, all dynamics is considerably slowed down, and a “memory” of the previous stage is then naturally retrieved when heating back. However, this memory can be affected by the low-temperature evolution,\(^ {9,11,12}\) yielding a non-monotonic behavior of the susceptibility after the negative cycling (this is denoted as a “memory effect” in the case of polymers\(^ {9}\)). In the dielectric orientational glass $KTaO_3NbO_5$ (KTN\(^ {22}\)), however, rejuvenation and partial memory effects are found, in a very similar way as observed in the disordered ferromagnet Fig.9.

§5. Summary

A universal feature of the aging properties of disordered materials seems to be the existence of two intricated types of dynamic processes, whose relative importance changes from one system to the other. On the one hand, cumulative processes bring the system towards the same ground state whatever the temperature. The time evolutions at different temperatures are adding up, yielding important cooling rate effects. On the other hand, temperature specific processes (rejuvenation and memory effects) correspond to a restart of aging at each different temperature, but the previous relaxations at higher temperatures remain imprinted, and can be retrieved as the system is heated back to these temperatures. In polymers,\(^ {10}\) in supercooled glycerol\(^ {11}\) and in the orientational
Fig. 9. Rejuvenation and partial memory effect in the disordered ferromagnet $CdCr_{1.9}In_{0.1}S_4$. The procedure is similar to that of Fig. 8, except that the low-temperature excursion was limited to either $T_d = 64K$ (crosses), $60K$ (squares), $50K$ (triangles) or $30K$ (circles). For a short enough low-temperature excursion (e.g. $T_d = 64K$, crosses), the re-heating curves show a partial memory effect, which progressively faints out for lower and lower $T_d$ values.

glass KLT, the cumulative part was the most important. In disordered ferromagnets and in KTN, both contributions appear almost equally. In spin glasses, this cumulative part is almost negligible: the spin glass dynamics is mostly characterized by the rejuvenation and memory effects.

§6. Analysis and conclusion

With reference to the ferromagnetic behavior, the cumulative part of the dynamics may be ascribed to slow domain coarsening in a medium with randomly distributed pinning centers. The second (non-cumulative) part could then be due to domain wall reconformations, the domain walls trying to find the most favourable path through the nearby pinning centers with the smallest loss of elastic energy. Such an interplay between elastic forces acting in walls and random local pinning forces has been shown to lead to a hierarchical free energy structure. The reconformations indeed involve rearrangements over a large spectrum of characteristic length scales, the small length scales being affected by the large ones. Such reconformations can occur even at almost constant domain sizes.

It was already clearly demonstrated that a hierarchical free energy structure well accounts for the rejuvenation and memory effects. These can also be directly understood within the scheme of wall reconformations in the following way. Each rearrangement over a characteristic length $l$ is associated to an energy barrier $E(l)$ increasing with $l$. This energy is itself associated to a
characteristic time through thermal activation. A given temperature $T$ selects out a length $l^*$ such that $E(l^*) \sim k_B T$. Then all the modes corresponding to shorter length scales, hence smaller barriers, will be equilibrated in short times, whereas modes with larger length scales will correspond to much longer times and thus experience slow aging or almost complete freezing for the largest lengths.\(^{25}\) This accounts for the coexistence of equilibrium (stationary) and aging (non-stationary) dynamics. As $T$ is decreased, $l^*$ decreases. The slow modes freeze out, while the fast modes slow down and fall out of equilibrium due to the change of Boltzman weights. Some of these now enter the experimental time window and are seen as restarting aging processes (rejuvenation effect). As $T$ is brought back to the previous higher value, the frozen modes become active again and continue their former aging, while the other modes again become fast and equilibrate in short times (memory effect). If a significant domain coarsening has occurred during the temperature cycling, the wall reconformations take place in a different pinning environment after the end of the cycle, and the previous rearrangements are not relevant any more. The memory of aging at the initial temperature is lost.

The implication of this picture for spin glasses would be that domain coarsening is almost irrelevant, whereas the predominant part of dynamics is related to wall reconformations. The spin glass phase would thus appear as a dense network of domain walls.\(^{13, 25-27}\) In the absence of ferromagnetism, the exact nature of such walls and of the domains themselves (are they made of only two or many phases?) remains an open question. The growth of a spin-spin correlation length during aging at constant temperature has been characterized in simulations\(^{28}\) and in experiments.\(^{29}\) If spin-glass dynamics is to be seen as wall dynamics, the observed growth of a correlation length should be related to a progressive increase of the largest length scales involved in the domain wall reconformations. Rejuvenation effects probably imply that other (smaller) length scales are simultaneously involved.

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