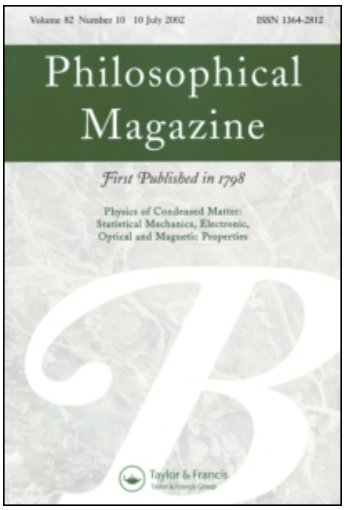

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Contrasting effects of field and temperature variations on ageing in spin glasses

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ABSTRACT

We present in this paper some new experimental results on the effect of temperature and field cyclings on the out-of-phase susceptibility of an insulating spin glass. The temperature-cycling experiments are consistent with a previously derived hierarchical scenario in which the probed metastable states continuously split into new substates as the temperature is lowered. The new results, however, suggest that the states of lowest free energy are not the same at different temperatures, supporting the idea of chaotic behaviour. Changing the magnetic field makes the system flow from one hierarchically organized set of states to another set of states with a different magnetization. An analysis of the field effect on the escape times of the states is proposed. It accounts for the observed behaviour of the a.c. susceptibility. It, moreover, predicts a scaling form for the time decay of the thermoremanent magnetization in terms of the probing field, which is nicely satisfied by the available experimental data.

§ 1. INTRODUCTION

Remarkable theoretical and experimental progress has been made recently on the problem of disordered systems and particularly on spin glasses. Many aspects of the complex structure of the phase space have been clarified. In the case of spin glasses, the most prominent conclusions were obtained from the investigation of the so-called ageing effects, that is the non-stationary dynamic properties (Hammann *et al.* 1992, Vincent, Hammann and Ocio 1992). Ageing, however, not only is observed in spin glasses, but also is a well known characteristic of the mechanical properties of amorphous polymers (Struik 1978). It is found in many other complex systems and seems to be related to a very common property of such systems.

A simple stochastic trap model has shown that ageing should, indeed, appear in any system in which the dynamics is governed by a distribution of relaxation times with a non-finite mean value (Bouchaud 1992). It takes such a system an infinite time to explore the accessible part of its phase space at any temperature below the freezing temperature T_g. In a given finite time (waiting time t_w), only a small subspace can be probed. The size of this subspace steadily increases with increasing t_w.

While the system slowly scans the phase space, its macroscopic properties evolve with time, reflecting the specific organization of the probed metastable states. A detailed investigation of the effects of small temperature or field changes on this evolution has been made. It showed the existence of a hierarchical structure and a continuous ramification of the metastable states with decreasing temperature (Refregier, Vincent, Hammann and Ocio 1987). A coherent picture could be derived in consistency with some predictions of the mean-field solution, and particularly with the idea of a continuous sequence of microphase transitions (Hammann *et al.* 1992).

After having reviewed the main features of the hierarchical picture, we discuss in

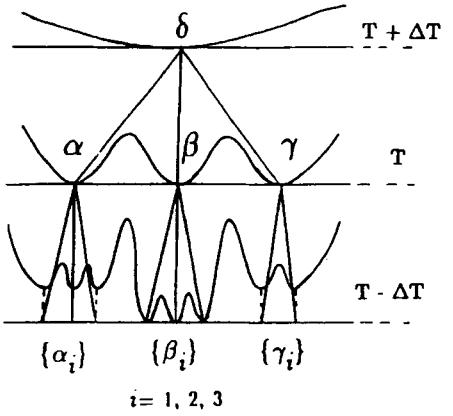
this paper some recent experimental results on the out-of-phase susceptibility and its dependence on temperature and field variations. We show that these results support and refine the proposed picture. Finally we develop an analysis which accounts for the observed effects and we suggest a scaling form for the time decay of the thermoremanent magnetization (TRM) in terms of the probing field, which is nicely satisfied by the available experimental data.

§ 2. The hierarchical picture

Various TRM and a.c. susceptibility experiments, on a metallic as well as on an insulating compound, led to a picture of a continuously ramifying organization of metastable states (Hammann *et al.* 1992, Vincent *et al.* 1992). These states were defined as local minima of a coarse-grained free energy, separated by finite barriers. They act as traps in which the system remains stuck for certain trapping or escapes times, related to the heights of the corresponding barriers. The observed properties, indeed, suggested a scenario in which lowering the temperature splits the metastable states into a large number of new states, these new states merging again when the temperature is raised back. The trapping times of the states increase as the temperature is lowered; so the most newly created states have the smallest trapping times. The proposed scenario is described in fig. 1.

The picture is, in many aspects, close to (and inspired from) the Parisi mean-field solution of the infinite-range Ising model (Sherrington–Kirkpatrick model). This solution predicts the existence of a low-temperature phase with a large number of nearly degenerate states (pure states). The number of these states increases exponentially with decreasing temperature. At a given temperature, the space of the pure states has an ultrametric topology. It can simply be represented by a hierarchical organization between the states in terms of their respective overlaps, where the overlaps are defined as a measure of the number of spins having the same orientation (Mézard, Parisi and Virasoro 1987).

Fig. 1



Sketch of the profile of the coarse-grained free energy and its evolution with temperature. As T decreases, each valley subdivides into new valleys between which the system slowly seeks out a new equilibrium. As T increases, the new valleys merge back into their ancestors. The states corresponding to each local minimum at a given T are represented as nodes on a horizontal line. The ramification of the states for decreasing T defines a hierarchical tree.

The Parisi solution does not predict the temperature dependence of the tree. There are, however, many theoretical approaches which suggest that the tree might develop continuously with temperature. In particular the so-called 'adiabatic cooling' approach directly derives an ultrametric organization of the pure states as a function of temperature (Dotsenko, Feigel'man and Ioffe 1990, Freixa-Pascual and Horner 1990).

However, the hierarchical structure described in experiments concerns only states separated by finite barriers, whereas, in theory, the pure states are all delimited by barrier heights increasing with increasing N (total number of spins in the system). These huge barriers prevent any transition between pure states in laboratory time scales. Yet, we have shown in previous publications that the finite barriers probed increase very steeply with decreasing temperature (Hammann *et al.* 1992). An extrapolation of their variation suggests a divergence at lower temperatures. As a consequence, the probed metastable states become pure states at lower temperatures and the experimentally observed structure is a preview of the organization of the pure states.

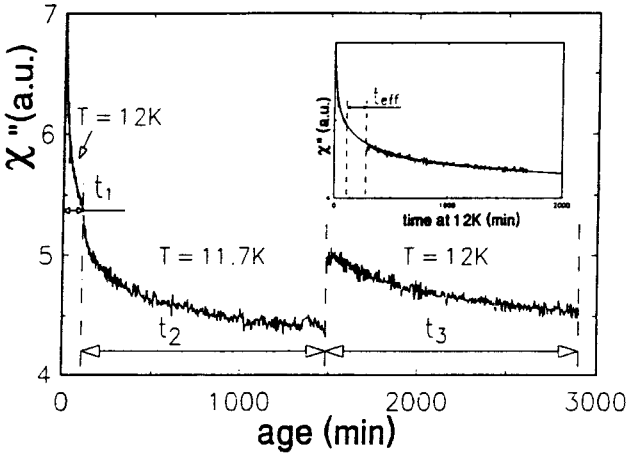
The suggested picture is finally that of a continuous series of microphase transitions as described by Mézard *et al.* (1984). As the temperature is lowered down to T_g, the first divergence of a barrier takes place, separating the phase space into a certain number of mutually inaccessible regions (ergodic components). There are diverging barriers at any temperature below T_g; the phase space is gradually divided into more and more such ergodic components. Within each region, there exists a whole distribution of smaller regions separated by finite barriers growing as the temperature is further lowered. The distribution of barrier heights extends up to infinite values and the time needed to explore a whole ergodic region is infinite. There is a coexistence of weak and true ergodicity breaking, as described by Bouchaud (1992) and found recently in a simple mean-field model (Cugliandolo and Kurchan 1993).

§ 3. Temperature cycles

The a.c. susceptibility experiments discussed in this paper have been undertaken on the well characterized chromium thiospinel compound $CdCr_{1.7}In_{0.30}S_4$ ($T_g = 16.7$ K) with an applied a.c. field in the range 0·01–0·1 Oe. Figure 2 presents the time dependence of the out-of-phase component χ'' at a frequency $\omega = 0.1$ Hz. In this experiment, the sample was quenched from above T_g down to $T = 12$ K ($T/T_g = 0.72$) at time (age) zero. The temperature was maintained constant for a time $t_1 = 110$ min, after which it was lowered to 11·7 K for a time $t_2 = 1400$ min and raised back to 12 K. The relaxation of χ'' was recorded during the whole procedure and for a time $t_3 = 1400$ min after the end of the temperature cycle.

During the first period the standard relaxation characteristic of ageing is observed. At the first temperature change, a new more drastic decay occurs. At the second step, χ'' returns almost immediately to the standard relaxation at a slightly smaller value than reached at the end of t_1. This is shown in the inset of fig. 2 where the data points during t_1 and t_3 have been superposed onto a standard relaxation curve. This standard curve following a simple quench, was determined in a separate experiment for long times (solid curve). The decay during t_1 is, of course, the same as the standard curve. The data points during t_3 can be superposed onto this curve if a shift in time is applied. In the inset, the t_3 decay curve is seen to be in good continuation with the t_1 curve after a time t_{eff}, much smaller than t_2. The large relaxation observed during t_2 at 11·7 K has only contributed an effective time $t_{eff} = 110$ min to the evolution of the system at 12 K. For large temperature variations (for instance $dT = 2$ K), t_{eff} becomes negligible and the

Fig. 2



Effect of a negative temperature cycling ($dT = -0{\cdot}3$ K) on the time dependence of $\chi''(\omega = 0{\cdot}1$ Hz) in the chromium thiospinel compound ($T_g = 16{\cdot}7$ K) (a.u., arbitrary units). The inset shows a plot of the data points recorded at $T = 12$ K during times t_1 before the dT cycle and t_3 after the dT cycle. The points taken after the cycle have been superposed on a standard relaxation curve (——) by shifting the time scale. The decay during t_3 is a continuation of the initial decay with an effective time interval $t_{eff} = 110$ min.

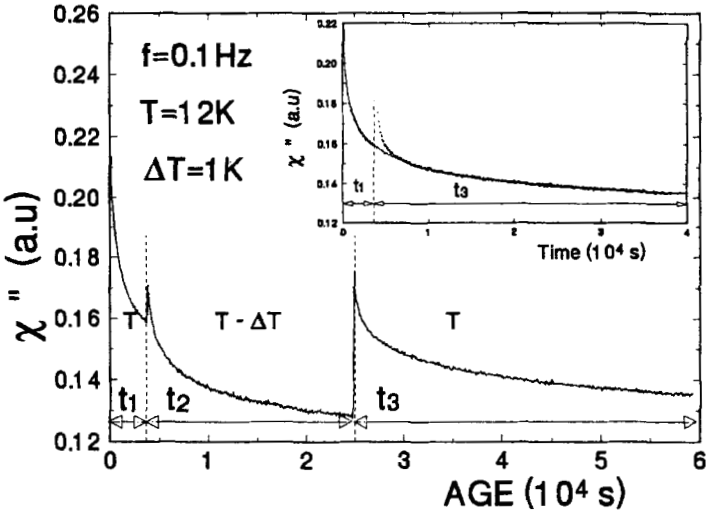
t_3 relaxation is just the exact continuation of the t_1 relaxation (Lefloch, Hammann, Ocio and Vincent 1992a). In that case, the relaxation during t_2 has been quite inefficient and did not bring the system closer to its equilibrium at 12 K, but the initial evolution t_1 has not been erased.

The behaviour supports the picture of ramifying states, schematized in fig. 1. The scenario, indeed, implies new ageing processes any time that the temperature is lowered, because of the existence of new states among which the system tries to equilibrate. It also yields a memory effect when the temperature is raised back since, as long as only processes between the newly born states occur at $T - dT$, the system will recover its initial state when it is brought back to its former ageing temperature. If dT is small enough or t_2 large enough that processes between higher states occur, then the system, once back to its former temperature, will have effectively aged for an equivalent time t_{eff}. The very small value of t_{eff} compared with t_2 is due not only to the decrease in the temperature but also to the large increase in the barriers, mentioned in the previous section. This increase, indeed, diminishes the probability of processes across barriers existing at the higher temperature.

However, the experimental situation is more complicated for intermediate dT values. In the range where t_{eff} has become very small but has not yet completely vanished, the system does not immediately recover the standard relaxation at the end of the temperature cycle. There is a transient faster relaxation for a certain time before the relaxation comes back to its normal time dependence. This behaviour was first mentioned by Anderson, Mattson and Nordblad (1993).

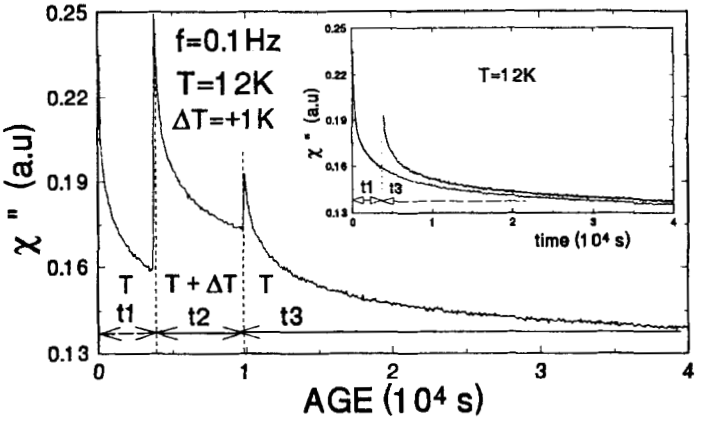
Such a behaviour is seen in fig. 3 which shows the results of an experiment with the same protocol as in fig. 2 but with $dT = -1$ K. From the inset, it is clear that the relaxation during t_3 is the normal continuation ($t_{eff} = 0$) of the relaxation during t_1 apart from a singular regime of about 35 min after the heating step. Again the temperature

Fig. 3



Effect of a negative temperature cycling ($dT = -1$ K) on the time dependence of $\chi''(\omega = 0{\cdot}1$ Hz) in the thiospinel compound ($T_g = 16{\cdot}7$ K) (a.u., arbitrary units). The inset shows a plot of the data points recorded at $T = 12$ K during times t_1 before the dT cycle and t_3 after the dT cycle. The points taken after the cycle have been plotted in continuation with the initial decay during t_1: (——) standard relaxation curve.

Fig. 4



Effect of a positive temperature cycling ($dT = 1$ K) on the time dependence of $\chi''(\omega = 0{\cdot}1$ Hz) in the thiospinel compound ($T_g = 16{\cdot}7$ K). (a.u., arbitrary units). The inset shows a plot of the data points recorded at $T = 12$ K during times t_1 before the dT cycle and t_3 after the dT cycle. The points taken after the cycle have been plotted in continuation with the initial decay during t_1: (——), standard relaxation curve.

cycle has not erased the initial ageing, since the t_3 relaxation can be superposed with the standard decay curve in continuity with the t_1 relaxation, if one forgets about the jump and the fast transient decay which appear at the beginning of t_3.

These singularities seem to occur in the region where the ageing at $T - dT$ still has some small effect on the ageing at T. If we consider fig. 1, we suppose that some processes over barriers, which already exist at T, can occur at $T - dT$ during t_2. That these processes do not contribute an effective time t_{eff} but lead to a singular relaxation may indicate that they did not make the system evolve towards the correct equilibrium distribution. It suggests that, as T is changed, the distribution of free energies is reshuffled. The states of lowest energy are not the same at different temperatures, meaning that the thermodyamic equilibrium phase is completely different from one temperature to the other.

The conclusion reached supports the idea of the chaotic nature of spin glasses as a function of temperature in the limit of thermodynamic equilibrium.

Finally, we shall illustrate the qualitative difference between positive and negative temperature cyclings. Figure 4 shows the behaviour of χ'' in an ageing procedure with a positive temperature cycle $dT = +1$ K at $T = 12$ K. The large overall value of χ'' during t_2 is related to the increase in its equilibrium value. A fast decay is observed during t_2. It is due to processes between states which were not accessible at 12 K and whose barriers have been lowered by the temperature step up to 13 K. When the initial temperature is recovered, a new relaxation appears. The inset of fig. 4 shows this t_3 relaxation shifted in time by t_2, that is plotted in continuity with the t_1 relaxation. In contrast with the behaviour observed in fig. 3, the t_3 relaxation remains here at a higher level than the standard relaxation. A larger shift towards smaller times must be applied to superpose the t_3 decay on the standard curve. The effective age of the system after the cycle is smaller than the initial waiting time t_1. This is consistent with the idea of renewed processes coming in during the temperature decrease, related to the growth of new barriers as expected from our suggested scenario.

It must be emphasized that this qualitative difference between increasing and decreasing temperature cycles is not due to the specific working temperature used; it exists in a large temperature region. It was actually checked down to 8 K ($T/T_g = 0{\cdot}48$).

§ 4. Field cycles

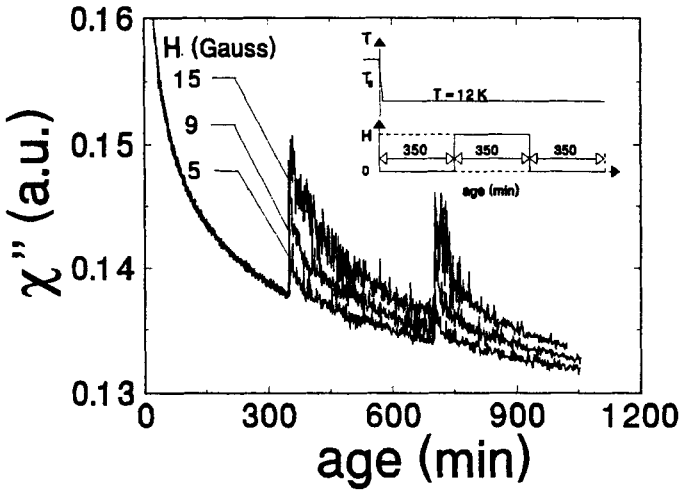
The asymmetrical behaviour characteristic of temperature cyclings does not exist in field cycling experiments (Lefloch, Hammann, Ocio and Vincent 1992b). During a relaxation measurement at a given temperature, both increasing and decreasing field cycles, around a given value, reinitialize most of the relaxation. After any field cycle, the system is brought back to an earlier stage of its expansion. No continuous evolution of the phase space structure as a function of field can be demonstrated.

The procedures used in the experiments presented in this paper are close to those described in the previous section. After an initial quench from above T_g down to the working temperature T, the χ'' relaxation is recorded for a time t_1. A d.c. field $h_{d.c.}$ is then applied for a time t_2. χ'' is continuously measured during the duration of the field cycle and for another time t_3 after the cycle.

Figure 5 shows the observed behaviour for $T = 12$ K and different fields $h_{d.c.}$. After each variation in the field, χ'' jumps to higher values and a renewed downward relaxation occurs. Ageing processes are restarted. The magnitudes of the jumps gradually increase with increasing field.

It must be noted that the scenario of ever-ramifying states as a function of

Fig. 5



Effect of field cyclings ($dH = 5$, 9 and 15 G) on the time dependence of $\chi''(\omega = 0{\cdot}1$ Hz) in the thiospinel compound (a.u. arbitrary units). At age zero the sample is cooled in zero field from above $T_g = 16{\cdot}7$ K to $T = 0{\cdot}72\,T_g$. The inset shows the experimental procedure.

temperature remains valid in a d.c. field when this field is applied before the initial quench and kept constant throughout the experiment (Lefloch *et al.* 1992b). The important difference is the different value of the magnetization. This value, however, remains constant along the tree, since the field-cooled magnetization is independent of temperature and time.

So, for each value of the field, there is a corresponding set of quasi-equilibrium states with equal magnetization and a hierarchical organization developing as a function of temperature. There is no obvious one-to-one relationship between the states of such two sets at different fields. Changing the field changes the active hierarchical tree corresponding to a different magnetization. Whether these trees are subsets of a unique tree relating all possible states, as suggested by Dotsenko (1993), cannot be answered in these experiments. The question now arises of how the system goes from one set to the other.

One can imagine the actual phase space as a mountain landscape with a valley corresponding to a minimum-energy path. At a given field, the minimum-energy path corresponds to an axis of constant magnetization in phase space. The local minima along this path are states or traps with a given lifetime which are hierarchically organized, that is smaller traps (holes along the valley) are located inside deeper holes which themselves are inside still deeper holes and so on. At constant field, the system only probes the traps along the valley. When the field is changed, a new valley is formed along an axis corresponding to a different magnetization, the former being lifted up the hillside. As the system escapes from one of the former traps, it has a high probability of flowing into the new set of traps.

§ 5. Tentative model for the behaviour of the susceptibility

Let us forget about the specific structure of the metastable states and only retain the existence of a wide distribution of characteristic lifetimes. Using the free-energy distribution function predicted by the mean-field theories, and relating the lifetimes to

these free energies, one obtains a power law distribution of the form $\psi(\tau) \propto \tau_0^x \tau^{-(1+x)}$ with $x < 1$. Such a distribution has no finite mean value. This is the starting basis of the stochastic model proposed by Bouchaud (1992) and used for the analysis of the thermoremanent magnetization (Bouchaud, Vincent and Hammann 1994).

The system is supposed to probe all the accessible states at random; it stays in each state for a time of order τ, the corresponding characteristic lifetime, and, when it gets out, it randomly falls into another state. Under these conditions, it was shown that the largest characteristic time encountered during a waiting time t is of order t, and the states of order $\tau = t$ are the most probable. The probability density of finding a given state τ after time t is $P(\tau, T) \propto \tau^{-x} t^{x-1}$ for $\tau < t$.

The out-of-phase susceptibility χ'' can then be calculated from

$$\chi''(\omega, t) = \int \frac{\omega\tau}{1 + \omega^2\tau^2} P(\tau, t)\, d\tau,$$

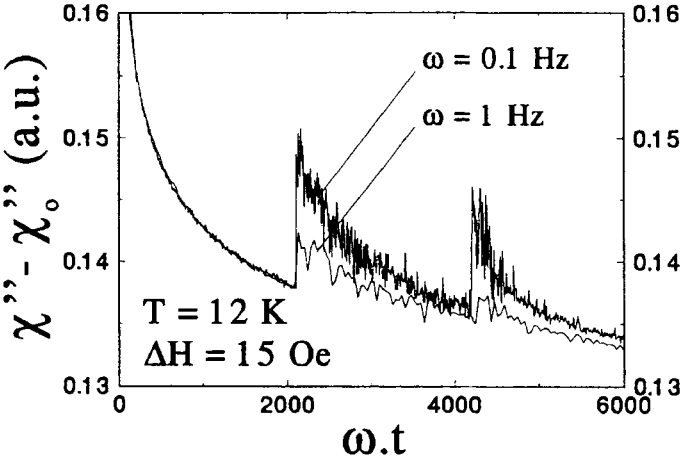
which leads to

$$\chi''(\omega, t) \propto (\omega t)^{x-1}, \quad x < 1.$$

This relation accounts well for the standard observed decay of χ'', which can be simply understood in the following way. At a certain frequency ω, χ'' mainly measures processes related to a characteristic time $1/\omega$, that is it probes essentially states with a lifetime $\tau = 1/\omega$; as it takes at least a time $t = 1/\omega$ to measure a given point, the state $\tau = 1/\omega$ has its largest probability of occupation at the first point; χ'' is thus at its maximum value. As the time evolves, the probed state has a population rate decreasing with time as t^{x-1}.

The expected power-law decay of χ'' can indeed be fitted to the observed relaxations. Its dependence on the reduced variable ωt can also be checked. In fig. 6, χ'' relaxations measured at two different frequencies are plotted as a function of ωt. In the initial time interval, the curves, indeed, fall on top of each other after vertical shift which allows

Fig. 6



Effect of a field cycling ($dH = 15$ G) on the time dependence of χ'' at two different frequencies $\omega = 1$ and 0·1 Hz, but the same ωt_1 value (a.u., arbitrary units). The χ'' data are plotted in terms of ωt (frequency times age). A vertical shift is applied in order to account for the different equilibrium values of χ''.

for the different equilibrium values, but the jumps at the change in field and the subsequent decays during t_2 do not scale. The jump is quite larger for a lower frequency.

The following step of the analysis is an attempt to understand these features.

As already stated previously, while ageing, the system evolves among states with same magnetization (M states) as inferred from the time independence of the field-cooled magnetization. When changing the field by dH, a new Zeeman term will be added which increases the free energy of the M states by $M\,\mathrm{d}H$ and tends to bring the system into a set of $M + \mathrm{d}M$ states. The simplest assumption is that this results in a decrease in the barriers by the same amount $M\,\mathrm{d}H$. In that case, the escape times $\{\tau\}$ of the whole set of states are lowered to $\{\tau'\}$ by the factor $\alpha = \exp(-b\,\mathrm{d}H)$: $\tau' = \alpha\tau, \alpha < 1$.

At the sudden field change, the τ distribution is shifted and the measurement now probes states which have a larger probability being closer to the most probable state αt_1. This induces a jump in χ''. The system looks younger as if it had only reached time αt_1. At t_1, a new decay is started with an apparent origin at time $t_1 - \alpha t_1$.

Within the previous assumption the decay curves during t_2 should be superposable on the t_1 curves with a certain adjustable horizontal shift smaller than t_1. This, however, does not work well in the experiments; the t_2 decays (see fig. 5) yield a slope which is always larger than in the initial t_1 decays. The assumption also predicts an ωt scaling for the jumps at the field change. The plot against ωt, shown in fig. 6, indicates that this is not the case, the jump being larger for smaller frequencies at a constant ωt_1.

Another mechanism must thus be added to account for the enhanced value and decay of χ'' after the field change. It looks as if a reinforced population rate of the probed traps occurs at the field variation. This enhancement is larger for smaller frequencies. A tentative explanation of this phenomenon is that the energy barriers are not equally affected by the change in field. Instead, we shall assume that deeper energy levels, that is higher barriers, are more influenced by the change in Zeeman energy.

The explanation finds its natural origin in the idea that escaping from a deeper state should involve more spin flips. This *ansatz* can be schematically understood by introducing a relation between the depth of a state and its extension in phase space. In that sense, to escape a deeper trap, one must go a longer distance to reach the top of the barrier. The Zeeman energy, brought in by the field change, depresses the top of the barrier mostly along the direction where the magnetization is changing. The probability of escaping the trap is largest along the direction and increases as the top of the barrier moves away from the centre of the trap, the difference in magnetization becoming larger.

In short, we suggest that the escape times τ of the traps are reduced by a factor $\alpha(\tau) = \exp[-b(\tau)\,\mathrm{d}H]$ where $b(\tau)$ is now an increasing function of τ. With this assumption, the increase in the jump of $\chi''(\omega t)$ with decreasing frequency but constant ωt_1 (see fig. 6) can be understood. At the change in field, the most populated trap of order t_1 is reduced to $\alpha(t_1)t_1$. As t_1 increases, this most probable trap is shifted towards smaller escape times and, for a constant ωt_1, comes closer to the probed trap $1/\omega$, yielding an enhanced response.

§ 6. Scaling of the remanent magnetization with field

With the previous analysis, it should also be possible to account for the field effects on the relaxation of the TRM. This remanent magnetization is obtained after field cooling the sample from above T_g down to the working temperature T and removing the field after a certain waiting time t_w at T. The TRM decays with time and the decay

curves depend on the applied field and on the waiting time t_w. For vanishingly small fields, the curves scale approximately as t/t_w.

In these procedures, the system first ages at constant field for a time t_w. The most populated traps just before removing the field are thus of order t_w. At the field change, however, the escape times of these traps are suddenly reduced from t_w to $\alpha(t_w)t_w$. This is equivalent to decreasing the age of the system by the same factor. The resulting relaxation curves should then be a function of $t/\alpha(t_w)t_w$ rather than of t/t_w only:

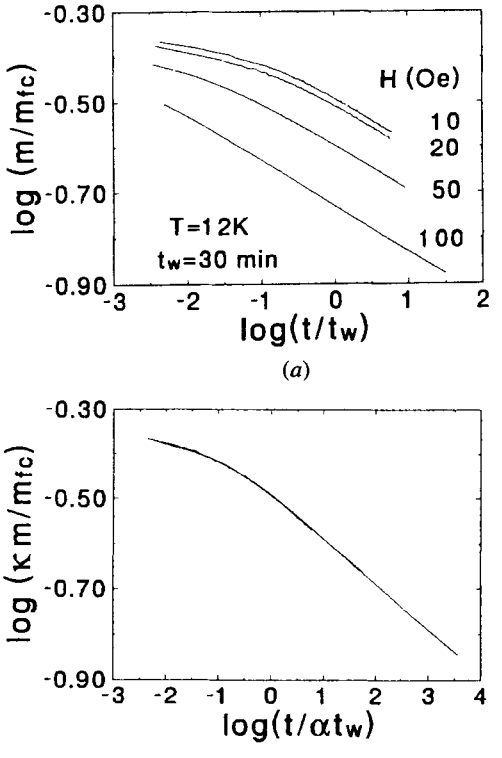
$$\frac{m}{m_{fc}} = kF\left(\frac{t}{\alpha(t_w)t_w}\right) \quad \text{with} \quad \alpha(t_w) = \exp[-\chi_0(t_w)H^2].$$

The present H^2 dependence of $\alpha(t_w)$ has been tentatively put in as an integrated form of the previously written term $b\,dH \propto \chi_0 H\,dH$. The factor k allows for a field and/or a t_w dependence of the initial fast response.

If plotted on a log–log scale, the relaxation curves at different applied fields should fall on a universal curve after application of the appropriate vertical and horizontal shifts.

Figure 7 (*a*) presents a set of thermoremanent curves measured at $T = 12\,K$ for

Fig. 7



Field dependence of the decay of the TRM at $T = 12\,K$ and t_w (waiting time) = 30 min in the chromium thiospinel compound: (*a*) log–log plots of the TRM over the field-cooled magnetization m_{fc} against the observation time t over t_w; (*b*) the curves corresponding to the four values of the probing field ($H = 10$, 20, 50 and 100 G) have been shifted horizontally (factor α) and vertically (factor k), in the log–log plot, to fall in continuity with each other.

$t_w = 30$ min in the insulating thiospinel compound. They correspond to four different values of the probing field. The above-described scaling has been applied and the resulting plot is shown in fig. 7 (*b*). In the log–log plot of this figure, the various TRM decay curves have been shifted horizontally and vertically in order to make them coincide as accurately as possible. Obviously the curves fall together nicely and a universal behaviour can be derived. The horizontal and vertical shifts determine the factors α and k.

The current available results point to the following conclusions.

(1) The field dependence of $\ln \alpha$ seems to be H^2 as suggested, but more than the present four values of the field should be probed.
(2) The values of α definitely depend on t_w, as derived from experiments at various t_w values, but a more quantitative analysis is needed in order to determine the actual dependence.
(3) The vertical shift (k factor) which must be applied to bring the curves together also depends on the value of the field, but the dependence is much smaller than for α and is currently being investigated.

We finally wish to point out that the proposed field scaling of the TRM relaxations is in agreement with a more general scaling form suggested by Parisi (1995), namely

$$\frac{m}{m_{fc}} \propto F\left(\frac{t}{t_w}, h^2 t_w^{\lambda}\right).$$

This form suggests a t_w dependence of α as $\alpha = \exp(-Ah^2 t_w^{\lambda})$. From our present incomplete analysis, this would be consistent with the experimental data for an estimated value of λ of order 0·1–0·2. Yet this result must be confirmed and worked out more quantitatively.

§ 7. Conclusions

The a.c. susceptibility experiments described above addressed the question of the stability of the hierarchical scenario against temperature or field perturbations.

The results of temperature-cycling experiments are at the origin of the picture of continuously ramifying states as the temperature decreases. A positive temperature cycle from T to $T + dT$, during a relaxation measurement at T, does not bring short-time processes closer to equilibrium; rather it restarts ageing at short scales in agreement with the growth of new states. A negative temperature cycle does not erase the initial evolution. For small values of the temperature variation, the cycle contributes a very small effective time, compared with the cycle duration, to the ageing at T. For large values, the effective time is zero; the evolution at the lower temperature $T - dT$ is completely wiped out as the system is raised back to T. After the cycle is completed, the system immediately resumes its initial behaviour.

However, for the intermediate values of dT presented in this paper, the a.c. susceptibility does not immediately return to its initial relaxation just after the cycling; there is a jump and a transient fast decay before the normal relaxation is resumed. We suggest that this singularity is due to an interchange, with temperature, in the relevant lowest-lying states, due to a redistribution of the free energies. This would be consistent with the predicted chaotic behaviour of the thermodynamic equilibrium properties as a function of temperature. It would also imply that, as the temperature is

lowered and the system goes down the hierarchical tree, it falls farther and farther away from thermodynamic equilibrium.

The field-cycling experiments do not reveal any particular relationship between the states at different fields. The hierarchical structure seems only to hold within states at a given field, that is of a given magnetization. Changing the field corresponds to flowing from one set of hierarchically organized states with a given magnetization to another set with a different magnetization.

We suggest an analysis of the field effects on the out-of-phase susceptibility χ'' in the framework of the simple stochastic model which was already shown to account for the ageing behaviour of the TRM. The jump in χ'' after a small change in field and the subsequent decay cannot be well fitted if one assumes that all the states are equally affected by the field. Another contribution must be considered. We have assumed that deeper states are more sensitive to the field because many more spins must be flipped to cross their barriers.

From our analysis, a scaling of the time decay of the TRM in terms of the amplitude of the probing field is predicted. Such a scaling is shown to work quite well for the few relaxation curves analysed in this paper (see fig. 7). A more comprehensive investigation is currently under way.

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