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► **To cite this version:**

G.J.M. Koper, H.J. Hilhorst. A domain theory for linear and nonlinear aging effects in spin glasses. Journal de Physique, 1988, 49 (3), pp.429-443. 10.1051/jphys:01988004903042900 . jpa-00210713

**HAL Id: jpa-00210713**

**<https://hal.science/jpa-00210713>**

Submitted on 4 Feb 2008

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Classification  
 Physics Abstracts  
 75.40 — 75.10 — 05.40

## A domain theory for linear and nonlinear aging effects in spin glasses

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(Reçu le 21 octobre 1987, accepté le 2 décembre 1987)

**Résumé.** — Nous présentons une théorie sur le vieillissement dans les verres de spin soumis à une température et un champ magnétique extérieur dépendant du temps. On suppose qu'un état de verre de spin arbitraire hors équilibre peut être décomposé en une collection de « domaines  $(T_1, H_1)$  » (pour tout couple  $(T_1, H_1)$ ) par comparaison de cet état avec un état d'équilibre à une température  $T_1$  et dans un champ  $H_1$ . On se donne des équations d'évolution dans le temps décrivant à la fois la croissance et la fracturation de domaines, ainsi qu'une équation pour la relaxation magnétique à l'intérieur d'un domaine. L'interaction entre les deux longueurs caractéristiques suivantes est d'une importance cruciale : i) la dimension linéaire d'un domaine (qui dépend du temps), et ii) une longueur de recouvrement  $l(\Delta T, \Delta H)$  ; celle-ci indique jusqu'à quelle échelle de longueur on ne peut différencier deux états d'équilibre thermodynamiques qui diffèrent par  $\Delta T$  et  $\Delta H$ . Nous montrons que la théorie explique une variété d'effets de vieillissement expérimentaux comme en ont observés en particulier Refregier *et al.* et Lundgren *et al.*

**Abstract.** — We present a theory of aging in spin glasses subjected to a time-dependent temperature and external magnetic field. An arbitrary nonequilibrium spin glass state is imagined to be decomposable into a collection of «  $(T_1, H_1)$ -domains » (for any pair  $(T_1, H_1)$ ) through a comparison of this state to an equilibrium state at a temperature  $T_1$  and in a field  $H_1$ . The theory postulates a time evolution for the domains (comprising both growth and breakup), as well as an equation for the magnetic relaxation within a domain. Of crucial importance is the interplay between two characteristic lengths : i) the time-dependent linear size of a domain, and ii) an overlap length  $l(\Delta T, \Delta H)$  ; the latter indicates up until which length scale two thermodynamic equilibrium states differing by  $\Delta T$  and  $\Delta H$  are indistinguishable. We show that the theory explains a variety of experimental aging effects as have been observed in particular by Refregier *et al.* and by Lundgren *et al.*

### 1. Introduction.

The phenomenon of aging in spin glasses was discovered in 1983 by Lundgren *et al.* [1]. It has since then been confirmed and investigated by many workers on a variety of spin glasses. Here we present a theory of aging in spin glasses. In subsection 1.1 we briefly discuss the context within which aging is observed and in subsection 1.2 we give a summary of the paper.

**1.1 AGING IN SPIN GLASSES.** — In a typical spin glass relaxation experiment (« field jump » experiment) one rapidly cools a sample in a magnetic field  $H$  to a temperature  $T$  below the freezing temperature  $T_g$ . One then cuts the field and observes the relaxation of the magnetization  $M(t)$ , which in that case is called the *thermoremanent* magnetization :  $M(t) = M_{\text{TRM}}(t)$ . Alternatively one may cool the sample in zero field to the temperature  $T$ , then

switch a field on and watch the increase of  $M(t)$ , then called the *zero field cooled* magnetization :  $M(t) = M_{\text{ZFC}}(t)$ . Obviously

$$\lim_{t \rightarrow \infty} M_{\text{TRM}}(t) = 0, \quad \lim_{t \rightarrow \infty} M_{\text{ZFC}}(t) = M_{\text{eq}}(H), \quad (1.1)$$

where  $M_{\text{eq}}(H)$  denotes the equilibrium magnetization in a field  $H$ . Experimentally one finds that  $M_{\text{TRM}}(t)$  and  $M_{\text{ZFC}}(t)$  relax extremely slowly (the relaxation is spread over a microscopic, a macroscopic and an astronomical time scale !) and nonexponentially. Except for temperatures very close to  $T_g$ , the system is still far from equilibrium even after the longest possible observation time, which is of the order of days.

Several theories have attempted to find the functional dependence of  $M(t)$  on time (see e.g. the review article [2]). In particular, logarithmic, power law and stretched exponential (or Kohlrausch) relax-

ation have been proposed and used to fit experimental data. It gradually appeared, however, and was first pointed out by Lundgren *et al.* [1], that in very small magnetic fields these relaxation curves are not uniquely determined: they turn out to depend on the *waiting time*  $t_w$  that has elapsed between the moment  $t = 0$  at which the temperature  $T_g$  was reached, and the moment  $t = t_w$  at which the field was cut (in a TRM experiment) or switched on (in a ZFC experiment).

During the waiting time one has not detected experimentally any changes in the system; in particular its magnetization remains constant (except for a slight ( $\sim 1\%$ ) decrease after field cooling). Nevertheless, the *response* of the system to a jump in the magnetic field at  $t = t_w$  becomes *slower* as  $t_w$  gets larger [1]. This indicates that during the waiting time the system is not in equilibrium but only slowly evolves towards it: it *ages*. Aging effects also occur when not the field but the temperature is varied (see Nordblad *et al.* [3] and Refregier *et al.* [4]), and can be detected by measurements not only of the magnetization but also of the ac susceptibility (see Lundgren *et al.* [5, 6], Svedlindh *et al.* [7], and Refregier *et al.* [4]) and of magnetic noise (see Ocio *et al.* [8]).

**1.2 SUMMARY.** — In section 2 we succinctly review the phenomenological linear response description of aging and some of its implications. We argue that there are experimentally observed aging phenomena for which this description fails. In the remainder of this work we then develop a theory which is also capable of describing a variety of experiments in which nonlinearities dominate.

The theory describes the magnetic response of the system in the presence of an arbitrary (but small) time-dependent magnetic field  $H(t)$  and temperature  $T(t)$ . Its general framework is given in section 3. The theory postulates a mesoscopic picture of time-dependent domains of correlated spins and assumes that the magnetic response behaviour of a spin depends on the size of the domain in which it is located. A fundamental feature of the theory is the interplay between two lengths. The first one is the time-dependent linear dimension of a domain. The second one is an overlap length  $l(\Delta T, \Delta H)$  which indicates up until which length scale we cannot distinguish two thermodynamic equilibrium states whose temperature and magnetic field differ by amounts  $\Delta T$  and  $\Delta H$ , respectively; this length generalizes a length scale  $l_{\Delta T}$  introduced by Bray and Moore [9] to nonzero field.

In section 4 we present the simplest application of the theory, viz. to the field jump experiment at a constant temperature. In the limit of small magnetic field jumps one recovers the linear response phenomenology. We then turn towards nonlinear

phenomena. These are due to restrictions on the domain growth imposed by the overlap length. Our results for both the linear and the nonlinear effects are found to agree qualitatively with the experimental data [10]. In section 5 we apply the theory to more complicated recent relaxation experiments [3, 4] in which one carries out both magnetic field and temperature jumps. We review the experimental data and show how the theory accounts for the observed phenomena. In section 6 we consider experiments [4, 6] in which one observes the time dependence of the ac magnetic susceptibility following temperature jumps. We obtain again theoretical curves in good agreement with the experiments.

To our knowledge, the only earlier theoretical work which specifically focusses on aging in spin glasses is a mean field approach by Ginzburg [11]. In section 7 we comment on the relation of our work to Ginzburg's as well as to some more general work on spin glass dynamics, and we make a few concluding remarks.

## 2. Linear and nonlinear response.

In subsection 2.1 we briefly expose the phenomenological linear response description of a spin glass. We emphasize that all relevant quantities can be expressed in terms of a single response function  $R(t, t')$ . This is done in particular for the time-dependent ac magnetic susceptibility in subsection 2.2. Although most of the ideas of these two subsections occur and have been used in the literature, a coherent presentation has, to our knowledge, not been given. In subsection 2.3 we indicate how experimentally the limits to the validity of the linear response description manifest themselves.

**2.1 LINEAR RESPONSE.** — The waiting time effect mentioned in section 1.1 has been the subject of detailed studies. In field jump experiments one has varied the waiting time  $t_w$  from a few seconds to a day and over. One has also studied the response  $M(t)$  to time-dependent fields  $H(t)$  more general [12, 13] than the step functions of the TRM and ZFC experiments.

It has been firmly established by several groups and in a variety of experiments that for reasonably small field values (say  $\leq 10$  G) the response  $M(t)$ ,  $t > 0$ , is *linear* in the applied field  $H(t')$ ,  $t' > 0$ . (We recall that  $t = 0$  is the instant of time at which the sample was cooled to the temperature  $T$  below  $T_g$ .) The most general relationship which expresses this fact is

$$M(t) = M(0) + N\chi_{\text{eq}} \int_0^t dt' G(t, t') \dot{H}(t') \quad (2.1a)$$

$$= N\chi_{\text{eq}} \left( H(t) - \int_0^t dt' R(t, t') \dot{H}(t') \right). \quad (2.1b)$$

Here  $\chi_{\text{eq}}$  is the equilibrium dc susceptibility in zero field and  $N$  the number of spins in the sample. The response function  $G(t, t')$  measures the response at time  $t$  to a unit increase in magnetic field at time  $t'$ . The two expressions (2.1a) and (2.1b) are equivalent, the relaxation function  $R(t, t')$  being defined as

$$R(t, t') \equiv 1 - G(t, t'). \quad (2.2)$$

We have the properties

$$R(t, t) = 1, \quad \lim_{t-t' \rightarrow \infty} R(t, t') = 0. \quad (2.3)$$

Furthermore, when the time following the quench tends to infinity,  $R$  tends to the equilibrium relaxation function :

$$\lim_{t \rightarrow \infty} R(t, t - \tau) \equiv R_{\text{eq}}(\tau) \equiv 1 - G_{\text{eq}}(\tau). \quad (2.4)$$

The relations (2.1a) and (2.1b) essentially already occur in reference [12], but their significance has been insufficiently stressed : in the linear regime the function  $G(t, t')$  gives a complete description of the spin glass response to small magnetic field changes, including the aging behaviour.

In what follows we shall frequently consider the field jump experiment described by

$$H(t) = \begin{cases} H_0 & (t \leq t_w) \\ H_1 = H_0 - \Delta H & (t > t_w). \end{cases} \quad (2.5)$$

According to the linear response relationship (2.1b) the excess magnetization

$$\Delta M(t) \equiv M(t) - N\chi_{\text{eq}} H_1 \quad (2.6)$$

decays as

$$\Delta M(t) = N\chi_{\text{eq}} \Delta H R(t, t_w) \quad (t > t_w). \quad (2.7)$$

The thermoremanent magnetization  $M_{\text{TRM}}(t)$  and the zero field cooled magnetization  $M_{\text{ZFC}}(t)$  discussed in section 1.1 are special cases of equations (2.5) and (2.7) obtained by setting  $H_1 = 0$ ,  $H_0 = H$  and  $H_1 = H$ ,  $H_0 = 0$ , respectively. As a consequence

$$M_{\text{TRM}}(t) + M_{\text{ZFC}}(t) = N\chi_{\text{eq}} H, \quad (2.8)$$

a relation satisfied to a high degree of accuracy in experimental results [13, 14].

**2.2 TIME-DEPENDENT AC SUSCEPTIBILITY.** — Within the above formalism one can discuss the time-dependent ac susceptibility. We suppose that a sample is cooled in zero field at time  $t = 0$  and that after a waiting time  $t_w$  an ac field with frequency  $\omega$  is applied :

$$H(t) = \begin{cases} 0 & (t \leq t_w) \\ H_0 e^{-i\omega t} & (t > t_w). \end{cases} \quad (2.9)$$

Upon applying a partial integration to (2.1b) and using (2.9) we find

$$\begin{aligned} M(t) &= N\chi_{\text{eq}} \left( R(t, 0) H(0) + \int_0^t dt' \frac{dR(t, t')}{dt'} H(t') \right) \\ &= -N\chi_{\text{eq}} H(t) \int_0^{t-t_w} d\tau \times \\ &\quad \times \frac{dR(t, t-\tau)}{d\tau} e^{i\omega\tau} \quad (t > t_w). \end{aligned} \quad (2.10)$$

If the coefficient of  $H(t)$  in (2.10) varies only little over a period  $2\pi/\omega$ , then it can be identified with the *time-dependent ac susceptibility*,

$$\chi(\omega; t) = -\chi_{\text{eq}} \int_0^{t-t_w} d\tau \frac{dR(t, t-\tau)}{d\tau} e^{i\omega\tau}. \quad (2.11)$$

Lundgren *et al.* [6] were the first to report on such time-dependence of the ac-susceptibility. They found power law decay towards the equilibrium value with an exponent  $\approx 0.5$  in their experiments on Cu (4 at % Mn). (This power is identical to the one occurring in  $R$  if  $R$  is assumed to decay as a stretched exponential ; see Ocio *et al.* [8].)

In the limit  $t \rightarrow \infty$  the expression (2.11) reduces to the ac susceptibility of the system in equilibrium,

$$\begin{aligned} \chi(\omega) &\equiv \lim_{t \rightarrow \infty} \chi(\omega; t) \\ &= -\chi_{\text{eq}} \int_0^\infty d\tau \frac{dR_{\text{eq}}(\tau)}{d\tau} e^{i\omega\tau}, \end{aligned} \quad (2.12)$$

where (2.4) has been used ; with the aid of (2.3) one sees that  $\chi(0) = \chi_{\text{eq}}$ .

In order to proceed we now exploit the fact that the experimental data (e.g. [10, 15]) are well described by a relaxation function  $R(t, t')$  of the form

$$R(t, t') = \left( 1 + \frac{t-t'}{t_0} \right)^{-\alpha} F(t, t'), \quad (2.13)$$

in which  $\alpha$  is a temperature dependent exponent in the range from 0.05 to 0.2 and  $F$  is a function for which  $F(t, t) = 1$  and

$$\lim_{t \rightarrow \infty} F(t, t-\tau) = 1 \quad (\tau \text{ fixed}). \quad (2.14)$$

The nontrivial implication is that from (2.4), (2.13) and (2.14) one has

$$R_{\text{eq}}(\tau) = \left( 1 + \frac{\tau}{t_0} \right)^{-\alpha}. \quad (2.15)$$

Such power law relaxation in a system in equilibrium has also been found, e.g., in Monte Carlo simulations [16]. The expression (2.15) satisfies  $R_{\text{eq}}(0) = 1$  ; however, the time constant  $t_0$  is so small

that for all times  $\tau$  for which data are available (i.e.  $\tau \geq 10^{-6}$  s [12]) one has  $\tau/t_0 \gg 1$ , and hence  $R_{\text{eq}}(\tau) \approx (\tau/t_0)^{-\alpha}$ .

In the remainder of this work the form (2.15) of the equilibrium relaxation function will be taken for granted, and we shall be concerned with the mechanisms by which deviations from it are generated when the system is out of equilibrium. These deviations are represented by the function  $F$ ; in the following sections explicit expressions for  $F$  will be derived on the basis of an underlying physical picture.

Upon using that  $|\omega|t_0 \ll 1$  for all frequencies of interest we find from equation (2.12) and equation (2.15)

$$\chi(\omega) \approx \chi_{\text{eq}} \left\{ 1 - (|\omega|t_0)^\alpha \Gamma(1-\alpha) \times \right. \\ \left. \times \left( \cos \frac{\alpha\pi}{2} - i \operatorname{sgn} \omega \sin \frac{\alpha\pi}{2} \right) \right\}. \quad (2.16)$$

It was first noticed by Lundgren *et al.* [5] that the real part  $\chi'$  and the imaginary part  $\chi''$  of the ac susceptibility approximately satisfy a «  $\frac{\pi}{2}$  law » :

$$\frac{\pi}{2} \frac{\partial \chi'(\omega)}{\partial \log \omega} \approx -\chi''(\omega) \quad (\omega > 0). \quad (2.17)$$

In reference [5] the relation (2.17) was proved to hold approximately for a spin system having a broad spectrum of relaxation times. It was subsequently derived and discussed by Pytte and Imry [17] in the wider context of scaling in random systems governed by thermally activated processes. In the present case we may verify explicitly that (2.17) is also valid for the equilibrium susceptibility (2.16), provided  $\alpha$  is not too large with respect to unity.

**2.3 NONLINEAR RESPONSE.** — We shall especially be interested in explaining what happens beyond the limits of the linear response regime. Experimentally these limits appear, for example, in the field jump experiment described by equation (2.5) as we shall briefly discuss now. Equation (2.7) shows that in the linear response regime  $\Delta M(t)/\Delta H$  is independent of both  $H_0$  and  $\Delta H$ . When  $\Delta H$  grows one expects, of course, nonlinearities associated with those of the static susceptibility, i.e. with the higher-order terms in the expansion

$$\frac{1}{N} M_{\text{eq}}(H) = \chi_{\text{eq}} H + \chi_{3,\text{eq}} H^3 + \dots \quad (2.18)$$

These nonlinear terms can no longer be neglected when  $\Delta H$  becomes comparable to  $\sqrt{|\chi_{\text{eq}}/\chi_{3,\text{eq}}|}$ . Recent experimental evidence [12, 13, 14] indicates, however, that when  $\Delta H$  increases a different and more important nonlinearity manifests itself in the dynamics well before  $\Delta H$  becomes comparable to  $\sqrt{|\chi_{\text{eq}}/\chi_{3,\text{eq}}|}$ . Its effect is to accelerate the relaxation in a way which *does* depend on  $\Delta H$ .

In the following sections we shall construct a theory capable of explaining both the linear and the nonlinear observed behaviour. We shall do so on the basis of a few physical hypotheses, in particular the one of time-dependent domains. Our hypotheses will lead us to a theory for the spin glass response which is in form identical to the linear response relation (2.1b). There is again a « relaxation function »  $R(t, t')$  which contains all basic information. The theory is, however, linear only in appearance, since  $R(t, t')$  will in general depend on the size of the magnetic field jumps.

In section 3 we present our general formalism. In section 4 we apply it to experiments with field jumps at a constant temperature, and in section 5 to experiments with both field and temperature jumps. In each case we describe the relevant experiments and show how these can be explained by the present theory.

### 3. A mesoscopic theory of spin glass response.

In this section we present the theoretical framework capable of explaining, at least qualitatively, all the observed phenomena. As we shall see when discussing the experimental results (in particular in Sect. 5 and 6), past values of temperature and magnetic field leave traces in the structure of the spin glass which at a later time may manifest themselves in the measurements. The main difficulty to be resolved is how to characterize this structure at the mesoscopic level. Our theory will be constructed in agreement with a few postulated principles that we shall list first, in section 3.1. In sections 3.2 to 3.4 these principles will then be converted into an operational tool.

**3.1 PRINCIPLES.** — i) A thermodynamic state of a spin glass at a temperature  $T_1$  and in a magnetic field  $H_1$  is characterized by a specific set of spin correlations. We shall, for short, refer to these as the  $(T_1, H_1)$ -correlations. In a system in equilibrium at  $(T_1, H_1)$  the range of the  $(T_1, H_1)$ -correlations is infinite.

ii) The correlations of two different thermodynamic equilibrium states, one at  $(T_1, H_1)$  and one at  $(T_2, H_2)$ , are nearly identical up to their « overlap » length  $l(T_1 - T_2, H_1 - H_2)$ . This length scale diverges as  $(T_2, H_2) \rightarrow (T_1, H_1)$ . In this sense, in a system (whether or not in equilibrium) which possesses  $(T_1, H_1)$ -correlations there also exist  $(T_2, H_2)$ -correlations, and *vice versa*.

iii) At any time, a nonequilibrium spin glass may be analysed with respect to its  $(T_1, H_1)$ -correlations for an arbitrary choice of  $(T_1, H_1)$ . One then finds, in general, that the system is composed of domains within which there exist  $(T_1, H_1)$ -correlations, but beyond which the  $(T_1, H_1)$ -correlations are des-

troyed. We shall, for short, refer to these as  $(T_1, H_1)$ -domains. For a system in equilibrium at  $(T, H)$  one finds a single infinite  $(T, H)$ -domain. However in the same system one finds a distribution of  $(T_1, H_1)$ -domains centred around  $l(T_1 - T, H_1 - H)$ . In a general nonequilibrium system there will be, at a time  $t$ , a distribution  $p(s, t; T_1, H_1)$  of the sizes  $s$  of the  $(T_1, H_1)$ -domains. (This distribution does not itself play a predominant role in what follows, but we will comment on it in the appendix.)

iv) At the time  $t = 0$ , i.e. just after the quench to a temperature  $T < T_g$ , the system is totally disordered and all domain sizes are zero.

v) In a nonequilibrium system placed at a certain time  $t$  in a heat bath of constant temperature  $T$  and in a constant magnetic field  $H$ , the  $(T, H)$ -domains will start growing without limit. Furthermore, for any  $(T_1, H_1) \neq (T, H)$ , the  $(T_1, H_1)$ -domains will grow until they reach the overlap length  $l(T_1 - T, H_1 - H)$ . If at time  $t$  any  $(T_1, H_1)$ -domains larger than this size are present, these will subsequently be broken up into domains of this maximum size. The laws governing the growth and breaking-up have to be specified.

vi) We assume that magnetic relaxation within a domain is described by a linear response relation. Small (large) domains will be associated with short (long) relaxation times of the magnetization. The response function of a spin in a domain with a time-varying size  $s(t)$  is a complicated functional of  $s(t)$ , to be specified below.

The necessity to consider, *simultaneously and within one and the same system*,  $(T, H)$ -domains for more than one value of  $(T, H)$  (or even for a continuum of such values) appears when there are time-dependent fields or temperatures. Such a description is essential for a correct qualitative understanding of e.g. the modified field jump experiment of section 5 and the susceptibility experiments of section 6. But it is also logically unavoidable : if e.g. the field  $H(t)$  keeps alternating between two values  $H_1$  and  $H_2$ , then neither the  $(T, H_1)$ -domain structure nor the  $(T, H_2)$ -domain structure should be privileged.

We have tacitly assumed, for simplicity, that only a single equilibrium state exists for each pair  $(T, H)$ . If more states exist, labeled by an index  $\alpha$ , then we have to denote the equilibrium by a triplet of parameters  $(T, H, \alpha)$ , and the present theory has to be generalized accordingly. It would remain true, however, that for  $t \rightarrow \infty$  at constant  $T$  and  $H$  the system will tend to form a single infinite domain with correlations which are characteristic of a triplet  $(T, H, \alpha)$  for some  $\alpha$ .

3.2 THE OVERLAP LENGTH  $l(\Delta T, \Delta H)$ . — We first discuss a length which was introduced by Bray and

Moore [9, 18] (but see also Ref. [19]) within the context of the Fisher and Huse [19] droplet model for the spin glass phase. In this droplet model one supposes that spin glass behaviour can be understood in terms of reversals (« excitations ») of spin clusters (« droplets »). For large  $l$ , the free energy  $F_l$  associated with a typical low-lying excitation of a droplet of linear size  $l$  is assumed to scale as  $F_l \sim J(l/a)^y$ , where  $J$  is the typical coupling strength between neighbouring spins and  $a$  the typical spin spacing.

Bray and Moore, on the basis of this droplet model, associate with any temperature change  $\Delta T$  of the system in the spin glass phase a characteristic « overlap » length  $l_{\Delta T}$ . For small  $\Delta T$  this length behaves as

$$l_{\Delta T} \sim a \left| \frac{J}{\Delta T} \right|^{1/y} \quad (3.1)$$

where

$$\zeta = \frac{d_s}{2} - y, \quad (3.2)$$

with  $y$  a constant and  $d_s$  the fractal dimension of the droplets. The meaning of  $l_{\Delta T}$  is that two identical systems, one in equilibrium at a temperature  $T$  and one at a temperature  $T + \Delta T$ , have nearly identical spin-spin correlations over distances less than  $l_{\Delta T}$ , but completely different correlations beyond  $l_{\Delta T}$ .

Next we consider the more general case where there is both a change of temperature  $\Delta T$  and a change of magnetic field  $\Delta H$ . Our argument generalizes the one given by Bray and Moore [9]. We focus on a particular low-lying droplet excitation of excitation energy  $F_l$ . Due to the change  $\Delta T$  the free energy of this same excitation will change by an amount  $\Delta F_l^{(T)}$  which is proportional to  $\Delta T(l/a)^{d_s/2}$ , being the sum of  $\sim (l/a)^{d_s}$  contributions with uncorrelated signs. Similarly, the change  $\Delta H$  in the magnetic field will bring about a change  $\Delta F_l^{(H)}$  which is proportional to  $\mu \Delta H(l/a)^{d/2}$ , being the sum of the Zeeman energies of  $\sim (l/a)^d$  randomly oriented spins. The droplet excitation in question loses its property of being a low-lying excitation if  $\Delta T$  or  $\Delta H$  becomes so large that

$$F_l \sim |\Delta F_l^{(T)}| + |\Delta F_l^{(H)}|, \quad (3.3)$$

that is, if

$$c_1 \frac{|\Delta T|}{J} \left( \frac{l}{a} \right)^{\frac{d_s}{2} - y} + c_2 \frac{\mu |\Delta H|}{J} \left( \frac{l}{a} \right)^{\frac{d}{2} - y} = 1, \quad (3.4)$$

where  $c_1$  and  $c_2$  are numerical constants. Our length  $l(\Delta T, \Delta H)$  is defined as the solution of equation (3.4). For  $\Delta H = 0$  it reduces to the Bray and Moore length (3.1).

**3.3 MAGNETIC RELAXATION IN TIME-DEPENDENT DOMAINS.** — We start from the idea that a spin in a hypothetical *finite* domain in *equilibrium* responds linearly to magnetic field variations, the response being described by a relaxation function  $R(t, t')$  which depends on the domain size  $s$  and can be expressed as

$$R(t, t') = R_{\text{eq}}(t - t') F(t, t'). \quad (3.5)$$

Here  $R_{\text{eq}}$  is the relaxation function for an infinite domain in equilibrium, and  $F(t, t')$  is a cutoff reflecting the system size. For an equilibrium domain of size  $s$  a plausible choice is

$$F(t, t') = e^{-(t-t')/\tau_{\text{max}}(s)} \quad (3.6)$$

where  $\tau_{\text{max}}(s)$  is the maximum relaxation time in the relaxation spectrum of a size  $s$  domain. The equations (3.5) and (3.6) are of the same form as (2.13) and (2.15), except that now the function  $F$  has been given a physical interpretation. Due to the spin coherence within a domain,  $\tau_{\text{max}}(s)$  will increase with  $s$ . In this work we shall use the form

$$\tau_{\text{max}}(s) \approx t_1 \left( \frac{s}{a} \right)^z, \quad (3.7)$$

where  $t_1$  is a microscopic time and  $z$  a constant. A dependence of this kind is used in various other discussions of spin glass dynamics (see e.g. [20, 21, 22]).

We postulate now that when the domain size varies with time, the magnetic response of a spin is still determined by a relation which has the form of the linear response relationship (2.1b), but with a relaxation function  $R(t, t'; [s(t'')])$  which is a *functional* of the time-dependent size  $s(t'')$  (with  $t' \leq t'' \leq t$ ) of the  $(T(t), H(t))$ -domain of which that spin was part. We shall call  $s(t'')$  the  $(T(t), H(t))$ -*history* of that spin.

The total magnetization  $M(t)$  is then related to the field  $H(t')$  by a relationship which is formally equal to (2.1b), but with a relaxation function which is now a weighted sum on all possible  $(T(t), H(t))$ -histories of the spins in the system :

$$R(t, t') = \int D[s(t'')] f(T(t), H(t); [s(t'')]) \times \\ \times R(t, t'; [s(t'')]). \quad (3.8)$$

Equation (2.1b) together with equation (3.8) yields a relation between magnetization and magnetic field which is linear only in appearance. Nonlinearities come in implicitly *via* the mechanism which governs the evolution of the domain sizes and hence the distribution of the  $s(t'')$ , and which we shall, in turn, make dependent upon  $T(t'')$  and  $H(t'')$ .

It is natural to put, in analogy to (3.5),

$$R(t, t'; [s(t'')]) = R_{\text{eq}}(t - t') F(t, t'; [s(t'')]) \quad (3.9)$$

while a plausible generalization of (3.6) is

$$F(t, t'; [s(t'')]) = \exp \times \\ \times \left[ - \int_{t'}^t dt'' \tau_{\text{max}}^{-1}(s(t''); T(t''), T(t)) \right]. \quad (3.10)$$

Here the relaxation time  $\tau_{\text{max}}(s; T'', T)$  is a generalization of (3.7) in which the first temperature argument,  $T''$ , is the actual temperature of the sample and the second one,  $T$ , indicates that we are considering  $(T, H)$ -type domains. We shall write

$$\tau_{\text{max}}(s; T'', T) \approx t_1(T'', T) \left( \frac{s}{a} \right)^z \quad (3.11)$$

and specify only later the precise way in which  $t_1$  depends on  $T''$  and  $T$ . It is assumed throughout that magnetic field variations, which are associated with very small energy differences, do not affect the relaxation times.

**3.4 DOMAIN DYNAMICS.** — The weight function  $f(T, H; [s(t'')])$  introduced in (3.8) follows from the domain dynamics. We shall discuss this dynamics while trying to avoid any too detailed assumptions. Nevertheless, two dynamical processes have to be distinguished : the growing and the breaking-up of domains. We shall assume that the characteristic size  $\bar{s}(t)$  of a growing domain increases as a power of time,

$$\bar{s}(t) \approx a \left( \frac{t}{t_2} \right)^p, \quad (3.12)$$

where  $t_2$  is a microscopic time and  $p$  a constant. (Growth laws of domains in spatially uniform as well as disordered systems have been reviewed by Binder *et al.* [23]). For the purpose of dealing with situations in which the temperature varies with time it is useful to observe that (3.12) is the solution of the autonomous differential equation

$$\frac{d\bar{s}(t)}{dt} = pt_2^{-1} a^{\frac{1}{p}} \bar{s}^{\frac{p-1}{p}}. \quad (3.13)$$

We now postulate that in the general case of an arbitrary time-dependent temperature  $T(t)$  and field  $H(t)$  the  $(T_1, H_1)$ -history  $s(t'')$  is a stochastic process governed by the following rules (which implement (v) of Sect. 3.1) :

i) Whenever  $s(t) < l(T(t) - T_1, H(t) - H_1)$ , the domain size  $s(t)$  increases deterministically according to

$$\frac{ds(t)}{dt} = pt_2^{-1} (T(t), T_1) a^{\frac{1}{p}} s^{\frac{p-1}{p}}. \quad (3.14)$$

This is just the differential equation (3.13), but with  $t_2$  temperature dependent in order to take account of the temperature variations. The solution, with initial condition  $s(t_i) = s_i$ , is given by

$$s(t|t_i, s_i) = \left[ s_i^p + a^{\frac{1}{p}} \int_{t_i}^t dt'' t_2^{-1}(T(t''), T_1) \right]^p. \tag{3.15}$$

ii) When  $s(t) > l(T(t) - T_1, H(t) - H_1)$ , the domain size  $s(t)$  no longer increases but, in each time interval  $\Delta t$ , sticks to its value with probability  $1 - \tau_b^{-1} \Delta t$ , and jumps to the lower value  $l(T(t) - T_1, H(t) - H_1)$  with probability  $\tau_b^{-1} \Delta t$ . For  $\tau_b$  we take the maximum relaxation time of a domain of size  $l(T(t) - T_1, H(t) - H_1)$ ,

$$\tau_b = \tau_{\max}(l(T(t) - T_1, H(t) - H_1); T(t), T_1). \tag{3.16}$$

The two rules above fully determine the probability distribution  $f(T_1, H_1; [s(t'')])$  of the  $(T_1, H_1)$ -histories. One can, in principle, convert without difficulty the path integral in equation (3.8) into a sum on the number  $n$  of jumps with in the summand a multiple integral on the jump times  $\hat{t}_1, \hat{t}_2, \dots, \hat{t}_n$ . However, we shall not present any further formalism here but rather consider a variety of experimental situations.

**4. Field jump experiments at constant temperature.**

We shall apply the theory of section 3 to field jump experiments performed at a constant temperature  $T < T_g$ . We consider the experiment described by equation (2.5), where after a waiting time  $t_w$  the magnetic field is changed from  $H_0$  to  $H_1 \equiv H_0 - \Delta H$ . Of interest is the decay of the excess magnetization  $\Delta M(t)$  (see Eq. (2.6)). In a constant temperature experiment the general theory simplifies considerably. In particular, the time constants  $t_1(T'', T)$  and  $t_2(T'', T)$ , defined by equations (3.11) and (3.14) respectively, remain unaltered during the experiment and hence can simply be denoted by  $t_1$  and  $t_2$ . The overlap length discussed in section 3.2 will be denoted by  $l_{\Delta H}$  as it only depends on  $\Delta H$ :

$$l_{\Delta H} \sim |\Delta H|^{-2/(d-2y)}. \tag{4.1}$$

The calculation of the response  $M(t)$  after the field jump requires the knowledge of the relaxation function  $R(t, t_w)$  for  $t > t_w$  (Eq. (3.8)) which, in turn, only requires the knowledge of the  $(T, H_1)$ -histories for times after the field jump. Hence a single weight function,  $f(T, H_1; [s(t'')])$ , is involved. In order to proceed we shall assume that this weight function is centred around an average domain size  $\bar{s}(t)$ , and that therefore the functional integral in equation (3.8) is well approximated by the integrand evaluated in  $\bar{s}(t)$ . Together with equation (3.9) this leads to

$$R(t, t_w) = R_{\text{eq}}(t - t_w) F(t, t_w; [\bar{s}(t'')]). \tag{4.2}$$

According to the domain dynamics postulated in section 3.4, in the time interval  $(0, t_w)$  the domain size  $\bar{s}(t)$  cannot grow larger than the overlap length  $l_{\Delta H}$ . The interplay between these two lengths leads to the distinction of two cases that will turn out to correspond to linear and nonlinear behaviour, respectively: small field jumps (i.e.  $\Delta H$  small enough so that  $\bar{s}(t_w) < l_{\Delta H}$ ), discussed in subsection 4.1, and larger field jumps, discussed in subsection 4.2.

**4.1 SMALL FIELD JUMPS,  $\bar{s}(t_w) < l_{\Delta H}$ .** — After the quench to  $T < T_g$  in a field  $H_0$  the characteristic dimension  $\bar{s}(t)$  of the  $(T, H_1)$ -domains will begin to increase according to equation (3.12). We consider here field jumps which by hypothesis are small enough so that at  $t = t_w$  the characteristic dimension  $\bar{s}(t_w)$  is still less than  $l_{\Delta H}$ . For  $t > t_w$  the growth law (3.12) will continue to hold, and from equations (2.7), (4.2) and (3.10) we have that the excess magnetization  $\Delta M(t)$  will decay as

$$\Delta M(t_w + \tau) = N \chi_{\text{eq}} \Delta H R_{\text{eq}}(\tau) \exp \times \left[ - \int_{t_w}^{t_w + \tau} dt'' \tau_{\max}^{-1}(\bar{s}(t'')) \right]. \tag{4.3}$$

Using the experimentally obtained equilibrium relaxation function (2.15) and the power law dependencies (3.7) and (3.12) we find from (4.3) that

$$\Delta M(t_w + \tau) = N \chi_{\text{eq}} \Delta H \left( 1 + \frac{\tau}{t_0} \right)^{-\alpha} \exp \times \left[ - \frac{t_2^{pz}}{t_1(1-pz)} \{ (t_w + \tau)^{1-pz} - t_w^{1-pz} \} \right]. \tag{4.4}$$

Equation (4.4) is the result of our theory. It predicts a definite waiting time effect on the magnetic relaxation. The corresponding relaxation curves have been plotted in figure 1a for a number of values of the waiting time  $t_w$ . (From Alba *et al.* [10] we estimate  $\alpha = 0.05$ ,  $t_0 = 1.5 \times 10^{-17}$  min,  $pz = 0.9$  and  $t_1 t_2^{-pz} = 1 \text{ min}^{1-pz}$ ).

In the analysis of experimental data on single jump experiments it has become customary to plot not only  $M(t)$ , but also the « logarithmic » decay rate  $S(\tau)$  here defined by

$$S(\tau) = \left| \frac{1}{N \chi_{\text{eq}} \Delta H} \frac{dM(t_w + \tau)}{d \log \tau} \right|. \tag{4.5}$$

In figure 1b we show the decay rates corresponding to the curves of figure 1a. It appears that the function  $S(\tau)$  exhibits a clear maximum for a value of  $\tau$  to be denoted as  $\tau_m$ . Hence  $\tau_m$  can be considered as the crossover point for  $\Delta M(t_w + \tau)$  between the slow power law decay (« equilibrium relaxation ») due to the function  $R_{\text{eq}}$ , and the more rapid decay imposed by  $F$  (see Eq. (4.2)). The



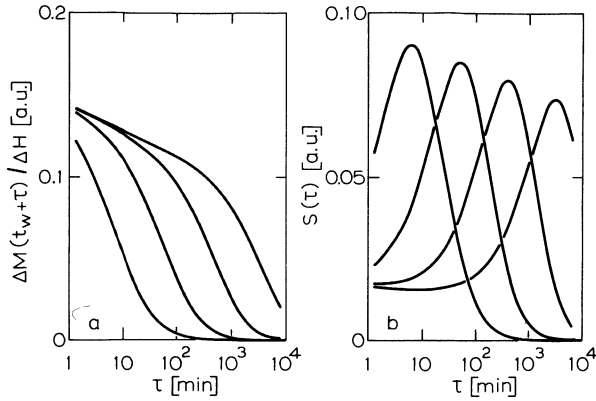


Fig. 1. — Theoretical curves for (a) the relaxation of the excess magnetization  $\Delta M(t_w + \tau)/\Delta H$ , equation (4.4), and (b) the logarithmic relaxation rate  $S(\tau)$ , equation (4.5), for small magnetic field jumps  $\Delta H$  and waiting times  $t_w = 10, 100, 1000, \text{ and } 10000$  min (from left to right). See the text for the parameter values.

experimental  $S(\tau)$  curves indeed exhibit a similar maximum [1, 15, 24]. Moreover, for several different types of spin glasses, such as Cu (5 at % Mn) [14, 15], Ag (2.6 at % Mn) [25], and  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  [4], experiments with waiting times  $t_w$  ranging up to  $10^4$  s show that this crossover takes place when the time  $\tau_m$  is roughly equal to the waiting time  $t_w$ , i.e.,

$$\tau_m = C(t_w) t_w, \quad (4.6)$$

with  $C$  a constant of order unity or at most a weakly varying function of  $t_w$ .

From equation (4.4) it follows that for the theoretical curves of figure 1b the proportionality (4.6) holds with  $C$  given by

$$C(t_w) \approx (1 - 2\alpha)/(pz + t_w^{1-pz} t_2^{pz} t_1^{-1}) \quad (4.7)$$

for small  $\alpha$ ,  $pz \approx 1$  and not too small  $t_w$  ( $t_w \geq 1$ ). For the parameter values given above and  $t_w$  in the experimental range we have that  $C(t_w)$  is of order unity and weakly varying with  $t_w$ . Hence we conclude that this kind of field jump experiments are well described by our theory.

**4.2 LARGER FIELD JUMPS.** — The second case to be considered is when the linear size of the  $(T, H_1)$ -domains reaches its upper limit  $l_{\Delta H}$  in the time interval  $(0, t_w)$ , so that  $\bar{s}(t_w) = l_{\Delta H}$ . After the field jump this limit is removed and the domain growth will proceed. With initial condition  $\bar{s}(t_w) = l_{\Delta H}$  the solution of equation (3.13) reads

$$\bar{s}(t) = a \left[ \left( \frac{l_{\Delta H}}{a} \right)^{\frac{1}{p}} + \frac{t - t_w}{t_2} \right]^p \quad (t > t_w). \quad (4.8)$$

The theoretical decay of the excess magnetization is

again given by equation (4.3), but now with the expression (4.8) for  $\bar{s}(t)$ . Hence the effect of the larger field jumps is reflected in the dependence of the characteristic domain size on  $\Delta H$  and this is the way in which the relaxation is *nonlinear* in the field jump  $\Delta H$ .

For this case one can also calculate the maximum in the logarithmic decay rate  $S(\tau)$  (Eq. (4.5)) and finds that now it is dependent on the size of the field jump  $\Delta H$  through

$$\tau_m = C(\tilde{t}_{\Delta H}) \tilde{t}_{\Delta H} \quad (4.9)$$

where the function  $C$  is as in equation (4.7) and

$$\tilde{t}_{\Delta H} \equiv t_2 \left( \frac{l_{\Delta H}}{a} \right)^{\frac{1}{p}}. \quad (4.10)$$

The equations (4.6), (4.9) and (4.10) can be merged into a single expression for the time  $\tau_m$  at which the logarithmic decay rate reaches its maximum, viz.

$$\tau_m = C(t_{\Delta H}) t_{\Delta H}, \quad (4.11)$$

where

$$t_{\Delta H} \equiv \min \left( t_w, t_2 \left( \frac{l_{\Delta H}}{a} \right)^{\frac{1}{p}} \right). \quad (4.12)$$

Hence, this picture of growing domains together with a domain size dependent relaxation process leads to

i) a *small  $\Delta H$  regime*, in which the relaxation is linear: curves for  $\Delta M(t)/\Delta H$  obtained at a fixed  $t_w$ , but at different values of  $\Delta H$  superimpose. In this regime the relaxation rate maximum occurs at a time  $\tau_m = C(t_w) t_w$  independent of  $\Delta H$ .

ii) a *regime of larger  $\Delta H$* , in which the relaxation is nonlinear: curves obtained at different  $\Delta H$  no longer superimpose. In particular, when  $\Delta H$  becomes larger the location  $\tau_m$  of the relaxation rate maximum begins to depend on  $\Delta H$  and shifts to smaller values according to

$$\tau_m \approx t_{\Delta H} \sim |\Delta H|^{-2/p(d-2y)}. \quad (4.13)$$

The crossover between the two regimes occurs at a value of  $\Delta H$  which satisfies  $\bar{s}(t_w) = l_{\Delta H}$ . This equation therefore divides the  $t_w - \Delta H$  plane into a region of linear and a region of nonlinear relaxation. In particular it predicts that the *longer* is the waiting time, the *smaller* is the field jump needed to provoke nonlinear relaxation [10, 13, 26, 27]. It is furthermore apparent from the above discussion that, if one accepts the power law dependencies (3.7) and (3.12) with exponents  $z$  and  $p$ , respectively, the *linear* relaxation depends only on the product  $pz$ , which should take a value close to unity. The *nonlinear*

relaxation, however, allows for a determination of  $p$  and  $z$  individually *via* equation (4.13). We have attempted an estimate based on the work of Alba *et al.* [10] on the insulating spin glass compound  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$ . Alba *et al.* report on field jump (TRM) experiments performed at a fixed temperature of  $0.72 T_g$  (12 K) where the field jump  $\Delta H$  is varied from effectively zero to 50 G for a range of waiting times  $t_w$  between 600 and 54,000 s and where the relaxation curves of the magnetization for times  $t > t_w$  are recorded. Subsequently their results are fitted to a master curve of which the parameter values for several  $t_w$  and  $\Delta H$  are given [10]. We have pursued this analysis in the following way. From the expression for the master curve we went back and calculated  $S(\tau)$ . We plotted  $S(\tau)$  and located the maximum  $\tau_m$  for the values of  $\Delta H$  and  $t_w$  using the parameters from [10]. Figure 2 shows a log-log plot of the  $\tau_m$  versus  $\Delta H$  for the given set of waiting times. We do not show  $\tau_m$  for  $\Delta H = 0$  and 10 G ; in

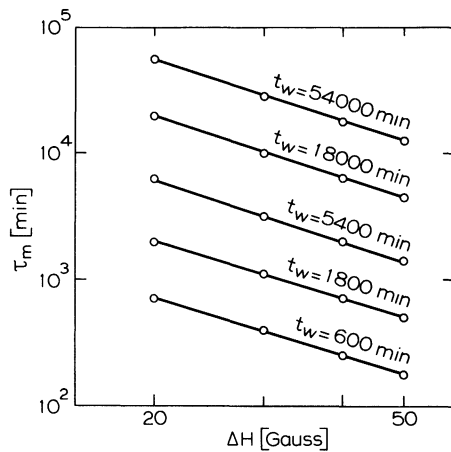


Fig. 2. — Location of the maxima  $\tau_m$  in the relaxation rate versus field jump size  $\Delta H$  for data taken from Alba *et al.* [10].

this field regime  $\tau_m$  levels off to a value independent of  $\Delta H$ . For each waiting time, the four values of  $\tau_m$  belonging to 20, 30, 40, and 50 G lie on a straight line whose slope is approximately  $-1.55$ . With this information we find the approximate value  $p \approx 0.5$  if we take  $y \approx 0.2$  for  $d = 3$  from reference [9]. This value of  $p$  for a real spin glass is, somewhat surprisingly, closer to the value  $\frac{1}{2}$  applicable to domain growth in spatially uniform Ising ferromagnets (see Blinder *et al.* [23]) than to the slow logarithmic growth law often found for random systems by theory [28] and by Monte Carlo simulation [29]. A possible explanation is hindered by the fact that, whereas ferromagnetic domains are easily observed experimentally, there is no direct way to see spin glass domains in the laboratory.

## 5. Relaxation experiments with field and temperature jumps.

In this section we shall consider a modification of the field jump experiment. The time dependence of the magnetic field is still given by equation (2.5), but in a time interval  $(\hat{t}_1, \hat{t}_2)$  during the waiting time period the temperature is kept at  $T + \Delta T$  instead of  $T$ :

$$T(t) = \begin{cases} T & (0 < t < \hat{t}_1) \\ T + \Delta T & (\hat{t}_1 < t < \hat{t}_2; \hat{t}_2 < t_w) \\ T & (\hat{t}_2 < t) . \end{cases} \quad (5.1)$$

Both positive and negative  $\Delta T$  will be of interest. In section 5.1 we shall first discuss the experimental data. Then we shall show, in section 5.2, how the theory developed above can account qualitatively for the observed phenomena. The formalism is *exactly the same* as was applied to the constant temperature experiments, except that we now have to allow for a temperature dependence of the times  $t_1$  and  $t_2$ . In section 5.3 we shall show how the extra parameters in the case of nonconstant temperature can be set to fit the experimental curves.

**5.1 EXPERIMENTAL RESULTS.** — The experiment described above has been performed by Nordblad *et al.* [3] and by Refregier *et al.* [4]. In the limit of sufficiently small  $\Delta T$  (for the examples of  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  [4] and Cu (10 at % Mn) [3] this means  $|\Delta T| \leq 0.1$  K) the magnetic relaxation curves coincide with the ones obtained at constant temperature  $T$  for a waiting time  $t_w$ . For sufficiently large *positive*  $\Delta T$  (which means in practice  $\Delta T \leq 2$  K for  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  when  $\hat{t}_2 - \hat{t}_1 = 300$  s [4]) the relaxation curve appears to coincide with the one at constant temperature  $T$  for a waiting time  $t_w - \hat{t}_2$ : at the time  $\hat{t}_2$  the system is « reborn » in the terminology of reference [3]. However, for intermediate values of  $\Delta T$  the situation is less simple: the relaxation curves obtained there do not coincide with the curves at constant  $T$  for any  $t_w$ , as demonstrated both by Nordblad *et al.* [3] and, in particularly clear way, by Refregier *et al.* [4]. The former authors speak of an « incompletely reborn system » and of the « coexistence of two separate and distinct aging states » in the same system. Our conclusion is that in this intermediate regime the system cannot be described by a single characteristic domain size, but that the distribution of domain sizes has more structure. This is what motivated the development of the full theory of section 3.

The modified field jump experiment with *negative* values of  $\Delta T$  [3, 4] shows a different picture. The situation is not symmetric with respect to  $\Delta T = 0$ . When  $\Delta T$  is sufficiently large negative, the relaxation

curves for  $t > t_w$  coincide with those obtained at constant temperature  $T$  for a waiting time  $t_w - (\hat{t}_2 - \hat{t}_1)$ , i.e. relaxation takes place as though the interval  $(\hat{t}_1, \hat{t}_2)$  of a decreased temperature had been completely nonexistent [4]. The correlations characteristic of the temperature  $T$  that were built up during the interval  $(0, \hat{t}_1)$  are not lost during the interval  $(\hat{t}_1, \hat{t}_2)$  but seem to remain frozen; then, once the temperature  $T$  is reestablished, their build-up continues. This shows that although a temperature jump  $\Delta T$  (or, by the same token, a field jump  $\Delta H$  [27]) may be large enough to require the breakup of existing correlations, this *breakup is not instantaneous* but should be described by a relaxation time.

**5.2 APPLICATION OF THE THEORY.** — We shall analyse the modified field jump experiments for values of  $\Delta H$  so small with respect to  $\Delta T$  that for all practical purposes we have

$$l(\Delta T, \Delta H) \approx l(\Delta T, 0) \equiv l_{\Delta T}. \quad (5.2)$$

We are again interested in the decay of the excess magnetization  $\Delta M(t)$  for  $t > t_w$ . Since we only need the relaxation function  $R(t, t_w)$ , equation (3.8), for  $t > t_w$ , a single weight function  $f(T, H_1; [s(t'')])$  for the  $(T, H_1)$ -histories is sufficient. We shall now derive an expression for this function.

We shall again assume that in the first time interval,  $0 < t < \hat{t}_1$ , the distribution of the sizes of the  $(T, H_1)$ -domains is well represented by a single characteristic length  $\bar{s}(t)$ . This length then grows according to the solution (3.15) of equation (3.14) and is given by

$$\bar{s}(t) = a \left( \frac{t}{t_2(T, T)} \right)^p \quad 0 < t < \hat{t}_1. \quad (5.3)$$

In the second time interval  $\hat{t}_1 < t < \hat{t}_2$ , there are two possibilities. The first one is that  $|\Delta T|$  is not too large so that  $\bar{s}(\hat{t}_1)$ , as given by equation (5.3), is less than  $l_{\Delta T}$ . In that case,  $\bar{s}(t)$  will continue to grow according to equation (5.3), albeit at a different rate due to the temperature dependence of  $t_2$ . Moreover, if  $\bar{s}(t)$  reaches the upper limit  $l_{\Delta T}$  before the time  $\hat{t}_2$ , then it will stick to that value. From  $\hat{t}_2$  on the domain size evolves again without limit, and hence we have for  $t > \hat{t}_2$

$$\bar{s}(t) = a \left[ \min \left\{ \left( \frac{l_{\Delta T}}{a} \right)^{\frac{1}{p}}, \frac{\hat{t}_1}{t_2(T, T)} + \frac{\hat{t}_2 - \hat{t}_1}{t_2(T + \Delta T, T)} \right\} + \frac{t - \hat{t}_2}{t_2(T, T)} \right]^p. \quad (5.4)$$

The second possibility is that  $|\Delta T|$  is so large that  $\bar{s}(\hat{t}_1) > l_{\Delta T}$ . In that case, according to the theory of section 3, during each time interval  $\Delta t$  the characteristic domain size  $\bar{s}(t)$  will stick to its value  $\bar{s}(\hat{t}_1)$  with probability  $1 - \tau_b^{-1} \Delta t$ , and jump to the lower value  $l_{\Delta T}$  with probability  $\tau_b^{-1} \Delta t$ . Here  $\tau_b$  is given by equation (3.16) as the maximum relaxation time of a domain of size  $l_{\Delta T}$

$$\begin{aligned} \tau_b &= \tau_{\max}(l_{\Delta T}; T + \Delta T, T) \\ &= t_1(T + \Delta T, T) \left( \frac{l_{\Delta T}}{a} \right)^z. \end{aligned} \quad (5.5)$$

Consequently, if  $\bar{s}(\hat{t}_1) > l_{\Delta T}$ , then for  $\hat{t}_1 < t' < \hat{t}_2$  there appear two different domain sizes: the « old » one  $\bar{s}_{\text{old}}(t') = \bar{s}(\hat{t}_1)$  and a « young » one  $\bar{s}_{\text{young}}(t') = l_{\Delta T}$ . The fraction  $f_{\text{young}}(t'; \hat{t}_1)$  of spins in domains of size  $l_{\Delta T}$  increases with time whereas the fraction  $f_{\text{old}}(t'; \hat{t}_1)$  of spins in domains of size  $\bar{s}(\hat{t}_1)$  decreases. At time  $t = \hat{t}_2$  we have

$$f_{\text{young}}(\hat{t}_2; \hat{t}_1) = 1 - \exp \left\{ - \frac{\hat{t}_2 - \hat{t}_1}{\tau_b} \right\}, \quad (5.6a)$$

and

$$f_{\text{old}}(\hat{t}_2; \hat{t}_1) = \exp \left\{ - \frac{\hat{t}_2 - \hat{t}_1}{\tau_b} \right\}. \quad (5.6b)$$

From  $\hat{t}_2$  on both characteristic domain sizes evolve again without upper limit according to the rate equation (3.14). This yields

$$\bar{s}_{\text{young}}(t) = a \left( \left( \frac{l_{\Delta T}}{a} \right)^{\frac{1}{p}} + \frac{t - \hat{t}_2}{t_2(T, T)} \right)^p \quad (t > \hat{t}_2) \quad (5.7a)$$

and

$$\bar{s}_{\text{old}}(t) = a \left( \frac{\hat{t}_1 + t - \hat{t}_2}{t_2(T, T)} \right)^p \quad (t > \hat{t}_2). \quad (5.7b)$$

Since the breaking-up process stops at  $t = \hat{t}_2$ , the fractions  $f_{\text{young}}$  and  $f_{\text{old}}$  remain constant for  $t > \hat{t}_2$ .

In the present case with *two* characteristic domain sizes the basic equations (3.8) and (3.9) for the relaxation function  $R$  reduce to

$$\begin{aligned} R(t, t_w) &= R_{\text{eq}}(t - t_w) \sum_{i = \text{young, old}} f_i(\hat{t}_2; \hat{t}_1) \times \\ &\quad \times F(t, t_w; [\bar{s}_i(t'')]) \quad (t > t_w). \end{aligned} \quad (5.8)$$

This expression also holds for the case  $\bar{s}(\hat{t}_1) < l_{\Delta T}$  provided that one take for  $\bar{s}_{\text{old}}(t)$  the expression (5.3), and put  $f_{\text{old}} \equiv 1$  and  $f_{\text{young}} \equiv 0$ .

The decay of the excess magnetization can be obtained by substituting equation (5.8) in equation (2.7) and using equation (3.10). In the next subsection we shall show how this leads to the theoretical curves for  $\Delta M(t)$  corresponding to the modified field jump experiment, equation (5.1).

**5.3 FIT TO EXPERIMENT.** — In order to obtain explicit theoretical curves it is necessary to specify the temperature dependence of  $t_1$  and  $t_2$  (Eqs. (3.11) and (3.14)), and of the constants in equation (3.4). We shall use the data from experiments carried out by Refregier *et al.* [4] on the insulating spin glass compound  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  to obtain values for the parameters that govern these temperature dependencies.

As discussed in section 3 the time constants  $t_i(T; T_1)$ ,  $i = 1, 2$ , refer to the dynamics of  $(T_1, H_1)$ -domains in a system at temperature  $T$ . For  $T = T_1$  we shall use the abbreviation  $t_{i0}(T_1) \equiv t_i(T_1; T_1)$ ,  $i = 1, 2$ . If domain growth is a thermally activated process, one would expect the  $T$ -dependence of the dynamics to manifest itself as an Arrhenius factor in the  $t_i$ . The experiments [4] show, however, a rather stronger asymmetry between positive and negative  $\Delta T$ . We therefore put, for general  $T$  and  $T_1$

$$t_i(T; T_1) = t_{i0}(T_1) A(T - T_1) \quad (5.9)$$

in which  $A(0) = 1$  and  $A$  increases steeply when  $T - T_1$  becomes negative. All strongly varying functions  $A(\Delta T)$  give qualitatively similar results, and we have obtained a good fit with the three parameter function

$$A(\Delta T) = x e^{-\gamma_+ \Delta T} + (1 - x) e^{-\gamma_- \Delta T}, \quad (5.10)$$

where  $\gamma_- = 18 \text{ K}^{-1}$  is large compared to  $\gamma_+ = 2.4 \text{ K}^{-1}$  and where  $x = 0.1$ . We set  $t_{10}(T) t_{20}^{-pz}(T) = 1 \text{ min}^{1-pz}$ ,  $\alpha = 0.05$  and  $t_0 = 1.5 \times 10^{-17} \text{ min}$  as in section 4.

Only two combinations of parameters from equation (3.4) play a role, viz.

$$k \equiv t_{20}(T) \left( \frac{J}{c_1} \right)^q \quad (5.11)$$

and

$$q^{-1} \equiv \left( \frac{d_s}{2} - y \right) p. \quad (5.12)$$

A fit to an analysis of the experiment by Refregier *et al.* [4] yields the values  $q = 1.66$  and  $k = 4.77 \text{ K}^q \text{ min}$ .

With the parameters listed above and the choice for  $A(\Delta T)$  given by equation (5.10) the theoretical curves of figure 3 are obtained. Figure 3a shows the curves for positive  $\Delta T$ . For sufficiently large  $\Delta T$  the fraction of spins with the « young » characteristic

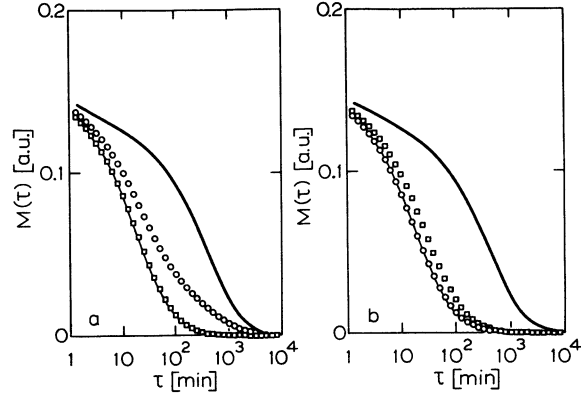


Fig. 3. — Theoretical curves for the relaxation of the excess magnetization where in a time interval  $(\hat{t}_1, \hat{t}_2)$  during the waiting time period  $(0, t_w)$  the temperature is kept at  $T + \Delta T$ . (a) shows the curves for positive  $\Delta T$  where  $\hat{t}_1 = 895 \text{ min}$ ,  $\hat{t}_2 = 900 \text{ min}$  and  $t_w = 930 \text{ min}$ . The solid lines correspond to TRM curves obtained for waiting times of 30 min and 930 min without temperature jumps. The squares ( $\square$ ) refer to the curve for  $\Delta T = 2 \text{ K}$  and the circles ( $\circ$ ) to the curve for  $\Delta T = 1 \text{ K}$ . (b) shows the curves for negative  $\Delta T$  where  $\hat{t}_1 = 15 \text{ min}$ ,  $\hat{t}_2 = 915 \text{ min}$  and  $t_w = 930 \text{ min}$ . Here the circles ( $\circ$ ) refer to  $\Delta T = -1 \text{ K}$  and the squares ( $\square$ ) to  $\Delta T = -0.3 \text{ K}$ . See the text for the parameter values.

domain size is dominant whereas for not too large positive  $\Delta T$  domains of the « young » and the « old » size coexist. This explains why the theoretical curve for  $\Delta T = 1 \text{ K}$  does not look like a « normal » TRM curve. A decomposition into two « distinct aging states » as performed by Nordblad *et al.* [3] clearly is possible here. Figure 3b shows the curves for negative  $\Delta T$ , where there is no breaking-up but where the domain growth simply freezes for sufficiently large  $\Delta T$ . For not too large negative  $\Delta T$  there is no freezing but domain growth at a lower rate. Hence the resulting decay curve is still similar to the « normal » TRM curves. All theoretical curves of figure 3 are qualitatively the same as the experimental curves obtained by Refregier *et al.* [4] as well as those of Nordblad *et al.* [3]. We conclude that the theory correctly accounts for this class of experimental data.

## 6. The ac susceptibility after temperature jumps.

A third type of experiment that has been performed by Lundgren *et al.* [6] and in greater detail by Refregier *et al.* [4] consists of observing the behaviour of the zero field ac susceptibility  $\chi(\omega; t)$  after temperature jumps. As discussed in section 2.2, this quantity relaxes towards an equilibrium value. In this section we shall consider the behaviour of the time-dependent ac susceptibility for three cases: i) immediately after the quench to a temperature

$T < T_g$  at  $t = 0$ , ii) after a second temperature quench to  $T - \Delta T$  ( $\Delta T$  positive) at some later time  $\hat{t}_1$ , and iii) after reheating the system to  $T$  at  $\hat{t}_2 > \hat{t}_1$ . This sequence of temperature jumps corresponds to an experiment performed by Refregier *et al.* on  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  [4]. Experimentally one observes a relaxation of the susceptibility after the first two temperature changes, but *not* after the third one. We shall show how our theory predicts a similar time dependence of  $\chi(\omega; t)$ . The amplitude of the ac magnetic field will be taken infinitesimally small.

The analysis of case i), the behaviour of the ac susceptibility immediately after the quench to the temperature  $T$ , makes use of results from subsection 4.1. Only the weight function  $f(T, 0; [s(t'')])$  is required (see Eq. (3.8)) and we describe the system again by a single characteristic domain size  $\bar{s}(t)$ . There is no upper limit on the domain growth so from equation (4.4) we have the response function

$$R(t, t - \tau) = \left(1 + \frac{\tau}{t_0}\right)^{-\alpha} \exp \times \left\{ -\frac{t_2^{pz}}{t_1(1-pz)} (t^{1-pz} - (t - \tau)^{1-pz}) \right\}. \quad (6.1)$$

Substitution of (6.1) into (2.11) and evaluation of the integral for  $t > 0$  results in the first curve shown in figure 4. The values for  $\alpha$ ,  $t_0$ ,  $pz$  and  $t_1 t_2^{-pz}$  are the same as in the sections 4 and 5.

Just like in section 2.2 we can calculate the asymptotic behaviour of this curve for long times. To this purpose we compute the derivative of

$$\frac{\chi(\omega; t)}{\chi_{\text{eq}}} \simeq 1 - (|\omega| t_0)^\alpha \Gamma(1 - \alpha) \left\{ \cos \frac{\alpha \pi}{2} - i \operatorname{sgn} \omega \sin \frac{\alpha \pi}{2} \right\} + c^{-1} t^{-pz} |\omega|^{-\alpha-1} t_0^\alpha \Gamma(2 - \alpha) \left\{ \sin \frac{\alpha \pi}{2} + i \operatorname{sgn} \omega \cos \frac{\alpha \pi}{2} \right\} - i \alpha \omega^{-1} t_0^{-1} \left(1 + \frac{t}{t_0}\right)^{-\alpha-1} e^{i\omega t}. \quad (6.3)$$

In practice the oscillating part is suppressed by averaging over one or more periods  $2\pi/\omega$  and subsequent low-pass filtering. (For the theoretical curves of Fig. 4 this suppression has been achieved by sampling  $\chi''(\omega, t)$  at times  $2\pi/\omega$  apart). Hence we find power law decay towards equilibrium with a power  $pz$ .

In case (ii), after the second temperature quench at time  $\hat{t}_1$  to a temperature  $T - \Delta T$  ( $\Delta T$  positive), the weight function to be considered is  $f(T - \Delta T, 0; [s(t'')])$ . We can describe the system by means of a characteristic domain size  $\bar{s}(t)$ . During the interval  $(0, \hat{t}_1)$  the growth of  $\bar{s}(t)$  is restricted to the overlap length. We will assume that we deal with the case, where the domain growth is actually limited by the overlap length  $\bar{s}(\hat{t}_1) = l_{\Delta T}$ ,

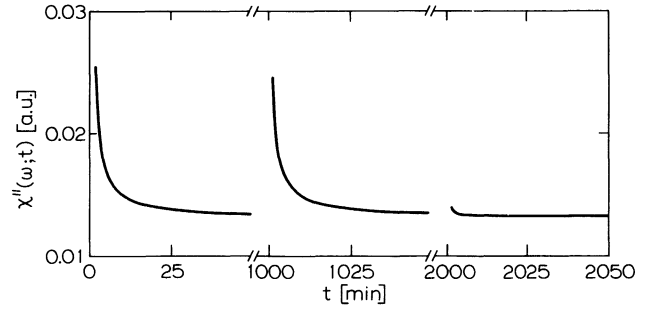


Fig. 4. — Theoretical curve for the imaginary part of the time-dependent ac susceptibility  $\chi''(\omega; t)$ . Shown is the relaxation to equilibrium immediately after the quench to  $T < T_g$  at  $t = 0$ , after a second quench to  $T - \Delta T$  at  $t = 1000$  min and after reheating to  $T$  at  $t = 2000$  min. The equilibrium values of  $\chi''$  at  $T$  and  $T - \Delta T$  have been assumed equal. See the text for the parameter values.

equation (6.1) and expand for large  $t$  and  $\tau$  fixed and find

$$\frac{dR(t, t - \tau)}{d\tau} \simeq -\frac{\alpha}{t_0} \left(1 + \frac{\tau}{t_0}\right)^{-\alpha-1} - \frac{1}{ct^{pz}} \left\{ \left(1 + \frac{\tau}{t_0}\right)^{-\alpha} - \frac{\alpha \tau}{t_0} \left(1 + \frac{\tau}{t_0}\right)^{-\alpha-1} \right\}, \quad (6.2)$$

where  $c = t_1 t_2^{-pz}$ . When (6.2) is substituted in (2.11) the first term gives rise to the equilibrium susceptibility  $\chi(\omega)$  that we calculated in equation (2.16) but due to the finite upper limit in the integral there will be an additional, rapidly decaying, oscillating part. The second term gives rise to a similar integral, so

where we used again the abbreviation  $l_{\Delta T} = l(\Delta T, 0)$ . This is the case for sufficiently large  $\Delta T$ . For times  $t > \hat{t}_1$  the domain growth is unrestricted and we read from equation (3.15):

$$\bar{s}(t) = a \left[ \left(\frac{l_{\Delta T}}{a}\right)^{\frac{1}{p}} + \frac{t - \hat{t}_1}{t_2} \right]^p \quad (t > \hat{t}_2). \quad (6.4)$$

The response function  $R(t, t')$  can be calculated by substituting equation (6.4) into equation (4.2). For sufficiently large  $\Delta T$  the overlap length  $l_{\Delta T}$  is so small that  $\bar{s}(t)$  is only influenced at very short times  $t - \hat{t}_1$ : the domain growth for the  $(T - \Delta T, 0)$ -domains will effectively set in at  $t = \hat{t}_1$ . Therefore the second theoretical curve in figure 4 is very similar to

the first curve for the quench at  $t = 0$ . (The parameters used are the same in Sect. 5.) For simplicity we have assumed  $\chi_{\text{eq}}$  not to depend on temperature.

Finally we will discuss the case iii) where the system is reheated to  $T$  at time  $\hat{t}_2$ . As was the case in section 5, breaking-up of the domains will occur in the interval  $(\hat{t}_1, \hat{t}_2)$  for sufficiently large  $\Delta T$ , and two fractions of spins will emerge after  $\hat{t}_2$ : a « young » fraction and an « old » fraction. As in section 5, the « young » fraction will be very small for  $\Delta T$  large enough and hence we can very well describe the system for  $t > \hat{t}_2$  with one characteristic domain size, which equals  $\bar{s}(\hat{t}_1)$  at  $t = \hat{t}_2$ . From equation (3.15) we then have

$$\bar{s}(t) = a \left[ \frac{\hat{t}_1}{t_2} + \frac{t - \hat{t}_2}{t_2} \right]^p \quad (t > \hat{t}_2). \quad (6.5)$$

The third theoretical curve in figure 4 clearly shows the absence of the relaxation behaviour of the previous curves. Similar curves have been obtained by Refregier *et al.* [4] for their experiments on  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$ . Our results for  $\chi''(\omega; t)$  are furthermore fully consistent with the experimental results of Lundgren *et al.* [6] on Cu (4 at % Mn) obtained during a sequence of temperature increases. A small discrepancy remains, nevertheless, when the relaxation behaviours of  $\chi''$  and  $\chi'$  are compared: Lundgren *et al.* [6] observe that in the nonstationary regime the  $\frac{\pi}{2}$  law (2.17) remains valid, whereas our asymptotic behaviour (6.3) does not satisfy this law.

We conclude this section by pointing out an interesting although only partial analogy. In an Ising ferromagnet in zero field, after a quench to the low-temperature phase, ferromagnetic domains appear whose average size increases with time [23]. Under these circumstances one would expect to find for the frequency dependent *staggered* susceptibility a relaxation behaviour analogous to the first curve in figure 4 for  $\chi''(\omega; t)$ .

## 7. Final remarks.

We wish to discuss the relation of the present study to existing work on spin glass dynamics. Virtually no theoretical work focusses specifically on the aging phenomena in spin glasses. An exception is the paper by Ginzburg [11], who, within a mean field approach, does address aging, starting from the dynamical theory of the spin glass by Sompolinsky and Zippelius [30]. Our work differs from Ginzburg's in that it does not start from a microscopic spin model, but rests on a set of assumptions at the mesoscopic level; it attempts to describe finite-dimensional systems possessing one or more characteristic lengths; it presents a quantitative

comparison with recent experimental findings, in particular by Refregier *et al.* [4] and by Nordblad *et al.* [3]; and it goes beyond the linear response regime.

A great deal of attention has been given to aging in polymers (see, in particular Ref. [31]). Alba *et al.* [25] and Ocio *et al.* [32], in order to fit their magnetic relaxation data in spin glasses, successfully use the same parametrization as employed for polymers. A physical picture underlying this procedure is, however, still lacking.

A theory which, like ours, addresses the spin glass dynamics at the mesoscopic level, is the « fractal cluster model ». This model was introduced and studied recently by Malozemoff and coworkers [20, 33] as well as by Lundgren *et al.* [21]. The fractal cluster model has been used to derive static and dynamic scaling laws in the vicinity of  $T_g$ , but has not been applied to aging. It postulates the existence of « clusters » inside of which the spins are randomly oriented but rigidly coupled together. In response to magnetic field changes such a cluster may be visualized as rotating in a frozen matrix [34]. With each cluster a relaxation time is associated *via* an equation identical to our equation (3.7). In this model the distribution law for the cluster sizes is assumed to be the equilibrium distribution, characterized by a temperature-dependent length. For  $T < T_g$  there is also an infinite cluster [20].

In this work we speak of *domains* of correlated spins rather than of « clusters ». The two concepts cannot be identified. Firstly, each pair  $(T, H)$  has a different domain structure associated with it. Secondly, a domain represents a less rigid and permanent aspect of the spin glass structure than does a cluster: it responds to a change of magnetic field or temperature by starting to grow or to break up. Hence the distribution of the domain sizes is *not* in equilibrium but evolves with time (it can tend towards an equilibrium distribution only when  $t \rightarrow \infty$  at fixed  $T$  and  $H$ ). It is precisely this feature which enabled us to explain the great variety of experimental aging effects.

A description of the nonequilibrium spin glass state as consisting of equilibrium domains separated by domain walls was given also by Kinzel [35]. In this work the domain size increases with time and Kinzel argues that there is a power law relation between the excess energy and magnetization during the equilibration process.

A cluster of spins that can flip between only two configurations is a two-level system. Models of spin glass dynamics involving a distribution of two-level systems and an associated distribution of relaxation times were considered by many authors, e.g. McMillan [22], Préjean and Souletie [36] and Hüser *et al.* [37]. In all these cases the distribution of relaxation times is fixed once for all. In contrast, the present

theory associates with each of the time-dependent distributions  $p(s, t; T_1, H_1)$  (see Sect. 3 and the appendix) a *time-dependent* distribution of relaxation times *via* the relation (3.7) between  $s$  and  $\tau_{\max}(s)$ .

The notion of a time-dependent spectrum to describe spin glass relaxation was used earlier by Lundgren *et al.* [1, 21, 24]. Our work bases this notion on a picture of domains that grow and break up, and makes the evolution equation for the spectrum explicit. Along the way we have needed several assumptions, which however we feel are all secondary. The agreement found with experiment does not prove that all these hypotheses are right; however it means that a simple picture like the one proposed here is capable of explaining the large collection of experimental data.

#### Acknowledgements.

The authors would like to thank J. Hammann, M. Ocio, Ph. Refregier, and E. Vincent for com-

municating to them experimental results prior to publication, as well as for many stimulating discussions. They have furthermore benefitted from discussions with P. Granberg and R. Orbach, and are pleased to acknowledge conversations with C. De Dominicis, B. Derrida, J. A. Mydosh and G. J. Nieuwenhuys.

Part of this research was supported by the *Stichting voor Fundamenteel Onderzoek der Materie (FOM)*, which is financially supported by the *Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek (ZWO)*.

#### Appendix.

In section 3.1 we have introduced the distribution  $p(s, t; T_1, H_1)$  of the  $(T_1, H_1)$ -domain sizes at time  $t$ . Although this distribution has not played an explicit role in the subsequent calculations, it is nevertheless interesting to remark that it satisfies the master equation

$$\begin{aligned} \frac{\partial}{\partial t} p(s, t; T_1, H_1) = & - \frac{\partial}{\partial s} [\lambda(s; T(t), T_1) p(s, t; T_1, H_1)] - \\ & - \tau_b^{-1} \theta(s - l(T(t) - T_1, H(t) - H_1)) p(s, t; T_1, H_1) \\ & + \tau_b^{-1} \delta(s - l(T(t) - T_1, H(t) - H_1)) \int_{l(T(t) - T_1, H(t) - H_1)}^{\infty} ds' p(s', t; T_1, H_1), \end{aligned} \quad (\text{A.1})$$

in which  $\lambda$  is the growth rate of a domain of size  $s$  at temperature  $T$ , which, according to equation (3.13), equals

$$\lambda(s; T, T_1) = \frac{p}{t_2(T, T_1)} a^{\frac{1}{p}} s^{\frac{p-1}{p}}. \quad (\text{A.2})$$

The time constant  $\tau_b$  is defined by equation (3.16). One easily checks that equation (A.1) is compatible with the normalization

$$\int_0^{\infty} ds p(s, t; T, H) = 1. \quad (\text{A.3})$$

According to (iv) in section 3.1, if at time  $t = 0$  the system is quenched to below  $T_g$ , we have that the initial condition is a delta peak at  $s = 0$ , regardless of  $(T, H)$ :

$$p(s, 0; T, H) = \delta(s). \quad (\text{A.4})$$

It is clear that in experiments in which after the quench only a finite number (say  $n$ ) of temperature or magnetic field jumps are applied, the function  $p(s, t; T, H)$  can be the sum of at most  $n$  delta peaks.

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