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Two-level-systems in spin glasses : a dynamical study of the magnetizations below T_G , application to CuMn systems

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Résumé. — Un modèle de distributions d'états à deux niveaux thermiquement activés qui s'impose sur la base des relaxations de l'aimantation et de l'énergie qu'on observe expérimentalement, permet sans autre hypothèse de prédire ou de justifier l'essentiel des phénomènes d'hystérésis dans les verres de spin. On distingue les processus où la température est variée à champ fixe qui conduisent à un état d'équilibre pratique du système et les processus isothermes qui conduisent à une situation hors équilibre et dont la description met en jeu des lois classiques (lois de Rayleigh) en fonction d'une variable réduite T/T_G où T_G comme toute température caractéristique du problème est une fonction du temps t de mesure $\left(T_G \sim \left(\text{Ln} \frac{t}{\tau_0}\right)^{-1}\right)$. Des considérations énergétiques fixent une limite supérieure T_c au domaine où ce *scaling* caractéristique d'une transition vitreuse est valable, ce qui permet de concilier les résultats apparemment contradictoires qui font état d'un maximum de la susceptibilité dépendant de la fréquence dans certains systèmes, indépendant de la fréquence dans d'autres. C'est l'existence de distributions bien caractérisées des états à deux niveaux qui est importante plutôt que la description microscopique des objets eux-mêmes cachés derrière le concept d'états à deux niveaux. C'est le désordre qui impose leur caractère aux distributions d'où la généralité du modèle.

Abstract. — A model of distributions of thermally activated two-level-systems (T.L.S.) which imposes itself on the basis of the observed relaxations of the magnetization and of the energy, allows, without further assumptions, to predict or to justify the main features of the hysteresis of spin glasses. We discern the processes where the temperature is varied at constant field which lead to a *quasi equilibrium* from the isothermal processes which lead to a non-equilibrium situation whose description involves classical laws (Rayleigh's laws) in terms of a reduced temperature T/T_G . T_G as any characteristic temperature in the problem is a function of the time t of the measurement : $T_G \sim \left(\text{Ln} \frac{t}{\tau_0}\right)^{-1}$. Energetic considerations fix a higher limit T_c to the domain where this *scaling*, characteristic of a glass transition, is valid, which permits to reconcile the apparently contradictory data which report a frequency dependent susceptibility maximum in some systems, a frequency independent one in others. It is the existence of well characterized distributions of T.L.S. which is important rather than the microscopical description of the objects themselves which are hidden behind the T.L.S. concept. This is the disorder which is responsible for the character of the distributions hence for the generality of the model.

1. **Introduction.** — The study of systems of magnetic impurities diluted in noble metal matrices has been a traditional subject of interest in magnetism. The *spin glass* problem deals with the study of the magnetic ordering occurring at low temperatures between randomly distributed magnetic moments interacting through the Rudermann-Kittel-Kasuya-Yoshida interaction. The discovery [1] of a sharp anomaly (the cusp) in the low field susceptibility at a temperature T_G has raised the question of the possibility of a new type of phase transition [2] and has

motivated theoretical interest [3]. At temperatures lower than T_G , the existence of time dependent effects in the magnetic properties has been known for a long time [4] but, although phenomenological descriptions exist [5], difficulties remain at several levels. In particular the very presence of unusually long time scales makes problematic the experimental determination of a fundamental state. Further, it appears difficult, at the present time, to reconcile the phenomenological concepts with the pathological variations of the susceptibility at T_G which are suggestive of a phase transition. From a mere experimental point of view contradictions appear :

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thus one finds [6, 7] or does not find [8] a time dependence of T_G depending on the system which is studied or even on the thermal treatment which the system has experienced [9].

In this paper we will try to clarify some notions and suppress some contradictions by recalling and developing phenomenological arguments of general interest to disordered system with time dependent properties. We will first justify the use of a model involving a distribution of thermally activated asymmetrical double well potentials, then derive the consequences and illustrate these consequences with the results of experiments performed mainly on the CuMn system. We will show that it is possible to reach, *in practice*, the fundamental state by field cooling the sample, although any modification of the field at low temperature brings the system out of equilibrium. Non linearities in field and time dependences of the magnetization as well as their consequences on the reversible susceptibility will also be discussed. We will afterwards speculate about the origin and the nature of the objects hidden behind the abstraction of the double well potential picture and show that it is possible to reconcile the proposed phenomenological description with pathological behaviour occurring at a fixed temperature.

2. Justification of a model of double well potentials.

— Remanent magnetization measurements [4, 5] were the first which showed the time dependent effects characteristic of the spin glass state. The observation of relaxation of the magnetization after several hours (or several days) implies the existence of unusually large potential barriers which delay the response of some kind of magnetic objects of moment M_g . The non exponential character of these relaxations (apparent for example on figure 4) implies, moreover, a broad distribution $P(W)$ of the values W of the energy of these barriers. This feature is not unexpected in a disordered system where no particular value of W is anticipated. We will therefore assume a distribution $P(W)$ with little or no dependence on W over a considerable range of energies limited to some value W_{\max} .

Recent measurements [10, 11] have established that an energy relaxation is always associated with the magnetization relaxations. This fact, among others, implies that the barriers W separate levels of different energies. Let ε_0 be the energy splitting. It is logical to postulate, in a disordered system, the existence of a distribution $P(\varepsilon_0)$ of the zero field splittings for the same reasons which justify the existence of a distribution $P(W)$ i.e. the absence of any privileged value of the energy. One is then logically led to the concept of a distribution of asymmetrical double well potentials (Fig. 1) for similar reasons to those that led to the models which have been proposed to explain the properties of glasses at very low temperatures [12].

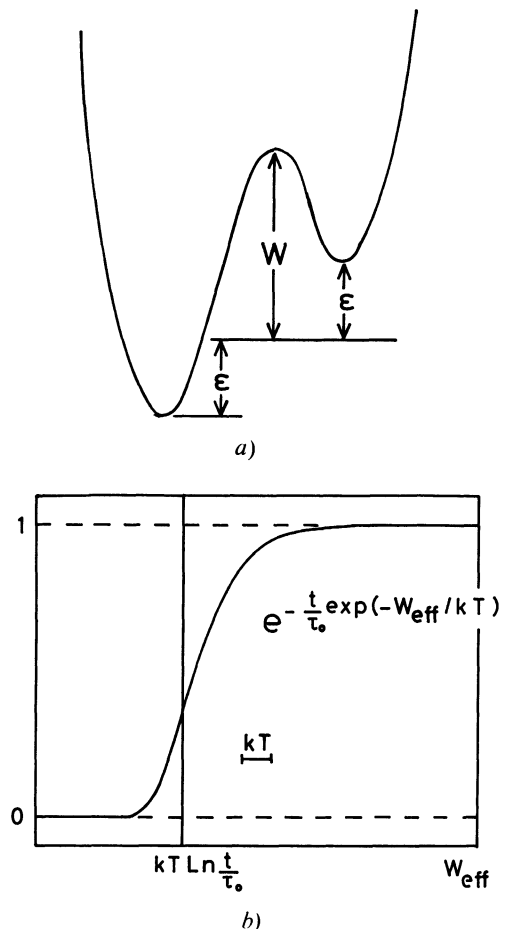


Fig. 1. — An asymmetrical double well potential is represented on figure 1a : W is the height of the barrier, ε represents the splitting. The effective barriers for the system are $W_{\text{eff}} = W \pm \varepsilon$. Figure 1b shows the variations of the relaxation

$$\exp\left(-\frac{t}{\tau}\right) = \exp\left(-\frac{t}{\tau_0} \exp\left(-\frac{W_{\text{eff}}}{kT}\right)\right)$$

vs. W_{eff} in an activated process. $W_c = kT \text{Ln} \frac{t}{\tau_0}$ is, with a good approximation, a cut-off in our problem (i.e.

$$\exp\left(-\frac{t}{\tau}\right) = \begin{cases} 0 & \text{for } W_{\text{eff}} < W_c \\ 1 & \text{for } W_{\text{eff}} > W_c \end{cases}$$

if $W_c \gg 2$ or $3kT$ (the width of the transition). In CuMn, the order of magnitude of W_c is $25kT$ in the usual time scale measurement.

A physical barrier can be crossed either by tunnelling or by thermal activation. While the first process is important in the models considered for the low temperature properties of glasses, it is easy to show experimentally that thermal activation is the phenomenon which governs the relaxations of a spin glass. The relaxation time in this case is given by the Arrhenius law $\tau = \tau_0 \exp\left(\frac{W - \varepsilon_0}{kT}\right)$. The remanent magnetization of objects of moment M_g involves an integration over the values of W and ε_0 . Formally we will have to integrate relaxations of the form $M_g \exp\left(-\frac{t}{\tau}\right)$ over the distributions of W and ε_0 .

The consideration of the variations of

$$\exp - \frac{t}{\tau_0 \exp \frac{W - \varepsilon_0}{kT}}$$

in terms of W shows that in practice and to a very good approximation (see Fig. 1b) :

$$\exp - \frac{t}{\tau_0 \exp \frac{W - \varepsilon_0}{kT}} \begin{cases} = 0 & \text{for } W - \varepsilon_0 < W_c \\ = 1 & \text{for } W - \varepsilon_0 > W_c \end{cases} \quad (1)$$

(where $W_c = kT \text{Ln} \frac{t}{\tau_0}$) and it is therefore possible to put $\exp - \frac{t}{\tau} = 1$ in the integration provided it is restricted to the values $W > W_c + \varepsilon_0$. Inasmuch as $P(\varepsilon_0)$ and $P(W)$ do not depend strongly on the temperature the remanent magnetization (as any other thermodynamic quantity) will be obtained after integration over W and ε_0 , as a function of the cut-off $W_c = kT \text{Ln} \frac{t}{\tau_0}$. The measurement time t

therefore is always associated with the temperature in such a way that it is possible to observe the same effects in terms of the temperature at fixed time or in terms of $\log t$ at fixed temperature. This time-temperature correspondance is characteristic of the Arrhenius law and of an activated process. One can check (Figs. 18 and 19) that it was possible to superimpose the temperature and the time dependences of the remanent magnetization of a CuMn 8 at. % in a unique diagram of the variable $kT \text{Ln} \frac{t}{\tau_0}$ (despite the fact that the observed variations which cover several decades in magnitude depart strongly from a simple linearity in T or $\text{Ln} \frac{t}{\tau_0}$). This correspondance allows the characteristic time τ_0 to be determined. We have found τ_0 to be of the order of 10^{-11} s in CuMn for concentrations between 1 and 8 at. % (see chapter 12).

We are now in possession of a model which allows us to make non trivial predictions (such as the above mentioned time-temperature correspondance). This model is by no means new. In the abstract shape in which it is presented, it covers descriptions which have been widely used to describe for example the plasticity of glasses, rubbers and polymers [13] near their glass transition or the magnetism of fine particles (Néel). This generality implies analogies, which can be checked by experiments between properties *a priori* as different as the magnetization of a spin glass and the extension of a rubber [14]. We will make use in what follows of the ideas and vocabulary which were developed by Néel [15], in his description of the properties of small particles (where $\varepsilon_0 = 0$: the double well potentials are symmetrical) or in

his theory of magnetism in the Rayleigh domain (in this case the model makes use of two independent random variables and can consequently be identified with the one we use here).

3. Isothermal processes and field cooling processes.

The (quasi) equilibrium state of the systems. — With the time entering as a third variable the specification of the field and the temperature is not sufficient to determine the magnetization. The hysteresis cycle exemplifies this indeterminacy. The evolution of the magnetization within the hysteresis cycle will depend not only on the time t which is spent on the measurement but also on the thermal path $C(H(t), T(t))$ through which the chosen initial state has been built up.

In other words the future behaviour of the sample is determined by its history. It is in principle always possible to express a thermal path as the sum of elementary paths where the magnetic field is modified at a constant temperature and where the temperature is modified at constant field ; such paths, because of their simplicity, are chosen by the experimentalist. It is therefore logical to study the isothermal response to a varying field and the response to a constant field when the temperature is varied.

In an isothermal process, the argument leading to equation (1) tells us (see Fig. 1) that a field h such that $M_g h + \varepsilon_0 > W - W_c$ is needed to overcome a barrier W (with $W_c = kT \text{Ln} \frac{t}{\tau_0}$). This means that at low temperature all the energy of the potential barrier must be supplied by the field.

In contrast, in a field cooling process, the magnetization is *blocked* at a temperature T_B such that

$$W + \varepsilon_0 - M_g h = kT_B \text{Ln} \frac{t}{\tau_0}.$$

The magnetic state of the object (the state in which it remains when the temperature is further decreased) is then determined by $th \left(\frac{M_g h - \varepsilon_0}{kT_B} \right)$. The ratio

$$\frac{M_g h - \varepsilon_0}{kT_B} = \frac{(M_g h - \varepsilon_0) \times \text{Ln} \frac{t}{\tau_0}}{W - M_g h + \varepsilon_0}$$

is larger than 1 (and $th \frac{M_g h - \varepsilon_0}{kT_B}$ is saturated) as soon as $\varepsilon = M_g h - \varepsilon_0$ is of the order of or larger than W/Q (where $Q = \text{Ln} \frac{t}{\tau_0}$, Q is of the order of 25 for usual values of t and τ_0) : the system has already chosen its equilibrium state (the lower side of the two-level-systems) at the blocking temperature. In other words we only need to provide an energy $M_g h - \varepsilon_0 = W/25$ to overcome a barrier W in a field cooling process (instead of W at low T). With

our assumptions on the distributions $P(\varepsilon_0)$ and $P(W)$ therefore, the field cooled magnetization $M_a(H, T)$ cannot differ from the thermodynamic equilibrium curve by more than 4% ($1/Q$) of the maximum deviation which can be obtained in the same field at temperature T (that is 4% of the actual width of the hysteresis cycle at temperature T in field H). Neglecting this difference we can define $M_a(H, T)$ as a *practical* determination of the fundamental state.

4. The field cooled magnetization as the equilibrium magnetization. Experimental evidence. — Figure 2 shows the magnetization curves obtained for decreasing fields (starting from 75 kOe) at different temperatures in CuMn 8 at.%. At temperatures below $T_G = 40$ K we observe hysteresis. At very

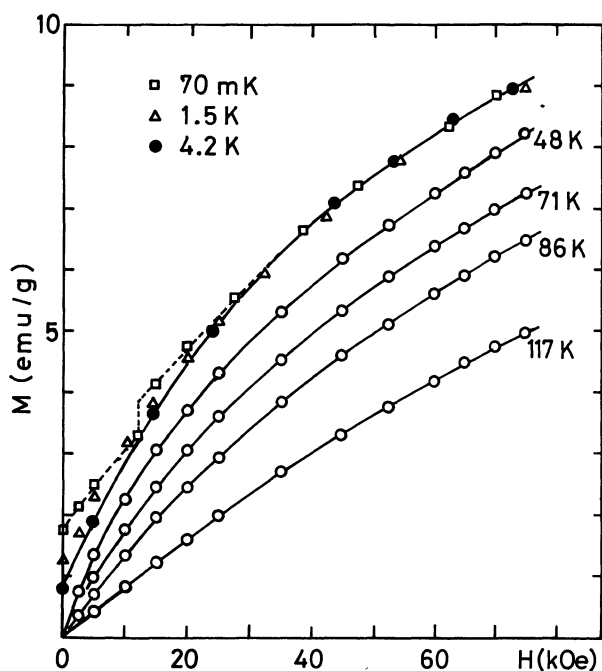


Fig. 2. — The magnetization of a CuMn 8 at. % vs. the field H is shown at different temperatures. For $T \geq T_G$ (40 K), the magnetization does not present any hysteresis. The magnetization curves represented for $T < T_G$ were obtained after cooling the sample in 75 kOe from $T > T_G$ down to $T = 4.2$ K (\bullet), 1.5 K (Δ) and 70 mK (\square) and decreasing the field.

low temperatures we also observe in positive fields the existence of *magnetization jumps* as reported by Tournier in AuFe [4]. These processes reminiscent of Barkhausen jumps in ferromagnetics, by which part of the remanent magnetization is globally reversed in the magnetic field, call for an interpretation different from the one which we propose here. Far from the jump, the general behaviour tends to recover the slower variations that our model attempts to describe, and obeys rules of general validity for all spin glasses.

Once a remanent magnetization has been built up the response of this magnetization to a small

transverse field or to an inverse field is very system dependent. In CuMn a very small inverse field H_r is sufficient to reverse completely the remanent magnetization as a whole. Very small quantities (a fraction of one at. %) of non magnetic impurities known to have strong spin orbit coupling are sufficient to increase very strongly the value of H_r to the point of making the magnetization reversal vanish completely, although the values of the magnetization are not greatly modified. This macroscopic response of the remanent magnetization is studied in detail in another work [16]. Here we are concerned only with the initial development and the evolution of the magnetization along the axis of the field (which has been used to build it up).

The field cooled magnetization (Fig. 3) exhibits the behaviour expected for an equilibrium magnetization : it is single valued and stable. When the system is pushed out of balance it tends to relax

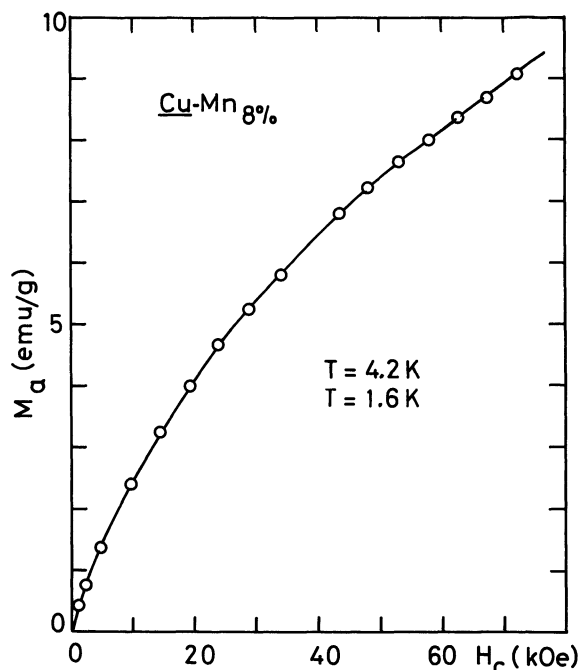


Fig. 3. — The field cooled magnetization $M_a(H_c)$ is shown vs. H_c at 1.6 K and 4.2 K. In a field cooling process, the field H_c is applied at $T > T_G$ and the temperature is subsequently decreased. $M_a(H)$ is time independent as long as H_c stays applied. In CuMn , $M_a(H)$ is not very sensitive to the temperature.

towards the equilibrium curve with liberation of energy [10, 11]. These properties are illustrated on figure 4 which shows $M_a(h, T = 20$ K) for a CuMn 8 at. %. Each point has been obtained by field cooling the sample from $T \geq 40$ K (T_G). $M_a(H, T)$ has a normal behaviour. It goes to zero in zero field : it can be developed in terms of odd powers of the field and remains time independent as long as the field stays applied. The application, at low temperatures, of a field variation ΔH (however small it may be) has the effect of creating a non equilibrium magnetization which always evolves as a function.

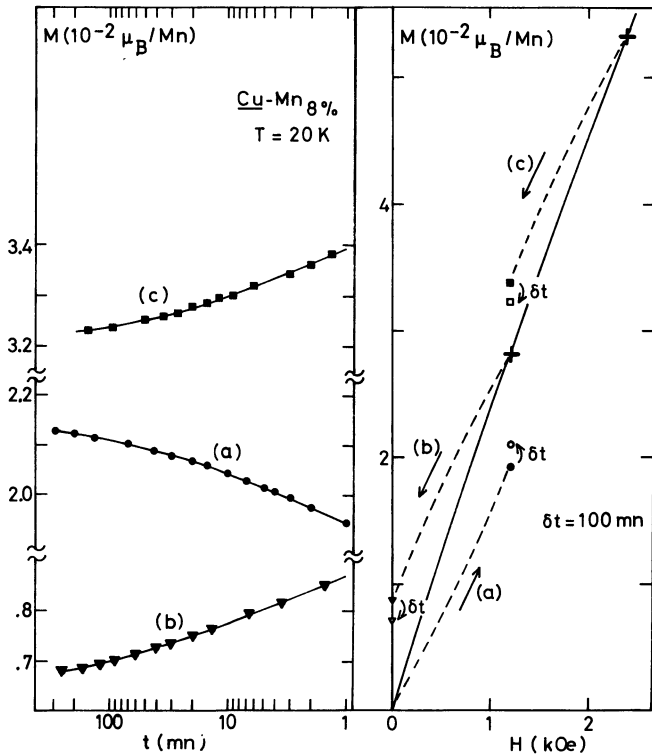


Fig. 4. — Starting from $M_a(H_c)$ (solid line), small isothermal variations ΔH of the field induce magnetization relaxations in the direction of $M_a(H_c + \Delta H)$: (a) $H_c = 0$, $\Delta H = 1.2$ kOe, (b) $H_c = 1.2$ kOe, $\Delta H = -1.2$ kOe, (c) $H_c = 2.4$ kOe, $\Delta H = -1.2$ kOe. The corresponding pseudologarithmic relaxations are shown vs. time on the left side of the figure (the origin of time is taken when the field reaches final value $H_c + \Delta H$).

of time towards the magnetization $M_a(H + \Delta H)$ which represents in principle the equilibrium in the new field. Notice in figure 3 the pseudologarithmic dependences which cannot be explained with a unique relaxation time. We have checked elsewhere [11] that these magnetization relaxations are always associated with an *energy relaxation* from the sample to the bath, irrespective of the fact that the magnetization may increase or decrease.

It seems therefore that there exists for each field an equilibrium corresponding to a fundamental state of minimum energy towards which the system tends to relax at long times. This equilibrium corresponds with a good accuracy to the situation which is reached by field cooling the sample. This, incidental-

ly, is consistent with the fact that no time dependent anomaly has been so far observed in specific heat measurements [17], since in this purely thermal procedure the system is always at equilibrium. It implies conversely that such time effects should be present if a field is applied or removed at low temperatures. Let us make it clear however that effects associated with a purely thermal treatment should become apparent either at a higher order of sensitivity or if the coefficient $Q = \text{Ln} \frac{t}{\tau_0}$ could be made much smaller than the usual value of 25. The practical interest of the curve $M_a(H)$ is evident since the knowledge of the response at infinite times amounts to the knowledge of the distribution $P(\epsilon_0)$. From now on, we will prefer to deal with the actual experimental curve $M_a(h)$ using only the additional hypothesis of a distribution $P(W)$ of barrier heights which determines the evolution of the system in time.

It can be seen in figure 3 that $M_a(H, T < T_G)$ in CuMn 8 at. % depends very little on the temperature. This behaviour which is shared with most of the archetypical spin glasses (AuFe, AuCr, AuMn, AgMn, ...) at low enough concentrations is, however, not general. A rather strong (but always monotonic) temperature dependence is found below T_G in systems such as PtCo [18], where a strong exchange enhancement of the lattice is known to exist; precipitations associated with a tendency to ferromagnetism may also be responsible for a similar behaviour.

Starting now from a point on the equilibrium curve $M_a(H_0, T)$ we will consider the isothermal response to the first application of a field variation ΔH_F , followed by a decrease of ΔH_R opposite and smaller than any previous variation, using the data of $M_a(H)$ at equilibrium and assuming a distribution $P(W)$ of barriers to fix the time dependences. The definitions which we introduce here are important since the model implies different responses to the first application and to a subsequent decrease [15]. (The first application is performed on a system at equilibrium.)

5. Isothermal application ΔH_F and decrease ΔH_R of a field. — Starting from $M_a(H_0)$ the response to the first application of a field variation ΔH_F followed by a decrease ΔH_R would be at infinite times :

$$M(H_0 + \Delta H_F - \Delta H_R) = M_a(H_0) + \int_{H_0}^{H_0 + \Delta H_F - \Delta H_R} M'_a(H) dH = M_a(H_0 + \Delta H_F - \Delta H_R). \quad (2)$$

At finite times we would have :

$$M(H_0 + \Delta H_F - \Delta H_R) - M_a(H_0) = \int_0^{W_c} P(W) dW \int_{H_0}^{H_0 + \Delta H_F - \Delta H_R} M'_a(H) dH + \int_{W_c}^{W_c + \bar{M}_s \Delta H_F} P(W) dW \times \int_{H_0}^{H_0 + \Delta H_F - \frac{W - W_c}{\bar{M}_s}} M'_a(H) dH + \int_{W_c}^{W_c + \frac{\bar{M}_s \Delta H_R}{2}} P(W) dW \int_{H_0 + \Delta H_F - \frac{W - W_c}{\bar{M}_s}}^{H_0 + \Delta H_F - \left(\Delta H_R - \frac{W - W_c}{\bar{M}_s}\right)} M'_a(H) dH \quad (3)$$

(where we have assumed that we can use some average value \bar{M}_g for the moments of the objects). The first term is the response of those objects which are at thermodynamic equilibrium, W being smaller than W_c . The second and the third terms account for the responses to the increase ΔH_F and to the decrease ΔH_R respectively, of those objects for which the effective barrier $W - W_c$ is overcome by the field variation. If the contribution \bar{M}_g was obtained at dH with $W - W_c = 0$, we would have to provide an extra field $\frac{W - W_c}{\bar{M}_g}$ in the direction of the field varia-

tion in order to overcome the energy of the barrier (Fig. 5). Thus we have to add the contributions of all W such that $\bar{M}_g dH + (W - W_c) < \bar{M}_g \Delta H_F$ for ΔH_F and subtract for ΔH_R ($|\Delta H_R| < \Delta H_F$) that of all objects $\bar{M}_g dH - (W - W_c) > \bar{M}_g (\Delta H_F - \Delta H_R)$. Those two relations fix the integration limits on dH and W in equation (3).

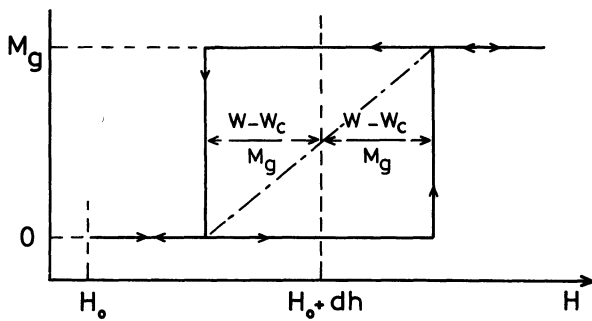


Fig. 5. — Schematic response of a double well potential to an external field. $(W - W_c)$ is the value of the effective barrier (at temperature T) which must be overcome by the applied field. The dashed dotted line shows the response to a field transverse to the anisotropy axis.

In the so-called *Rayleigh domain* i.e. for excursions $\bar{M}_g \Delta H_F \ll W_{\max} - W_c$, we obtain, assuming we can linearize the variation of $M_a(H)$ in the vicinity of $M_a(H_0)$,

$$M(H_0 + \Delta H_F - \Delta H_R) - M_a(H_0) \simeq A(\Delta H_F - \Delta H_R) + B(\Delta H_F)^2 - \frac{B}{2}(\Delta H_R)^2. \quad (4)$$

$$M(H_0 + \Delta H) = M_a(H_0 + \Delta H) - \int_{W_c}^{\infty} P(W) dW \int_{H_0 + \Delta H - \frac{W - W_c}{\bar{M}_g}}^{H_0 + \Delta H} M_a'(H) dH \simeq M_a \left(H_0 + \Delta H - \int_{W_c}^{\infty} P(W) \left(\frac{W - W_c}{\bar{M}_g} \right) dW \right). \quad (7)$$

The reversible susceptibility A is proportional to $T \text{Ln} \frac{t}{\tau_0}$:

$$A = M_a'(H_0) P(0) kT \text{Ln} \frac{t}{\tau_0} \quad (5)$$

in the low temperature limit. The coefficient B of the quadratic terms is independent of $T \text{Ln} \frac{t}{\tau_0}$ in the same limit

$$B = M_a'(H_0) P(0) / 2 \bar{M}_g. \quad (6)$$

Notice the classical results [15] known as Rayleigh's laws: the response is quadratic in the field variation (1st law); the coefficient of the quadratic term is twice as large during the first application of the field than in any subsequent decrease (2nd law). An illustration of this behaviour is shown in the figure 6.

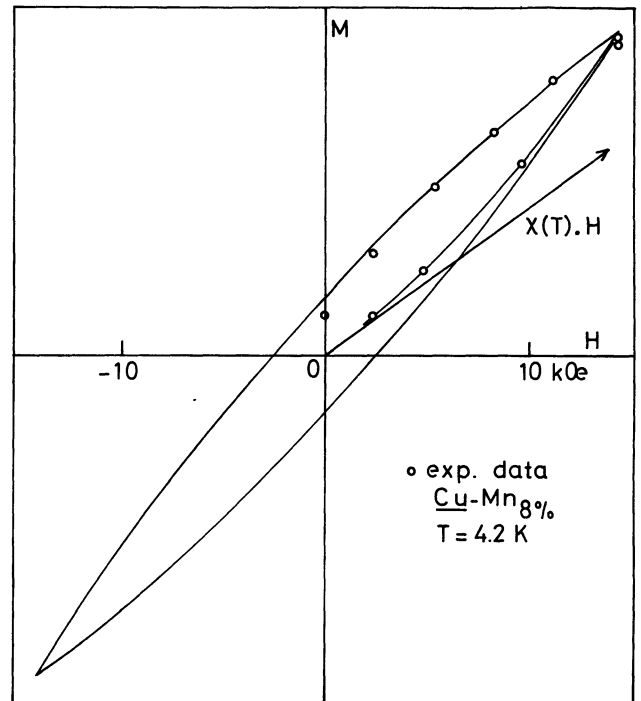


Fig. 6. — The magnetization of a $\overline{\text{CuMn}}$ 8 at. % in the Rayleigh domain at 4.2 K. The deviations to an ideal behaviour (solid line) noticed in zero field are accounted for by the relaxation of the magnetization during the time necessary to increase and decrease the field.

Out of the Rayleigh domain, when $\bar{M}_g \Delta H_F$ becomes larger than the maximum height $W_{\max} - W_c$ of the effective barriers the integration has to be extended over the whole distribution $P(W)$. We have then:

The magnetization tends therefore towards a limiting curve which is obtained by translating the equilibrium curve along the field axis by the quantity :

$$\delta H = \frac{1}{M_g} \int_{W_c}^{\infty} P(W) (W - W_c) dW. \quad (8)$$

6. Case of a constant density of energy barriers. — Let us assume

$$P(W) = P(0) = \frac{1}{W_{\max}} \quad \text{for } W < W_{\max}$$

$$= 0 \quad \text{for } W > W_{\max}.$$

This simplification allows us to obtain expressions which, despite the crudeness of the model, give a fairly good qualitative description of the phenomena.

Defining :

$$W_{\max} = \frac{1}{P(0)} = kT_G \text{Ln} \frac{t}{\tau_0} = \bar{M}_g H_g \quad (9)$$

and using reduced units $\theta = T/T_G$ and $\eta = H/H_g$ we have :

a) In the Rayleigh domain i.e. for $\eta < 1 - \theta$

$$\Delta M(\eta_F - \eta_R) = m'_a \left\{ (\eta_F - \eta_R) \theta + \frac{\eta_F^2}{2} - \frac{\eta_R^2}{4} \right\} \quad (10)$$

if η_F corresponds to the initial increase and η_R to the subsequent decrease of the field and where we have introduced

$$m'_a = \frac{dM_a(\eta)}{d\eta} = H_g M'_a(H).$$

b) Beyond the Rayleigh domain (i.e. for $\eta > 1 - \theta$) from (8) the limiting curve is obtained by a translation $(1 - \theta)^2/2$ of the equilibrium curve

$$M_a(\eta) \rightarrow M_a \left(\eta \pm \frac{(1 - \theta)^2}{2} \right). \quad (11)$$

These two expressions (10) and (11) allow a simple description of the behaviour of the magnetization below the glass temperature T_G of the system.

7. Description of the magnetization. — The equilibrium curve $M_a(\eta)$ is given by experiment.

— The limiting hysteresis cycle $M_a \left(\eta \pm \frac{(1 - \theta)^2}{2} \right)$ is obtained by translating $M_a(\eta)$ by $\pm \frac{(1 - \theta)^2}{2}$ along the field axis (Fig. 7).

— By cooling the sample in zero field we start from an equilibrium point with zero magnetization. The first application η_F of a field then allows, starting from equilibrium, the ascending branch of the limit-

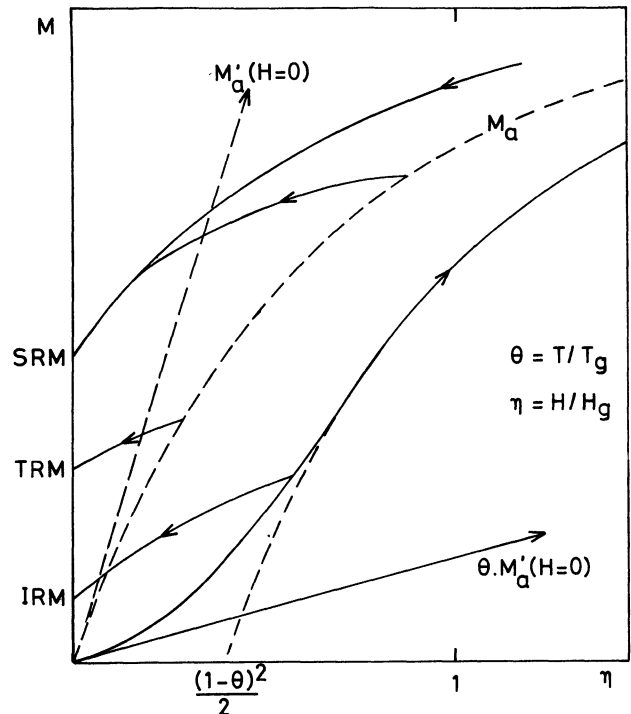


Fig. 7. — Schematic representation of the predicted behaviour of the different magnetizations below T_G . The limiting hysteresis cycle is obtained by translating the equilibrium curve $M_a(\eta)$ by $\pm \frac{1}{2}(1 - \theta)^2$ along the field axis. Starting from $M_a(\eta)$, a field variation opposite and equal to the cooling field η yields the *T.R.M.* The first magnetization curve is obtained by a field variation η , starting from $M_a(0)$. The *I.R.M.* is the result of a field decrease starting from the first magnetization curve. Notice the presence of a reversible susceptibility proportional to θ .

ing hysteresis cycle to be reached, for $\eta \geq 1 - \theta$, through a Rayleigh parabola $m'_a \eta_F \left(\theta + \frac{\eta_F}{2} \right)$. One thus describes the first magnetization curve. Notice the presence of a reversible thermodynamic initial susceptibility $m'_a \theta$ (proportional to $T \text{Ln} \frac{t}{\tau_0}$).

Starting now from the ascending branch, the descending branch of the limiting cycle can be reached through a Rayleigh parabola $m'_a \eta_R \left(\theta + \frac{\eta_R}{4} \right)$ (for $|\eta_R| \leq 2(1 - \theta)$) by decreasing the field.

The agreement with experiment is discussed in chapter 11. There are two features which the model fails to describe.

1. It does not account for the presence of a large reversible susceptibility at $T = 0$.
2. Experimentally the ascending and descending branches of the hysteresis cycle join in large fields. They can not be deduced from each other by a simple translation. We will discuss later the natural interpretations that one can make of these discrepancies.

8. The remanent magnetization. — Particular interest is attached to the remanent magnetization obtain-

ed after the applied field has been removed, because it inevitably affects the results of measurements in low fields (and the susceptibility).

— The thermoremanent magnetization, *T.R.M.* is obtained by cooling in a magnetic field and removing the field at low temperatures. It is therefore (on our definitions) the result of a first application η_F equal and opposite to the field in which the sample has been cooled.

— The isothermal remanent magnetization (*I.R.M.*) is obtained by a retrogression from the first magnetization curve. Both the *T.R.M.* and the *I.R.M.* saturate to the value *S.R.M.* where the descending branch of the limiting hysteresis cycle cuts the magnetization axis (Fig. 7).

Inasmuch, therefore, as one can neglect the curvatures of $M_a(\eta)$ over the range $0 < \eta < 2(1 - \theta)$ one obtains easily from equations (10) and (11) :

$$S.R.M. = m'_a \frac{(1 - \theta)^2}{2} \tag{12}$$

$$T.R.M. = m'_a \eta \left(1 - \theta - \frac{\eta}{2} \right) \tag{13}$$

= *S.R.M.*

$$I.R.M. = m'_a \frac{\eta^2}{4} \tag{14}$$

= *S.R.M.* - $m'_a \frac{(2(1 - \theta) - \eta)^2}{4}$

= *S.R.M.*

for $\eta \leq 1 - \theta$

for $\eta \geq 1 - \theta$

for $\eta \leq 1 - \theta$

for $1 - \theta \leq \eta \leq 2 - \theta$

for $\eta \geq 2 - \theta$.

The comparison of figure 8 with figures 15 and 19 shows that those predictions are, considering the roughness of the assumptions, in good agreement with the experimental results. In particular it is found, in agreement with experiment, that the remanent magnetizations are more easily saturated the higher the temperature. For a given field $\eta \ll 1$ there always exists a regime in the vicinity of T_G where $\theta + \eta > 1$ and the *T.R.M.* is saturated. Below this regime the *T.R.M.* is a linear function of the temperature in agreement with the experimental results (Fig. 8b).

Notice however that the model does not account for the occurrence of a maximum in the dependence of the *T.R.M.* on the magnetic field η (see Fig. 15 and previous data from Tholence [4]).

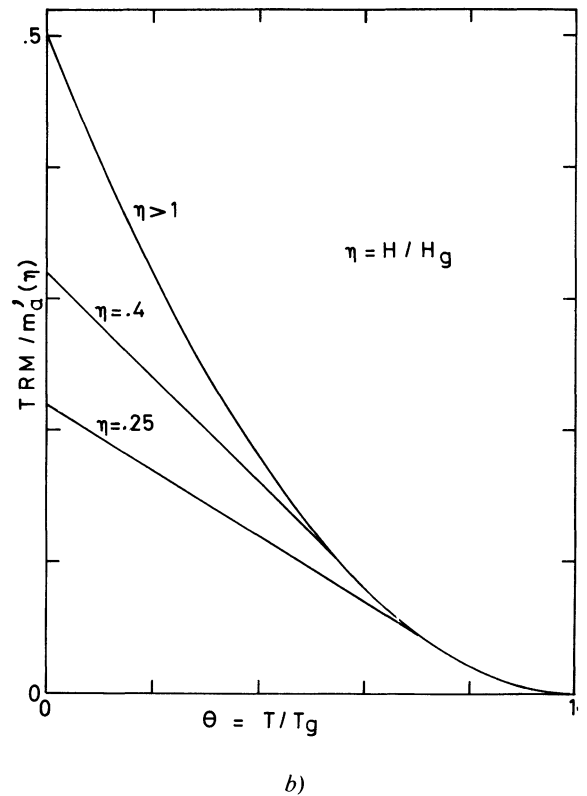
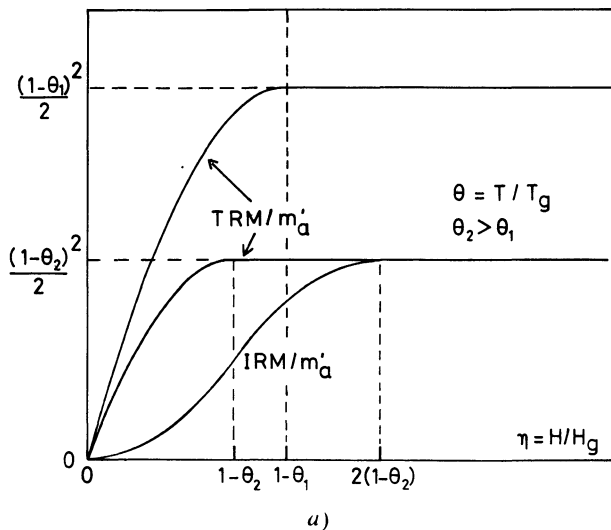


Fig. 8. — (a) The predicted variation of the *T.R.M.* and the *I.R.M.* in terms of the reduced field η at fixed values of the reduced temperature θ . (b) The predicted variations of the *T.R.M.* vs. θ for different values of the reduced field η . For $\eta > 1$, the *T.R.M.* is saturated and follows a parabola $\left(S.R.M. = M'_a \frac{(1 - \theta)^2}{2} \right)$. For $\eta < 1$, the *T.R.M.* is linear in θ for $\theta < 1 - \eta$ and reaches the saturated regime as soon as $\theta > 1 - \eta$.

9. **Susceptibilities.** — By cooling the sample in a small field η we obtain $M_a(\eta) \simeq m'_a \eta$. When the field η is withdrawn at low temperature we are left with the *T.R.M.* in the field η . *By definition of the low temperature reversible susceptibility χ_{rev} we have therefore :*

$$\chi_{rev} \eta = m'_a \eta - T.R.M. = m'_a \eta \left(1 - \frac{T.R.M.(\eta)}{M_a(\eta)} \right) \tag{15}$$

from which we get

$$\begin{aligned} \chi_{rev} &\simeq m'_a \left(\theta + \frac{\eta}{2} \right) \quad \text{for } \theta \leq 1 - \eta \\ &= m'_a \left(1 - \frac{(1 - \theta)^2}{2\eta} \right) \quad \text{for } 1 - \eta \leq \theta \leq 1. \end{aligned} \tag{16}$$

One would obtain the same result by defining $\chi_{rev} \eta$ as the first magnetization decreased from the *I.R.M.* in the same field. This result is made of two parts. $m'_a \theta$ is the thermodynamic contribution due to those barriers $W < W_c$ which are at thermal equilibrium at temperature T in the time of the measurement. The other contribution, which is anomalous, is due to those barriers $W > W_c$ over which the applied field can induce transitions. This magnetization, initially proportional to η^2 , is responsible for a susceptibility which is proportional to η for small values of η .

The figure 9 shows the expected variation of $\chi_{rev}(\eta)/m'_a(\eta)$ as a function of θ for different values of η between 0 and 1. The increasing curvatures which are expected for increasing fields account reasonably well for the experimental observations of for example Sarkissian *et al.* [19]. See also on figure 20 our results on CuMn 8 at. %. For η fixed however small it may be, there always exists a domain in the vicinity of T_G where $\theta + \eta > 1$ and deviations from linearity in θ are observed.

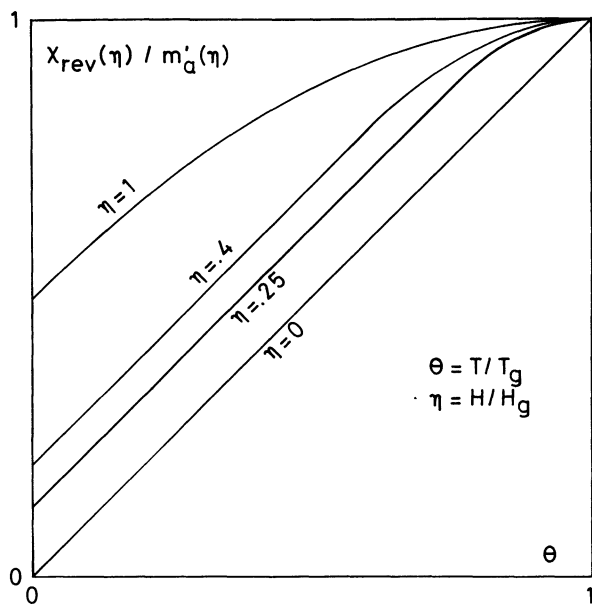


Fig. 9. — Evolution of the predicted reversible susceptibility vs. the reduced temperature θ for increasing values of the reduced field η .

We have expressed our data in terms of $m'_a(\eta)$, the susceptibility of the equilibrium magnetization. Figure 9 represents the actual susceptibility variations in systems like CuMn (or AuFe, AgMn, ... at low concentrations) where m'_a does not depend too much on the temperature. We show in figure 10 a and b the expected behaviours of χ_{rev} when m'_a is given by a Curie-Weiss and by a Curie law respectively. If, as in PtCo [18] and other exchange-enhanced or cluster-enhanced systems, the equilibrium susceptibility evolves from Curie like to Curie-Weiss like behaviour, the susceptibility maximum can appear at a temperature T_{max} different from T_G where the remanences vanish. Obviously in such cases, T_{max} is a totally irrelevant quantity.

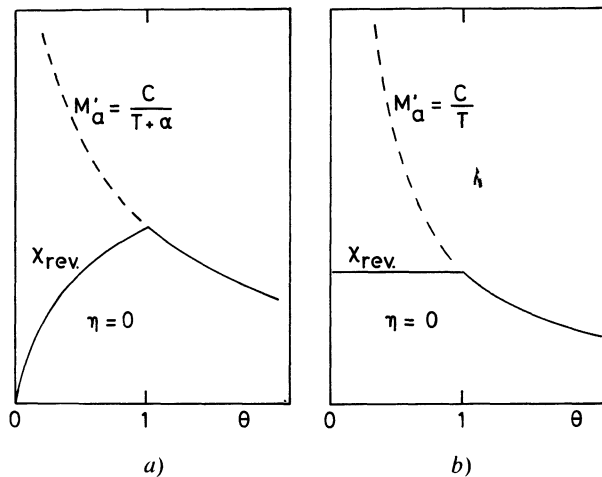


Fig. 10. — Predicted behaviour of the low field susceptibility when the equilibrium susceptibility M'_a follows a Curie-Weiss law (Fig. a) and a Curie law (Fig. b).

10. **Edwards and Anderson's q parameter.** — We have seen above that the reversible susceptibility can be expressed as $m'_a \left(1 - \frac{T.R.M.(\eta)}{M_a(\eta)} \right)$. This expression allows to identify in the limit $\eta \rightarrow 0$ the ratio $T.R.M.(\eta)/M_a(\eta)$ with the parameter $q(t)$ which measures the average $\langle S(t) S(0) \rangle$ of the correlation of the magnetization of a site at time t with the value that it had at time $t = 0$. Physically this formula implies that starting from an unmagnetized sample only those objects for which the correlation has vanished at time t , have acquired their equilibrium magnetization. The correlated objects on the contrary keep the memory of the initially unmagnetized state. Thus the *T.R.M.* in small fields provides a direct measurement of $q(t)$ whose long time limit is the parameter q which Edwards and Anderson proposed to use as the *order parameter* characteristic of an eventual phase transition to a *spin glass phase* [2]. With our assumptions ($P(W) = P(0)$ for $W < W_{max}$) $q(t)$ depends linearly on the temperature and vanishes at $\theta = 1$, i.e. at the temperature

$$T_G \sim \frac{W_{max}}{\text{Ln } \frac{t}{\tau_0}} \tag{Fig. 11}$$

long times and $q = 0$. This conclusion is in agreement with the analysis by Bray and Moore of their Monte Carlo results [20]. For comparison with the experiments, remember that $M_a(\eta)$ does not include the reversible contribution $\chi_0 \eta$ which is not accounted in our model and which will be considered later.

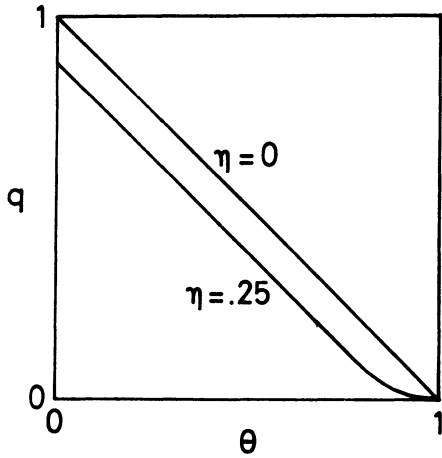


Fig. 11. — Predicted behaviour of the correlation function $q = \langle S(t) \cdot S(0) \rangle$ vs. the reduced temperature θ for two values of the reduced field η .

11. **Qualitative comparison with the experiment.** — The equilibrium curve $M_a(H)$ being given, figure 12 shows how the first magnetization curve is obtained by the first application of a field, starting from the point of zero magnetization. There is indeed an initial term quadratic in the field variation as shown on figure 13 but the presence of a large reversible susceptibility χ_0 is not predicted by the model. By translation along this reversible magnetization $\chi(T = 0) H$, the equilibrium curve $M_a(H)$ can be superposed on the ascending branch of the limiting cycle (in small and in moderate fields).

The *T.R.M.* is obtained, starting from $M_a(H)$, when the field variation is equal and opposite to the field applied during the cooling process. For large fields (above the Rayleigh regime) the descending

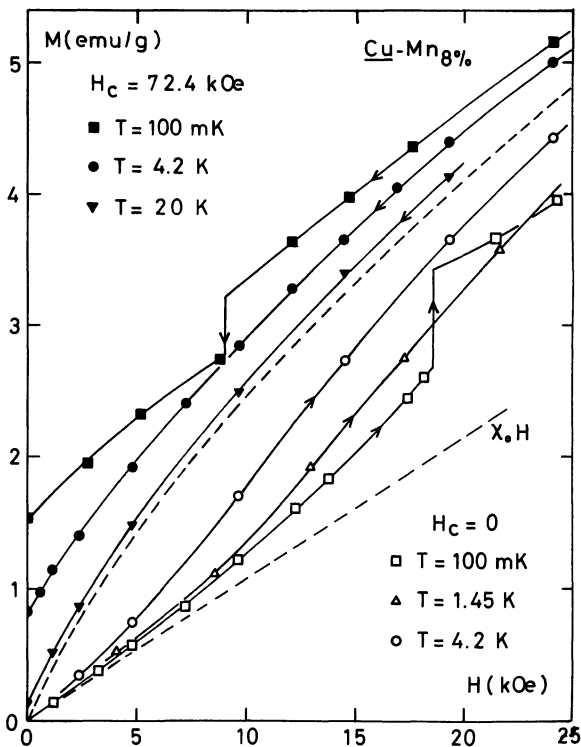


Fig. 12. — The magnetizations of a CuMn 8 at. % vs. field at different temperatures below T_G . The dashed curve is the equilibrium magnetization shown in figure 3. The first magnetization curves at 100 mK (\square), 1.45 K (Δ) and 4.2 K (\circ) are obtained in increasing fields, starting from the zero field cooled sample. The descending branches of the limiting hysteresis curves at 100 mK (\blacksquare), 4.2 K (\bullet) and 20 K (\blacktriangledown) are obtained in decreasing fields, starting from the field cooled magnetization in 72.4 kOe. Notice the unimpredicted reversible susceptibility at $T = 0$ (dashed line) and the magnetization jumps at the lowest temperature.

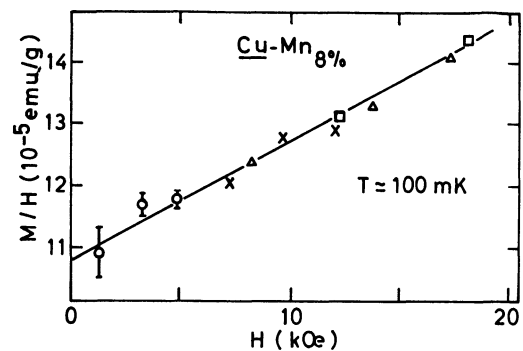


Fig. 13. — The first magnetization curve of a CuMn 8 at. % at 100 mK in a M/H vs. H diagram showing the quadratic Rayleigh behaviour and the existence of a reversible susceptibility χ_0 at very low temperature.

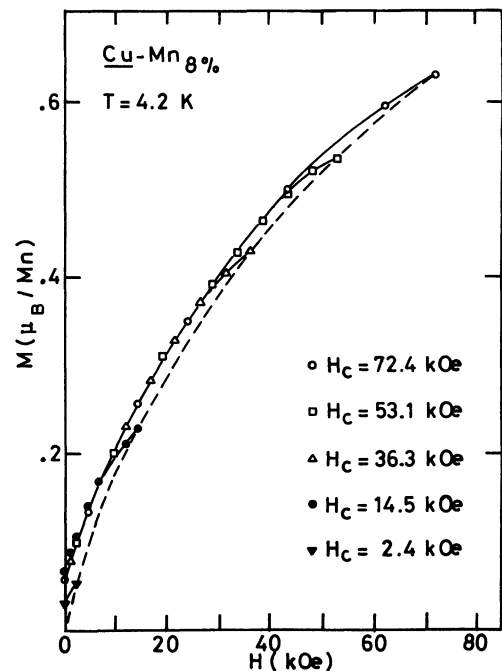


Fig. 14. — The descending branch of the limiting hysteresis cycle is reached through a Rayleigh parabola starting from different points of the equilibrium curve $M_a(H)$ (dashed curve). In small enough fields, the Rayleigh parabola intersects the magnetization axis and the thermoremanent magnetization is not saturated.

branch of the hysteresis cycle is reached (Fig. 14).

The *I.R.M.* is obtained, starting from the first magnetization curve with a decrease in field equal and opposite to the initial increase.

Figure 15 exhibits the experimental behaviour of the *T.R.M.* and of the *I.R.M.* vs. *H* at different temperatures, and should be compared with the predictions of figure 8. Notice the unexplained maximum of the *T.R.M.*

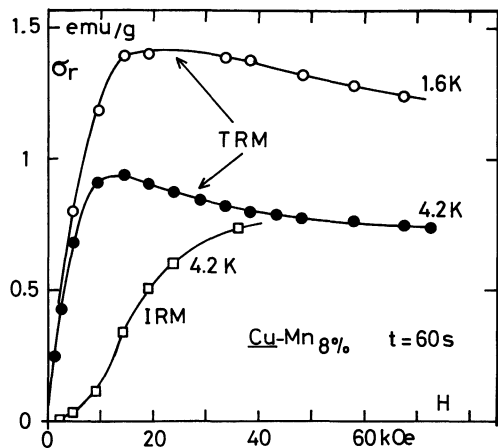


Fig. 15. — The *I.R.M.* and the *T.R.M.* at 4.2 K and 1.6 K vs. the field *H* in CuMn 8 at. %. The field was applied during 60 min. The measurements were performed 60 s after the field was withdrawn.

The temperature dependence of the saturated remanent magnetization measured at fixed times in the region of the plateau which follows the *T.R.M.* maximum, departs noticeably from the predicted parabolic behaviour in the vicinity of T_G (compare figures 8 and 18) : the hypothesis of a sharp cut-off at W_{max} is of course very bad in this domain.

A better fit is obtained with an exponential law :

$$S.R.M. \simeq S(0) \exp - \left(\frac{T}{T_0} \ln \frac{t}{\tau_0} \right) = S(0) \left(\frac{t}{\tau_0} \right)^{-T/T_0} \quad (17)$$

between $T_G/3$ and $2 T_G/3$ (Fig. 16) as was noticed previously [5], [21]. However we observe important deviations from exponential behaviour in both the high and the low temperature range whatever the concentration (see in particular the figure 17 where data at several concentrations have been superimposed in a reduced diagram $S.R.M./S(0)$ vs. T/T_G). The agreement with exponential behaviour is made somewhat better but still unsatisfactory if the value of the *T.R.M.* at its maximum rather than in the subsequent plateau is used as the determination of the *S.R.M.* For the time being therefore we believe that the expression (17) can be considered only as an *ad hoc* best fit in a restricted temperature range. More studies would be necessary to decide about its eventual generalization.

12. Determination of τ_0 : validity of the Arrhénius law. — We have used the excellent agreement which

is obtained with an exponential behaviour in the range $T_G/3, 2 T_G/3$ to make a determination of the characteristic time τ_0 which enters in the Arrhenius law. The parallelism of the straight lines which represent the variations of $\text{Log } S.R.M.$ vs. $T \text{ Log } t$ at fixed *T* and the observed linearity of $\text{Ln } S.R.M.$ vs. *T* (Fig. 16) are consistent with a unique expression

$$S.R.M. \sim S(0) \exp - \left(\frac{T}{T_0} \text{Ln } \frac{t}{\tau_0} \right) = S(0) \left(\frac{t}{\tau_0} \right)^{-T/T_0} \quad [21]$$

which describes both the time and the temperature dependences. The value of τ_0 obtained is $\tau_0 \sim 10^{-11}$ s, consistent with other estimates in this and in other systems.

Even in the high and low temperature ranges where deviations from the exponential behaviour have been noticed the *same* value of τ_0 does lead to a good superposition of the data in terms of $T \text{ Ln } \frac{t}{\tau_0}$ i.e. in terms of the reduced variable $\theta = T/T_G$ which we have introduced (Fig. 10). This scaling $S.R.M. \simeq f\left(T \text{ Ln } \frac{t}{\tau_0}\right) = f(\theta)$ implies the validity of the Arrhenius law and that we are dealing with thermally activated processes.

Figure 19, where the evolution of the remanent magnetizations in different fields is shown vs. $T \text{ Ln } \frac{t}{\tau_0}$, should be compared with figure 8b. Except for the previously considered deviations from parabolic behaviour the qualitative agreement is good. In particular a linear dependence of the *T.R.M.* on $T \text{ Ln } \frac{t}{\tau_0}$ is observed in small fields up to the vicinity of T_G where a saturated regime is always reached ($\eta + \theta > 1$).

We have noticed a deterioration of the above picture in *very small fields*. It would then be necessary to introduce values of τ_0 much larger than 10^{-11} s to restore the above mentioned time temperature correspondance. Typically we would need $\tau_0 \sim 10^{-6}$ s in the 10 Oe range. Other authors have also observed the disappearance of any sign of irreversibility if small enough fields are used [22]. We can comment within the framework of our model on some other consequences of such a change in the characteristic time τ_0 . For example we have seen that the error which was made when identifying the field cooled magnetization $M_a(H)$ with the equilibrium magnetization was of the order of $1/Q$ of the maximum disequilibrium which could be reached in this field (i.e. the *T.R.M.*). With $\tau_0 \sim 10^{-6}$ s this error becomes sizeable. We believe it can account for the fact that in many measurements which have been performed with Squid techniques in fields of the order of 1 Oe, the field cooled susceptibility itself does exhibit a weak

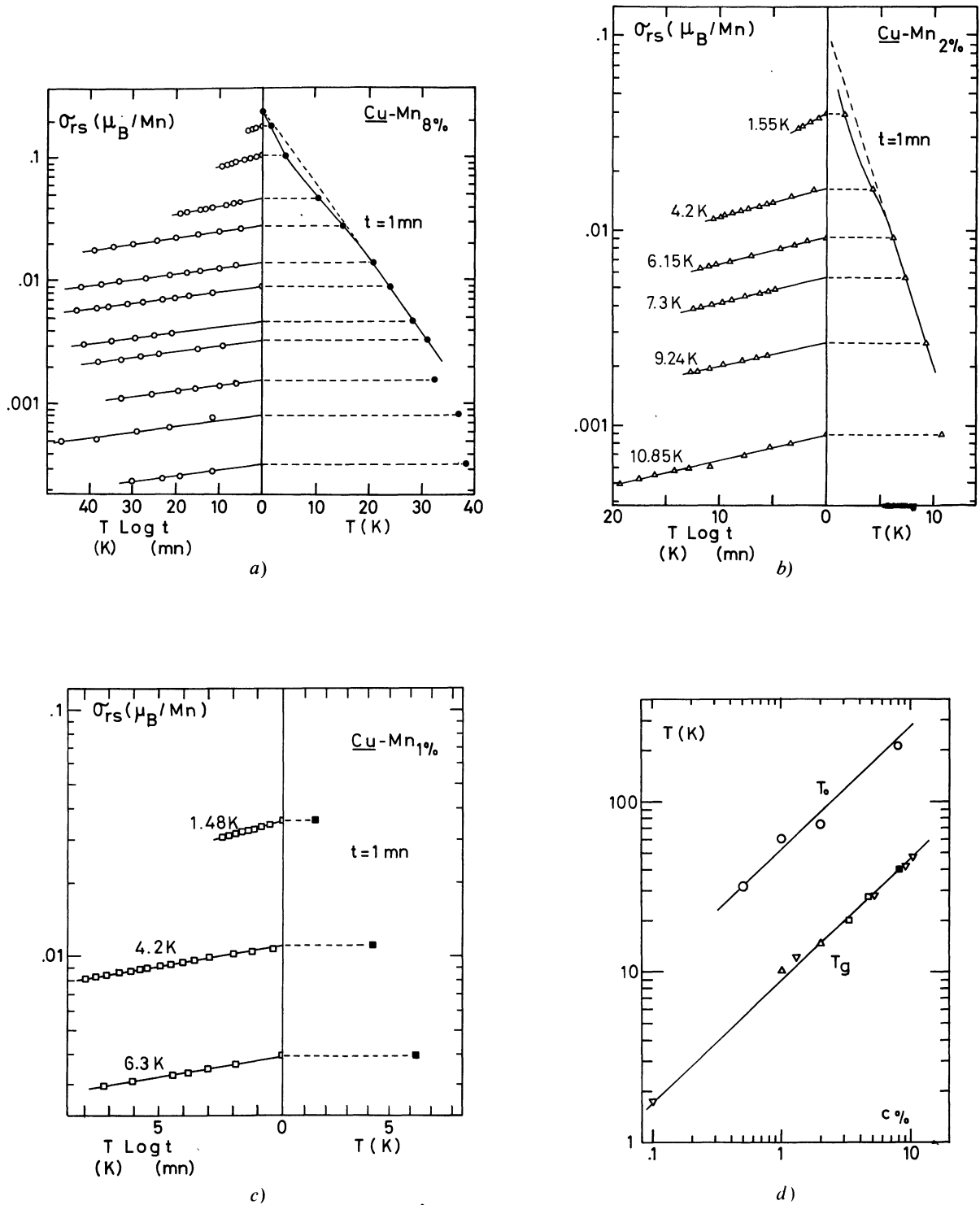


Fig. 16. — The Log of the saturated thermoremanent magnetization $\sigma_{rs}(T, t)$ of a CuMn 8 at. % is shown (Fig. a) at different constant temperatures (left side) vs. $T \text{ Log } t$ for times t varying from 1 to 100 min. after the field was removed. The right side shows the temperature dependence of $\text{Log } \sigma_{rs}(T, t = 1 \text{ min.})$. For

$$T_G/3 < T < 2 T_G/3,$$

we have (left side) $\text{Ln } \sigma_{rs}(T, t) = \text{Log } \sigma_{rs}(T, t = 1 \text{ min.}) - \frac{T}{T_0} \text{Ln } t$ with $T_0 \approx 210 \text{ K}$, and (right side)

$$\text{Ln } \sigma_{rs}(T, t = 1 \text{ min.}) = \text{Ln } \sigma_0 - \alpha \frac{T}{T_0}.$$

Those results can be condensed in the unique expression :

$$\sigma_{rs} = \sigma_0 \exp\left(-\frac{T}{T_0} \text{Ln } \frac{t}{\tau_0}\right) \text{ with } \tau_0 \approx 10^{-11} \text{ s.}$$

Figures b and c : same results for CuMn 2 at. % ($T_0 = 84 \text{ K}$) and 1 at. % ($T_0 \approx 59 \text{ K}$) respectively.

Figure d : the concentration dependence of T_0 (O) is compared with the concentration dependences of T_G (∇ : [32], Δ : [33], \square : [34], \blacksquare : our work and [35]). $T_G \sim 0.2 T_0$.

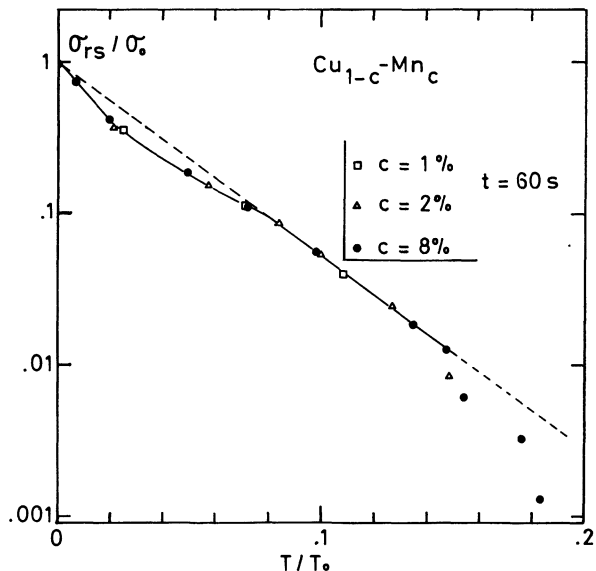


Fig. 17. — The same data as in figure 16 (for $t = 1$ min.) are superimposed in a unique reduced diagram $\sigma_{rs}/\sigma_0 = f(T/T_0)$ including all three concentrations (8 at. %, 2 at. %, 1 at. %) with σ_0 and T_0 defined in figure 16.

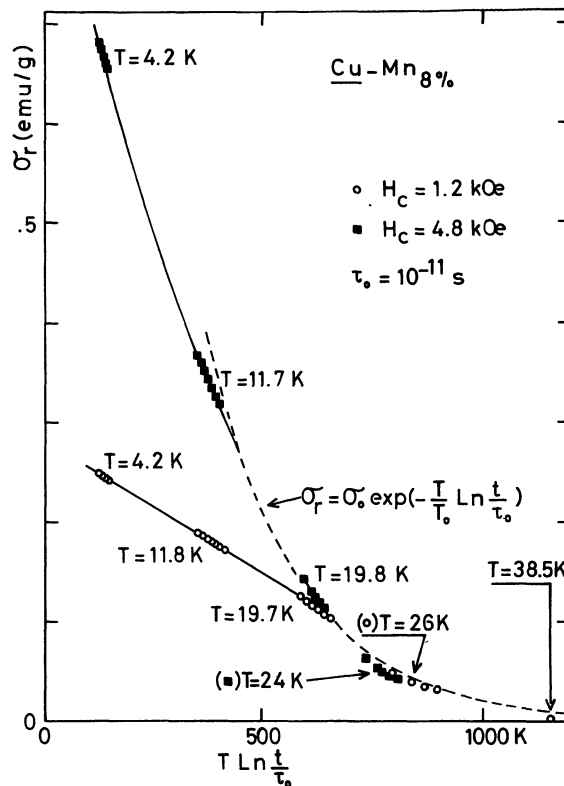


Fig. 19. — The thermoremanent magnetization of CuMn 8 at. % in different constant fields is shown vs. $T \ln \frac{t}{\tau_0}$. Notice the linear temperature dependence in the unsaturated regime at low $T \ln \frac{t}{\tau_0}$ values.

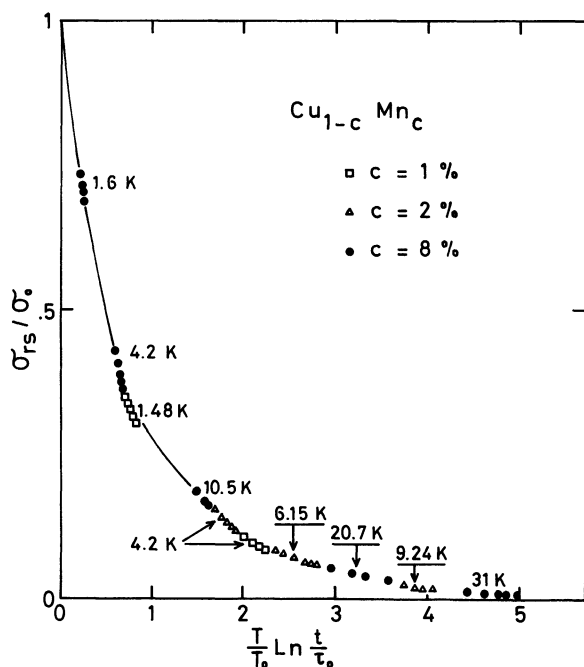


Fig. 18. — The same data as in figure 16 including the time ($1 \text{ min.} < t < 100 \text{ min.}$) and temperature ($T_G/25 < T < T_G$) variations of all three concentrations (8 at. %, 2 at. %, 1 at. %) are shown in a unique reduced diagram σ_{rs}/σ_0 vs. $\frac{T}{T_0} \ln \frac{t}{\tau_0}$ with $\tau_0 \approx 10^{-11}$ s determined as in figure 16.

maximum [23]. This argument suggests a correction of the form :

$$\chi_{eq} = M'_a(H) + \frac{M'_a(H) - \chi_{rev}(H)}{Q}$$

which seems of the correct order of magnitude to make the small structure noticed on $M'_a(H)$ vanish.

13. Discussion of some discrepancies. — We have quoted several features which the model fails to describe. They are :

- a) a finite and large reversible susceptibility at $T = 0$,
- b) the presence of a maximum in the $T.R.M.$,
- c) indications that τ_0 might become larger in small fields,
- d) the existence of jumps in the magnetization,
- e) another major difficulty is associated with the sharpness of the susceptibility cusps which have been reported in some cases (not all). The last point e), to us, is much more important than the others (a)-c) since whatever sharpness which is present in our description is obviously connected with the unphysical cut-off which we have assumed for the distribution $P(W)$ and would vanish with any reasonable assumption on the shape of $P(W)$.

Points a), b), c), d), on the contrary could conceivably be amended in a more realistic description. At least the discrepancy would not obviously increase in an attempt to make the model more realistic as in the case of e).

Take point *a*) for example. In order to account for the additional large initial susceptibility $\chi(T = 0)$ we would need to introduce an additional density of states $\mathcal{F}'(\varepsilon_0)$ the access to which would not be restricted by an associated distribution $P(W)$ of energy barriers. We need

$$\frac{\mathcal{F}'(\varepsilon_0)}{\mathcal{F}(\varepsilon_0)} = \frac{\chi(T = 0)}{m'_s(h)}$$

to be of the order of unity. This *ad hoc* modification to the model is *not* contradictory with our initial motivations (see § 2) : we introduced the distribution $P(W)$ in order to account for the observed large relaxation times of the magnetization and of the energy : this does not exclude the presence at the same time of additional reversible responses of other or of the same objects. In particular, the square cycle (shown Fig. 5) traduces the response to a field applied along the *easy axis* of the objects. The response transverse to this easy axis is in fact reversible (dash-dotted line figure 5). In an assembly of objects where the orientation of the anisotropy axis is presumably isotropic, the barriers restrict the response associated with the component of the external field along the easy directions of the objects. But there exists a reversible response of the same magnitude associated with the transverse component.

The fact that the model does not account for the maximum of the *T.R.M.* (point *b*)) may be a consequence of the very unsatisfactory assumption which we made that $P(W)$ and $\mathcal{F}(\varepsilon_0)$ are independent from each other and from the magnetic state of the sample. It would be tempting to introduce a correction on W_{\max} proportional to the deformation which has been applied to the system : $W_{\max} \rightarrow W_{\max} - \alpha \Delta M$ where ΔM is the variation of the magnetization starting from the initial equilibrium situation. Thus in the saturated regime the remanent magnetization associated with a larger field would relax faster than a remanent associated with a smaller field and would appear smaller at any finite measurement time.

We do not regard the apparent increase of the magnitude of τ_0 when very small measurement fields are used (point *c*)) as the manifestation of an inadequacy of the model. Physically τ_0^{-1} is related with the curvature of the potential well and it is expected that this curvature increases when W increases [15]. Of course it is reasonable to neglect (as we have done) the effect of the variations of τ_0 as compared with the huge variations in τ arising from the large distribution of W since W appears in the exponent of the Arrhenius law. When moderate or high fields are used a wide enough range of W is reached and τ_0 can reasonably be replaced by its average value. The small fields however only select those barriers which are small and which correspond to a large τ_0 very different from the average value.

We have stressed from the beginning that this model

has no ambition in explaining the occurrence of magnetic jumps (point *d*)) in positive fields (Fig. 12) nor in negative fields as those discussed in reference [16]. Concerning the latter however, provided the interaction which is responsible for $P(W)$ and $\mathcal{F}(\varepsilon_0)$ has the rotational symmetry (like the R.K.K.Y. interaction) the magnetizations which we described are conserved by rotation. In this case it should be possible to align at no cost the remanent magnetization along any direction of space. The results of reference [16] clearly show that the small reverse field which is necessary to abruptly reverse the magnetization of a CuMn alloy must be due to the anisotropy associated with the conduction electron spin flip cross section due to spin orbit coupling. This cross section is particularly small in CuMn but it can be modified by orders of magnitude through the addition of a very small amount of non magnetic impurities (such as Au or Pt) with little, if any, influence on the properties which we have attempted to describe in this paper.

This shows that the existence (or non existence) of magnetic jumps is not a characteristic property of the spin-glass state. On another hand it is clear that the spin-glass state can be constructed with an interaction which has the rotational symmetry. This observation eliminates any explanation which would imply at some stage the necessity of an anisotropic interaction (such as the dipolar interaction) to account for the observed relaxations (i.e. the energy barriers) [25].

14. What do the magnitudes of Q , W_{\max} and H_{\max} tell us about the objects ? — The main parameters of the problem are, apart from the additional initial susceptibility $\chi(T = 0)$, the width W_{\max} of the distribution $P(W)$ of the barrier heights, the associated field H_g and the parameter $Q \sim \text{Ln} \frac{t}{\tau_0}$ through which the experimental time affects the problem. We have measured $\tau_0 \sim 10^{-11}$ s and $Q \sim 25$. The energy $W_{\max} \simeq kT_G Q$ is therefore about 25 time larger than the apparent energy kT_G through which it manifests itself in usual measurements ($t \sim 1$ s). This is much larger than the R.K.K.Y. interaction at the average interimpurities distance. This has several consequences. The first, which has been often stressed, is that the considered objects are large size objects. Comparing

$$\overline{M}_g H_g \sim W_{\max} \sim QkT_G$$

one gets an estimation of $\overline{M}_g \sim 500 \mu_B$ in CuMn 8 at. %, $250 \mu_B$ in CuMn 1 at. % (to be compared with $95 \mu_B$ obtained from energy relaxation measurements in AuFe 4 at. % [11]).

This large value implies the association of a large number n of individual moments. This is an important feature which has to be taken into account in the microscopical description of the objects. A picture was proposed by Holtzberg *et al.* [5] who attempted

to extend to spin glasses Néel's ideas on fine anti-ferromagnetic particles. The suggestion was to attribute the origin of the barriers W to an internal dipolar energy proportional to the volume of the objects (called *magnetic clouds*); this volume term is bound to become larger than the interactions with neighbouring clouds increasing presumably with the surface of the cloud for some critical size, even if the magnitude of the R.K.K.Y. is much larger than that of the dipolar interaction. Although the interclouds interactions were thus thought to be determinant on the size of a cloud, their effect was in practice omitted in the description of the time effects which was subsequently performed.

The model which was in fact used was a model of symmetrical double well potentials which like equivalent superparamagnetic descriptions [24] can account for neither the energy relaxations nor the shape of the equilibrium curve $M_a(H)$. The model however succeeded in popularizing the idea of a distribution of potential barriers and the phenomenological discussion of time effects which are associated to this distribution. We have tried to go a step further by introducing phenomenologically the interactions between the objects through the distribution $\mathcal{F}(\epsilon_0)$ or the equivalent use of the equilibrium magnetization $M_a(H)$.

The main criticism which has been raised against the *clouds model* is that the microscopical mechanism which it proposes for the occurrence of *clouds* relies on the dipolar interaction a speculation which, as we saw, has been ruled out by recent measurements [25, 16]. We escape this criticism because, being less ambitious, we do not attempt any such description of the objects. Let us stress however, that whatever satisfactory it might be to have a static picture of the objects, the important point in the problem is not the objects themselves but the *distributions* and the fact that a reasonably *flat and constant density* of objects can be assumed in the conditions of time, temperatures and fields which we consider. This situation is of course automatically realized when the objects are well characterized material fine particles such as Néel considered [15]. But we *are not restricted* to this description. The deformation of the system associated with an experiment may well modify the initial objects, we need only that the new objects which are defined at each step are again characterized by the same distributions. Recall that there is a strong reason for the distributions to remain a white spectrum as it is that in an amorphous system no special value of the energy should be favoured. The additional requirement upon the constancy of the density probably implies little less than the presence of some typical size or correlation length in the problem which would be reasonably field and temperature independent. We believe that in the absence of obvious geometrical grains in a spin glass such a description in term of say *virtual fine grains* would be appropriate.

15. Frequency dependent maximum and frequency independent cusp. — Nothing in this model would account for the sharpness of the susceptibility data which first started the *phase transition* hypothesis. Even when, as in this paper, a sharp unphysical cut-off of the distribution $P(W)$ is assumed at W_{max} , the predicted susceptibility is rather unpathological : neither figures 8 nor 9 exhibit the upward curvatures which have been experimentally reported [1] and which can be seen on figure 20. Further, T_G is expected to be frequency dependent :

$$\frac{1}{T_G} = \frac{kQ}{W_{max}}$$

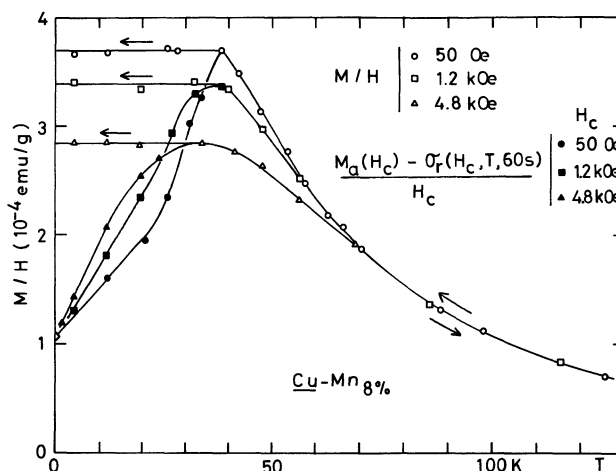


Fig. 20. — The initial susceptibility of CuMn 8 at. % vs. temperature in different constant fields. The equilibrium (field cooled) susceptibility does not show any anomaly. The reversible susceptibility $\left(\frac{M_{eq}(h) - T.R.M.(h)}{h}\right)$ shows a rounded maximum in increasing fields.

should be a linear function of $\ln t$ as shown on figure 21, the intercept of the linear variation with the $\ln t$ axis occurring for $\ln \tau_0$. In principle, varia-

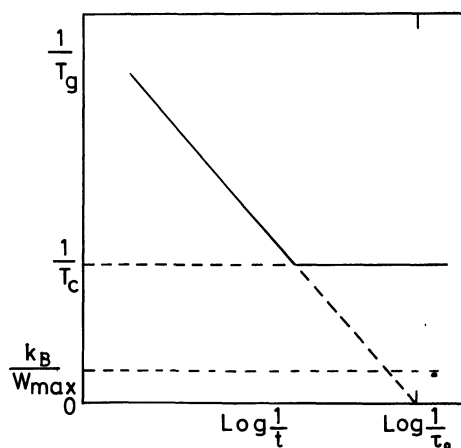


Fig. 21. — Predicted variations of $\frac{1}{T_G}$ vs. $\ln t \cdot \frac{1}{T_c} \approx \frac{k}{\Delta J}$ is the proposed high temperature limit of the variations of T_G (since, over T_c , the cohesion of objects having the necessary size to produce energy barriers as high as W_{max} cannot be maintained).

tions of T_G should be observed over the whole range between the long time limit ($T_G = 0$), and the short time limit ($T_G = W_{\max}/k$) which would occur for frequencies of the order of τ_0 .

This picture does agree with some of the experimental data which have been reported in insulators like $\text{EuSr}_{1-x}\text{S}_x$ or true glasses doped with metallic impurities [26] or in systems like LaAl_2Gd [6] which, although an f impurity is involved, are obviously to be classified among the archetypal spin glasses [27].

In low-concentration AgMn in contrast, experiments extended over decades in frequency [8] failed to show any sizeable frequency dependence, a feature which has been confirmed in several other systems [28]. In some cases it was possible, through a heat treatment, to induce a frequency dependence in a sample where no such dependence was present in the initial quenched state [9].

These contradictions we believe, raise the all important question of the validity range of our speculations. Remember that the experimentally determined values of W_{\max} are very large : much larger than the average magnitude $\Delta J = \sqrt{\langle J^2 \rangle}$ of the R.K.K.Y. interaction at the average interimpurities distance. This prompted us into considering the above discussed large size objects for which it is not unreasonable to argue that their rearrangements will involve energies much larger than ΔJ . Inasmuch as such objects exist it is possible to justify a $W_{\max} \sim 25 \Delta J$ and a T_G whose higher limit is W_{\max}/k . It is however obvious that the model collapses at temperatures of the order of $T_c \sim \Delta J/k$ when the system becomes paramagnetic and the R.K.K.Y. interaction is no longer able to sustain the necessary cohesion of the objects which are the basis of our model. This temperature T_c sets the higher frontier to the validity range of our model.

More precisely, consider an hypothetical alloy which would be built up with impurities in the same positions than in the real alloy and where the interaction would everywhere be positive and identical to the modulus of the real interaction. This alloy has a ferromagnetic transition at $T_c \sim \Delta J/k$. We may now construct the *associated Mattis alloy* by flipping (when necessary) the spins to their actual orientation in the real alloy : provided that for each spin we flip, we flip at the same time all its interactions with the $N - 1$ other spins, we conserve a *flat* model which still exhibits long range order below T_c . (This alloy differs from our real alloy by the fact that any closed loop of bonds contains an even number of antiferromagnetic bonds and frustration is absent.) We know (in particular from numerical experiments) [31] that this long range order is destroyed by the frustration effect [30] (and we enter the spin glass state) when a sufficient fraction of the bonds are flipped to their actual sign in the real alloy. There must be a qualitative difference between the situation over T_c where the temperature in any case prevents the occurrence

of long range order and the situation below T_c where the frustration prevents the occurrence of an otherwise infinite cluster. Only below T_c we can speculate about objects of the required (reasonably temperature independent) size to justify the magnitude of $W_{\max} \sim 25 T_G$. T_c therefore would limit the temperature range where the glass behaviour characterized by a scaling in terms of $\theta \sim T/T_G$ (with $T_G \sim (\ln t)^{-1}$) can be observed. In a similar way, we believe the fusion temperature of say crystalline silica sets the higher limit where vitreous silica can be observed and the Curie temperature of the magnetic material sets the limit where fine grains behaviour can be observed in an alloy with this metal.

We have represented in the figures 22, 23 the predicted variations of the susceptibility (assuming an equilibrium susceptibility M'_a given by a Curie-Weiss law and forgetting about $\chi(T = 0)$) and of the parameter $q(t) = \frac{T.R.M.}{M_a} (h \rightarrow 0)$ vs. T at different times and vs. time at different temperatures. We simply assume in this rough picture that the distribution $P(W)$ is unaffected up to temperatures of the order of $\Delta J/k$ where it suddenly vanishes. We expect a transi-

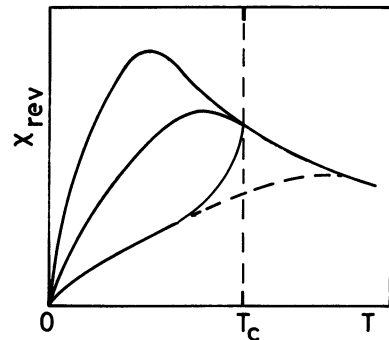


Fig. 22. — Schematic representation of the expected features of the reversible susceptibility vs. temperature for different measurement times. The scaling T/T_G (yielding a time dependent maximum) vanishes at T_c where the system becomes paramagnetic.

tion from a *glass regime* where all the properties scale with T/T_G (with T_G being frequency dependent) to a regime where T_G being larger than $T_c \simeq \Delta J/k$ the transition to the paramagnetic regime occurs at T_c (Fig. 22) with a sudden inversion of the curvature of the susceptibility. It is remarkable that reasonable estimations of ΔJ have been made in the past on the basis of the temperature of the susceptibility maximum : this implies that $kT_G \sim W/Q$ and ΔJ are actually of the same magnitude at the usual frequencies i.e. $W_{\max} \sim 25 \Delta J$. This can be checked using independent estimations of ΔJ which can be made very accurately from the magnetization curve in the vicinity of its saturation [29]. It is therefore not unexpected if, depending on the system, W_{\max}/Q can be found to be slightly larger than ΔJ (AgMn), or slightly smaller (LaAl_2Gd) for usual values of Q .

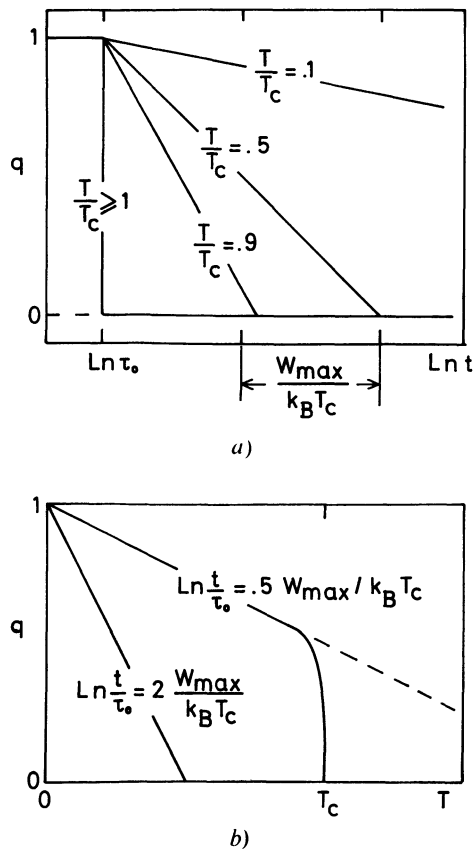


Fig. 23. — Predicted behaviour of the autocorrelation function $q = \langle S(0) S(t) \rangle$ vs. time for different temperatures (Fig. a) and vs. temperature for different measurement times (Fig. b).

This is supported by the fact that when a straight line is drawn through the experimental data showing the frequency dependence of T_G (like in Fig. 21) it very seldom provides a value of τ_0 (by extrapolation to the $\log t$ axis) in agreement with the low temperature data on the same samples, or even a value which can be easily justified on physical grounds ($\tau_0 \sim 10^{-20}$ s

in LaAl_2Gd vs. 10^{-11} s from remanent data at low temperatures). This suggests that this straight line was incorrectly drawn through data which are already in the cross-over regime.

16. Conclusions. — We have discussed the consequences of a model of a distribution of asymmetrical, thermally activated double well potentials and found that such a model gives a satisfactory description of the properties of spin glasses.

Some discrepancies would probably be explained by a modified model which would use more realistic distributions and would account more properly for the correlations. The consideration of the magnitudes of the characteristic energies which are required, implies the necessary existence of two regimes : a glassy regime $T < T_c$ where the properties can be expressed in terms of a reduced variable $\theta = T/T_G$ where T_G decreases as $(\log t)^{-1}$ and a regime $T > T_c$ where the system is truly paramagnetic or superparamagnetic. It is in the transition towards paramagnetism that the pathological effects, which eventually would need the definition of new phase, occur.

The model which has been used is very general since it is justified essentially on the basis that the structural disorder inherent in the system does not favour a particular value of the energy. This generality implies similarities between the magnetic properties of spin glasses and the properties of other disordered systems (for example the elasticity of rubbers). This justifies the name *spin glass* given to this type of magnetic system and also the application to spin glasses of a more practical definition which was established for glasses namely : systems which react like liquids at long times like solids at short times.

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