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Equilibrium magnetization of a spin glass : is mean-field theory valid ?

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Résumé. — A partir des mesures d'aimantation d'un alliage verre de spin AgMn 10,6 % en fonction de la température entre 4,2 K et 50 K et du champ magnétique de 26 à 9 000 gauss, nous tentons d'établir un diagramme de phase dans le plan (H, T) d'après l'analyse de De Almeida et Thouless ainsi que Parisi et Toulouse. Un examen plus approfondi de la région au-dessus de la température de transition verre de spin permet de comparer nos courbes d'aimantation avec les prédictions spécifiques concernant le terme non linéaire de l'aimantation faites par Suzuki à partir du modèle Sherrington-Kirkpatrick. Un accord qualitatif est trouvé.

Abstract. — Measurement of field cooled magnetization of a AgMn 10.6 % alloy as a function of temperature between 4.2 K and 50 K and magnetic field from 26 to 9 000 gauss is performed in order to link the low field and high field regimes of a spin glass. This can possibly help to define a phase diagram in the (H, T) plane as proposed by De Almeida and Thouless and Parisi and Toulouse. A closer investigation of the vicinity of T_g above T_g allows to compare the magnetization curves with the specific prediction of the mean-field model of Sherrington and Kirkpatrick. A qualitative good agreement is found.

Introduction. — In this letter [1] we present some systematic measurements of the equilibrium magnetization of a spin glass alloy above and below the spin glass temperature T_g as a function of temperature and magnetic field. The first purpose of these experiments stems from the need of a detailed survey, for a given system, of the « low field » behaviour of the magnetization together with the « high field » one : we define these two regions below. Indeed previous work on spin glass alloys has very often been confined in one of these two regimes : for instance the work of Nagata *et al.* [2] or Guy [3] is certainly a low field study ; on the contrary the work of Smit *et al.* [4] offers a very high field determination of M . A notable exception is the early work of Hirschkoﬀ *et al.* [5] continued by Thomson and Thompson [6]. However these authors work within a concentration and temperature range two orders of magnitude smaller, and the necessary connection between their results and those aforementioned rely on the validity of scaling laws for the spin glass parameters [7]. Finally the work of Tholence and Tournier [8] spans a quite large area of field and temperature but is not systematic enough.

The theoretical motivation of our work is based on the possible existence of a phase diagram

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for the spin glass phase in the (H, T) plane as originally proposed by De Almeida and Thouless [9]. Specifically, at any temperature T below T_g , there exists a magnetic field $H_c(T)$ above which the system can be described by the mean-field equations of Sherrington and Kirkpatrick [10], whereas below that field a specific hypothesis concerning the structure of the spin glass phase must be done. This line of thinking is entirely contained in the work of Parisi and Toulouse [11] and rests on the existence of a well defined line $H_c(T)$ in the (H, T) plane separating the spin glass phase from the paramagnetic phase. To what extent can this be made apparent in a real spin glass alloy ?

The second purpose of our work is to look very carefully at the vicinity of T_g , above T_g , in order to be able to compare the measured magnetization with the mean-field prediction from the Sherrington-Kirkpatrick model [10] as analysed by Suzuki [12]. This author has shown that the phase transition at T_g which manifests itself in small finite field by a divergence of the parameter $q(H, T)$ implies also a divergence of the non linear magnetization i.e. the term beyond the (non divergent) susceptibility. By measuring the magnetization *above* T_g one avoids experimentally the problem of the occurrence of irreversible effects and one also avoids theoretically the question linked to the symmetry breaking of the order parameter as it is well established [13] that *above* T_g a single value of $q(H, T)$ is obtained. It is thus very tempting to test the mean-field theory for spin glasses in this region. Curiously enough very little work has been devoted to this specific issue in the past : only the recent data of Miyako *et al.* [14], Berton *et al.* [15], Barbara *et al.* [16] are pertinent although a lot of older data could also be used in the same spirit [17]. The final aim of this type of measurement would be to establish experimentally the existence of an equation of state for the spin glass transition as originally proposed by Suzuki [18].

1. Equilibrium magnetization results. — In order to compare the magnetization with some specific predictions [11, 12, 19] we want to determine what is the equilibrium magnetization of our spin glass below T_g . From lack of a better criterion we take as a definition of equilibrium the time independent magnetization i.e. the *field cooled* magnetization below T_g . By doing so we do not prove it to be at equilibrium in the thermodynamic sense : this question is precisely being raised by the calorimetric high precision work of Fogle *et al.* at Berkeley [20]. However we do not know of any other means to obtain an other time independent magnetization state. We note that the results of Knitter and Kouvel [21] who addressed exactly this question by a different technique are not inconsistent with our definition : their result is similar to the extrapolation to infinite time of a time varying magnetization. The fact that the field cooled state is time independent and history independent has been established by many people : Tholence and Tournier [8], Guy [3], Nagata [2] and more recently Malozemoff *et al.* [22] over a longer period of time.

Our measurements were done on a single crystal of AgMn 10.6 % previously used for the X-ray determination of the short-range order [23]. It was obtained after quenching in water from below the melting temperature and after spark cutting a cylinder 8 mm in height and 5 mm in diameter. The homogeneity of the Mn concentration was checked by an X-ray microprobe analysis carried out on a separate piece. The magnetization was obtained with a vibrating sample magnetometer with an accuracy better than 1 % and a sensitivity of about 10^{-4} emu. It was equipped with a He gas flow variable temperature cryostat. The He gas flow temperature was stabilized to better than 0.1 K with reference to a platinum thermometer attached to the cryostat tail near the sample position. The sample temperature was continuously recorded with another platinum thermometer located a few millimeters above the sample position on the sample holder. In this way very reproducible results could be obtained although no absolute calibration of our temperature was carried out. The main source of systematic error came from small contributions to either M or H which tend to blow up the ratio M/H as $H \rightarrow 0$. The magnetic field of our magnet was determined by a fit to a Curie law at high temperature.

Figure 1 displays the ensemble of our results for our fixed field, variable temperature measurements. In order to exhibit the Curie-Weiss behaviour of the high temperature part ($T > T_g$) we have plotted the inverse of the apparent susceptibility, H/M , as a function of T , showing a Curie-Weiss temperature of $+13 \pm 0.5$ K for this alloy. The field dependence of this apparent susceptibility is remarkable : indeed a very flat ($< 1\%$) « plateau » appears below T_g for fields less than $\sim 1\,000$ gauss and a small « cusp » like minimum (5 % deep at most) is present right near T_g for the lowest field used (26 gauss). At higher fields the transition between the Curie-Weiss regime and the spin glass plateau is progressive and a smooth rounding is obtained. None of these magnetization data are time dependent provided that they are obtained with field cooling through T_g . Once the lowest temperature was obtained (4.2 K) the magnetization upon subsequent warming was observed to be identical to that measured during cooling (provided that the field was kept fixed at all times). This fact further stresses the equilibrium nature of the magnetization. These observations allow one to link the low field results obtained [2] to the « high » field ones, especially those of Hirschkoﬀ, Symko and Wheatley [5].

A qualitative fit of these results can be obtained for the region $T > T_g$ with the prediction of

Fig. 1. — Inverse of the apparent susceptibility H/M for AgMn 10.6 % as a function of magnetic field as indicated on each set of point symbols (in gauss). These data points have been obtained at fixed field and variable temperature i.e. by field cooling through T_g . As explained in the text these represent an *equilibrium* state of the spin glass phase. The insert shows an attempt to determine a phase diagram in the (H, T) plane. We define (arbitrarily) the boundary of the spin glass phase by the point of the $M(T)$ curve departing by 3 % from the low temperature value (arrows).

a simple Sherrington-Kirkpatrick set of equations including external field [26]. Such a qualitative fit is presented in figure 2 but no attempt is made to make this into a quantitative fit. In particular we have left the parameter $J_0 = 0$ whereas the experimental data clearly show a Curie-Weiss behaviour. The salient features from figure 2 are the extreme field sensitivity to the departure from the Curie law near T_g and the location of the De Almeida-Thouless instability point which occurs very near the maximum of magnetization of the S. K. solution as noted by Vannimenus, Parisi and Toulouse [19] (which corresponds to a minimum of the quantity H/M). If we further

Fig. 2. — Calculated [26] Sherrington-Kirkpatrick magnetization for fixed field and variable temperature expressed in units of T_g . The curves represent eqs. (1) and (2) until the De Almeida-Thouless point (open circle) below this point the magnetization is represented as independent of T according to the Parisi-Toulouse hypothesis [11]. A better qualitative fit can be obtained with figure 1 by introducing the S. K. parameter $J_0 = 0.33$ to account for the finite Curie-Weiss temperature observed.

assume, according to the Parisi-Toulouse hypothesis, that the magnetization is temperature independent below $H_c(T)$ then it becomes a quite difficult challenge to pin-point experimentally the De Almeida-Thouless line because the point where the magnetization plateau ends is experimentally ill defined. As a working criterion we have taken those points where the magnetization departs by 3 % from the low temperature plateau [24] : this is indicated by an arrow on figure 1 and enables one to draw a set of points in the (H, T) diagram (insert of figure 1) with admittedly increasing error bars along the T axis as H becomes larger. From this data no serious test of the De Almeida-Thouless relation is possible. We note however that the scale of fields is certainly wrong as the data presented for 9 kgauss mimic approximately the mean field result for $H = 1$ in reduced units i.e. $g\mu_B H = k_B T_g$ for a spin of $1/2$. In our case $T_g = 37.5$ K so this would correspond to a field of the order of 250 kgauss. At this stage we can only list the factors we should take into account to reduce this seemingly large discrepancy : i) the fact that Mn has a spin $S \simeq 2$ [1]; ii) the slope of the Curie law indicates that the effective moment corresponds to

3 Mn atoms because of the presence of short-range order as revealed by X-ray diffuse scattering [23]; iii) the finite Curie-Weiss temperature : we feel that these factors taken together will go in the right direction but will not be sufficient to account for all the discrepancy.

2. **Non linear magnetization near T_g .** — The Sherrington-Kirkpatrick equations [10] in presence of a field H and for $J_0 = 0$ are :

$$q = (2\pi)^{-1/2} \int_{-\infty}^{+\infty} dz e^{-z^2/2} \tanh^2 \left(\frac{Jq^{1/2} z + H}{kT} \right) \quad (1)$$

$$M/M_0 = (2\pi)^{-1/2} \int_{-\infty}^{+\infty} dz e^{-z^2/2} \tanh \left(\frac{Jq^{1/2} z + H}{kT} \right) \quad (2)$$

with $J = kT_g$ and M_0 the saturation magnetization. A straightforward expansion of these equations for small values of the arguments above T_g gives :

$$q = \frac{H^2}{k^2(T^2 - T_g^2)} - 0(q^2) \quad (3)$$

and

$$M/M_0 = \frac{H}{kT} - \frac{T_g^2}{kT^3} qH - \frac{1}{3} \frac{H^3}{k^3 T^3} + 0(H^5). \quad (4)$$

Combining (3) and (4) and dividing by H yields

$$\frac{1}{M_0} \frac{M}{H} = \frac{1}{kT} - \frac{H^2}{3k^3 T^3} \left(\frac{T^2 + 2T_g^2}{T^2 - T_g^2} \right) + 0(H^4) \quad T > T_g \quad (5)$$

where the divergence in the non linear magnetization is obviously related to that of $q(T, H)$. The same kind of expansion carried out at $T = T_g$ gives :

$$q = \frac{1}{\sqrt{2}} \frac{H}{kT_g} + 0(H^2) \quad (6)$$

$$\frac{1}{M_0} \frac{M}{H} = \frac{1}{kT_g} - \frac{1}{\sqrt{2}} \frac{H}{k^2 T_g^2} + 0(H^2). \quad (7)$$

Note the occurrence of a singular odd term in the field dependence of the susceptibility [19]. For the regime $T < T_g$ Parisi and Toulouse [11] predict that the magnetization at low temperature becomes temperature independent below the De Almeida-Thouless point. The magnetization is then given by the value of the magnetization at the critical line $H_c(T)$. This line is given by [9] :

$$\frac{T^2}{T_g^2} = (2\pi)^{-1/2} \int_{-\infty}^{+\infty} dz e^{-z^2/2} \cosh^{-4} \left(\frac{Jq^{1/2} z + H_c}{kT} \right). \quad (8)$$

Again, a small parameter expansion of (8) and (1) gives near T_g :

$$H_c^2 = \frac{4}{3} k^2 T_g^2 \left(1 - \frac{T}{T_g} \right)^3 \quad (9)$$

which is carried in (1) and (2) this time expanded below T_g to yield [19] :

$$\frac{1}{M_0} \frac{M}{H} = \frac{1}{kT_g} - \left(\frac{3}{4}\right)^{2/3} \left(\frac{H}{kT_g}\right)^{4/3} + o(H^3) \quad (10)$$

where the occurrence of fractional powers of H is due to the specific relation $H_c(T)$ of the De Almeida-Thouless line. Although we do not expect *a priori* the relations (4), (7) and (10) to give any reasonable fit to the experiment carried on a real spin glass because of the over simplifications inherent to the infinite range Ising system of spin 1/2 we intend to use them as a guide in order to analyse the experimental results. The application of more elaborate theories for the Heisenberg case would be required in our case at the cost of a more complex phase diagram [25]. We feel that, at this stage, our data do not warrant such a comparison.

We have carried out two series of measurements of the susceptibility M/H at constant temperature *versus* the magnetic field. The first series was done in a range of field 100 gauss-5 000 gauss and between T_g and $1.5 T_g$. A special run was done at 4.2 K where at every field the sample was field cooled to that temperature after being warmed up above T_g *prior* to changing the field to a new value. Needless to say that only the flexibility of the gas flow cooling system associated to the vibrating sample magnetometer could make this type of run possible. The major uncertainty was again the knowledge of the magnetic field of the magnet which was given by a Hall effect probe. As can be seen in figure 3 where some typical M/H data are shown, this affects seriously the low field region. We note that the M/H data at 4.2 K i.e. $T/T_g = 0.11$ is rather similar to

Fig. 3. — Susceptibility M/H for fixed temperature as a function of field for the range 50 gauss-5 000 gauss of a AgMn 10.6 % alloy. The data at 4.2 K represent the field cooled value of the magnetization i.e. the low temperature end of the « plateau » apparent in figure 1. In order to display the singular behaviour at $T = T_g$ and below of M/H as $H \rightarrow 0$ we have measured this quantity for positive and negative fields.

that taken right at T_g which of course is a manifestation of the « plateau » of the magnetization *versus* T at low field as shown in figure 1. However the scatter of the points does not allow us to quote a specific exponent for the singular part of the magnetization. We note nevertheless that for $T = T_g$ a linear term is certainly well acceptable in the range 0-500 gauss. In order to improve on this data and to explore more carefully the low field region as suggested from figure 3 we have replaced our magnet by a pair of iron free, water cooled Helmholtz coils capable of ≈ 900 gauss. There, the linearity of the field with the current is granted and the current is determined in 1 part in 10^5 from a fixed resistor in series with the coils. The second series of M/H measurements, shown in figure 4, can be *a priori* analysed in two ways. Either one would be tempted to fit a polynomial dependence of H for each temperature leaving the exponent of the second term of (5)

Fig. 4. — The apparent susceptibility M/H is plotted *versus* H for four temperatures above T_g for AgMn 10.6 % alloy on an enlarged scale. Note the linear behaviour of M/H *versus* H for $T = 37.6$ K and the subsequent rounding of the initial slope at higher temperature. By fitting these points to expression (11) we define a coefficient $b(T)$ for the non linear magnetization which tends to diverge as $T \rightarrow T_g$. The insert shows a log-log plot of b *versus* reduced temperature. The solid lines represent a slope of 1 and 2 respectively.

as a variable : (this approach has been carried out by Barbara *et al.* [16]) or we can force the mean-field equation (5) to be true at the expense of a rapidly diverging non linear susceptibility. To settle this point a comparison was made of the validity of the expansion equation (5) compared to the calculated S.K. exact solution [26]. Indeed this comparison shows that the proximity of an

inflexion point on the M/H versus H plot at low field when $T \rightarrow T_g$ makes the range of validity of the expansion [5] smaller and smaller in field. As an example for $T/T_g = 1.04$ we have checked that equation (5) departs by 2 % from the exact S.K. solution for a field of 300 gauss. We have nonetheless carried out such an analysis by the determination of the parameter $b(T)$ from :

$$M/H = \chi_0(T)(1 - b(T)H^2) \quad (11)$$

in the range $0 < H < 200$ gauss [27].

The parameter $b(T)$ is plotted on a log log scale in the insert of figure 4. By doing so we have avoided taking into consideration the non-diverging temperature dependence of this factor as appears in equation (5). Indeed the particular T coefficients of the non-divergent part of $b(T)$ are model dependent [25]. As shown by the two straight lines in this insert the divergence of $b(T)$ can be represented by $(T - T_g)^{-\gamma}$ with $1 < \gamma < 2$ and a rather large uncertainty. Although more work is needed along this line to have a better determination of γ we note that this range of values together with the value $\delta = 2$ extracted from the definition $q(T = T_g) = H^{2/\delta}$ (by fitting a linear term in the field dependence of M/H versus H) ensures that $-1 < \alpha < -4$. This is consistent with the absence of anomaly for the specific heat at T_g .

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