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Magnetocaloric investigation of (H, T) phase diagram of CuMn spin glass

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Résumé. — L'effet magnétocalorique a été utilisé pour déterminer le diagramme de phase (H, T) d'un verre de spin CuMn. Nos résultats suggèrent une ligne $H_c(T)$ frontière entre l'état paramagnétique et le verre de spins, et une ligne de croisement $H_m(T)$ séparant un paramagnétique de Curie d'un paramagnétique non-Curie. Les résultats obtenus pour $(\partial M / \partial T)_H$ obéissent à une loi d'échelle et conduisent à un exposant critique γ_2 différent de la valeur prévue par la théorie de « champ moyen ».

Abstract. — The magnetocaloric effect has been used to determine the phase diagram (H, T) of a CuMn spin glass. Our results suggest the presence of two lines : the spin glass-paramagnetic boundary line $H_c(T)$ and a crossover line $H_m(T)$ separating a pure Curie paramagnet from a non-Curie paramagnet. The data obtained for the derivative $(\partial M / \partial T)_H$ obey a scaling law and give an estimation of the critical exponent γ_2 different from its « mean field » value.

The nature of the transition from the paramagnetic to the ordered spin glass state remains one of the most interesting and exciting problems in solid state physics. For many years, theoreticians and experimentalists were faced with the same question : is the spin glass a new phase, to be distinguished from a paramagnet resulting from a thermodynamic phase transition, or trivially a metastable state : the result of a progressive freezing phenomenon ?

Recent theoretical work [1] on the infinite range model shows the richness as well as the subtlety of the techniques needed to answer this question. The following new features are of physical interest :

i) the breaking of linear response theory (at least for magnetic properties) in the spin glass phase ;

ii) the regular behaviour of linear response functions (first derivatives of free energy), with a singular behaviour of non-linear response functions (higher order derivatives, when the spin glass state is approached from the paramagnetic side) ;

iii) the importance of the entropy $S(T, H)$ as a key quantity in understanding the physics of spin glass.

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For a long time, attention was focused on the cusp at T_c of the low-field a.c. susceptibility, considered as the signature of a true phase transition at this temperature. The variations of T_c with frequency, observed later in many spin glasses [2], was the first evidence, showing the weakness of the cusp argument.

The presence of hysteresis and slow relaxation of remanent magnetization and of the associated energy [3] are now known to be characteristic properties of spin glasses below the temperature T_c of the susceptibility cusp.

A considerable amount of work has been needed [4] to clarify the situation giving rise finally to the following set of « golden rules » :

- 1) field cooling yields a quasi-equilibrium state (minor instabilities);
- 2) changing a magnetic field, by a small amount, at $T < T_c$ yields non-equilibrium and after-effects (major instabilities);
- 3) for a given procedure of application of the field variations, the induced non-equilibrium state is a function of the reduced variable $T \ln(t/\tau_0)$, of associated time and temperature.

Many illustrations of the validity of these golden rules have been given during the last few years [4]. In particular rule (1) gives the route towards the equilibrium state. Rules (2) and (3) indicate that any measurement obtained with a field variation must, because of its non-equilibrium character, be carefully considered.

In this letter we report the results of magnetocaloric measurements on a CuMn spin glass in a large range of applied magnetic fields ($0 < H < 5$ kG), and temperatures ($1 \text{ K} < T < 10 \text{ K}$). The measurements were made on a 6.65 g, 0.25 at. %, polycrystalline sample of irregular shape. The temperature of the χ_{ac} cusp is $T_c = 3.53 \text{ K}$, and the Curie-Weiss temperature θ obtained from very high temperature susceptibility measurements was $\theta \lesssim 0.1 \text{ K}$.

The basis for the magnetocaloric effect is nothing other than the variation of the temperature of an adiabatically isolated substance with external magnetic field. This variation is related to the magnetization by

$$\left(\frac{\partial T}{\partial H}\right)_S = -\frac{T}{C_H} \left(\frac{\partial M}{\partial T}\right)_H \quad (1)$$

where T is the temperature, H is the magnetic field, C_H is the total specific heat of the sample in a magnetic field and M the magnetization of the sample.

Bearing in mind the Maxwell relation

$$\left(\frac{\partial M}{\partial T}\right)_H = \left(\frac{\partial S}{\partial H}\right)_T \quad (2)$$

it is easy to see that the magnetocaloric effect $(\partial T/\partial H)_S$ gives directly the isothermal variation of the entropy with a magnetic field, as well as the variation of magnetization with temperature at a fixed field. It is interesting to recall that this effect was, in the early days of the century [5], the first experimental evidence for spontaneous magnetization in a ferromagnet, as well as the first check of the mean field theory. Usually, the adiabatic variation of temperature vs. applied magnetic field is a sensitive probe to determine changes in the nature of spin ordering as a function of temperature and applied magnetic field [6]. This is exactly our purpose here.

After a field cooling in a field H_0 , starting from $T \gg T_c$ down to $T \simeq 1 \text{ K}$, we measure the magnetocaloric effect ΔT associated with small and slow variations ΔH around H_0 (typically $\Delta H/H_0 \sim 0.5\%$ at 10% , this variation being established within 5s). The calorimetric measurements were made using apparatus and techniques to be described elsewhere [7]. The sensitivity of the temperature measurement was better than 10^{-6} K . Heating by eddy currents was eliminated in these measurements by taking into account the proportionality of this effect with ΔH^2 , in contrast to the measured magnetocaloric effect ΔT which has the same sign as ΔH .

Figure 1 shows the variation of the magnetocaloric effect ΔT in our sample against temperature at different fields ($\Delta H = 25$ G). Using equation (1) and specific heat data, we can deduce for each H_0 the temperature variation of the derivative $(\partial M/\partial T)_{H_0}$, (the relative variation of the specific heat due to the magnetic field H_0 is less than 2×10^{-3} [8] in the range of interest). In figure 2 we show $(\partial M/\partial T)_{H_0}$ scaled by the cooling field H_0 . The weakness of ΔH seems to allow the use of equation (1) below T_c , all the more as the magnetocaloric effect remains reversible in our experiment. However the verification of the Maxwell relation in the spin glass state must be carried out in the future.

Fig. 1. — Magnetocaloric effect at different values of H_0 . The arrow shows the position of $T_c = 3.53$ K.

Fig. 2. — $(\partial M/\partial T)_{H_0} \cdot H_0^{-1}$ deduced from equation (1) and experimental results of figure 1.

Starting from high temperature, we obtain a Curie regime ($C = 9.21 \times 10^{-5}$ K.emu/g.G) followed by a minimum of $\left(\frac{\partial M}{\partial T}\right)_{H_0}$ showing a departure from a Curie regime and finally the spin glass region with vanishing values. The variation $(\partial M/\partial T)_{H_0} \cdot H_0^{-1}$ vs. T near T_c becomes sharper in vanishing fields H_0 , which suggests a possible abrupt discontinuity at $(H_0 \sim 0, T \sim T_c)$.

For each value of H_0 we find two characteristic temperatures :

- 1) $T_m > T_c$ giving the position of the minimum of $(\partial M/\partial T)_{H_0}$ (i.e. inflexion point of magnetization $M(T)$ at H_0);
- 2) $T_c(H_0) < T_c$ defined by the point where the tangent at the inflexion point of $(\partial M/\partial T)_{H_0}$ intersects the temperature axis.

These characteristic values for each H_0 are plotted in the diagram (H, T) shown in figure 3. The two lines $H_m(T)$ and $H_c(T)$ start from $(T = T_c, H = 0)$ and divide the diagram into three regions : a spin glass phase ($H \sim 0, T \sim 0$), a Curie paramagnet ($T \gg T_c, H \sim 0$) and a non-Curie paramagnet in the intermediate region (see below). The line $H_m(T)$ is a crossover line, dividing the paramagnetic phase into two regions. The first of these is a Curie one, where the magnetization is a function of H/T only. In our case $M \sim CH/T$, because $\theta \simeq 0$ K. In the second region, the magnetization is expected to have a more complicated expression. For instance, at $T \gtrsim T_c$ and $H \sim 0$ we suspect the following [9-11] behaviour :

$$M(T, H) = \chi H - a(T) H^3 + O(H^5) \quad (3)$$

where χ denotes the Curie susceptibility (linear susceptibility) and $a(T)$ is a singular correction : $a(T) \sim (T - T_c)^{-\gamma_2}$ (non-linear susceptibility), which measures the rigidity of the condensed state. $H_m(T)$ is the field value where the singular correction becomes as important as the leading term (in « mean field » theory, $\gamma_2 = 1$).

Fig. 3. — Phase diagram (H, T) determined from magnetocaloric effect. Full line $H_c(T)$ is the boundary line spin glass-non-Curie paramagnet, and dashed line $H_m(T)$ is the crossover line Curie-non-Curie paramagnet.

More generally, $M(T, H)$ is the sum of two contributions : a regular contribution $M_{\text{reg}}(T, H)$ and a singular contribution $M_{\text{sing}}(T, H)$, obeying the universal scaling law [10]

$$M_{\text{sing}}(T, H_0) \sim (T - T_c)^\beta H_0 G[H_0^2/(T - T_c)^{\beta + \gamma_2}] \quad (4)$$

where β is the exponent of the order parameter q . A previous determination of β [12] gives $\beta \simeq 1$, close to the « mean field » value.

We deduce from (4) :

$$\left(\frac{\partial M_{\text{sing}}}{\partial T} \right) / (H_0(T - T_c)^{\beta-1}) = F(x), \quad \text{where } x = H_0^2/(T - T_c)^{\beta + \gamma_2}. \quad (5)$$

Taking into account the first order term in $F(x)$, we deduce two points from the preceding equation :

— the temperature of the minimum of $(\partial M/\partial T)_{H_0}$ (i.e. $(\partial^2 M/\partial T^2)_{H_0} = 0$) depends on the field cooling by the following relation :

$$H_0^2 \approx (T_m - T_c)^{\gamma_2 + 2} / T_m^3 \quad (6)$$

— the amplitude of $(\partial M_{\text{sing}}/\partial T)$ at this minimum is related to T_m through :

$$\partial M_{\text{sing}}/\partial T \approx (T_m - T_c)^{\frac{\gamma_2 + 2}{2}} / T_m^{4.5}. \quad (7)$$

From these two relations and our experimental results at $T = T_m(H_0)$, we determine the critical exponent $3.3 < \gamma_2 < 4$. Admitting (4) as the scaling law and assuming $\beta = 1$ [12], we obtain, with all our results for $T > T_c$, the best fit of $(\partial M_{\text{sing}}/\partial T) H_0^{-1}$ for $\gamma_2 = 3.5$ instead of $\gamma_2 = 1$ as in the « mean field » theory (Fig. 4). This universal behaviour supports our assumption $\beta \simeq 1$.

The line $H_c(T)$ is identified as the boundary line of the spin glass phase. $H_c(T)$ increases rapidly as T decreases from T_c . This critical line varies as predicted by the « mean field » theory in low fields.

$$H_c^2(T) \sim (T_c - T)^3. \quad (8)$$

Fig. 4. — Universal plot of $(\partial M_{\text{sing}}/\partial T)_{H_0} \cdot H_0^{-1}$ as function of $H_0^2(T - T_c)^{-4.5}$ from data obtained at $T > T_c$.

The strong variation of $H_c(T)$ at lower temperatures converges asymptotically towards the behaviour predicted by the « mean field » theory [13] :

$$H_c^2(T) \sim - \ln (T/T_c). \quad (9)$$

More details will be published elsewhere.

To conclude, the magnetocaloric effect was used for the first time to investigate the (H, T) phase diagram of a CuMn spin glass. We have shown the existence of two lines $H_c(T)$ and $H_m(T)$. The physical significance of the line $H_m(T)$ is the crossover between a pure Curie paramagnet, and a non-Curie one. The latter can be characterized by the presence of rigid clusters of spins, giving non negligible corrections to the magnetization law $M(T, H)$. The transition at $H_c(T)$ can be viewed as an abrupt discontinuity of $(\partial M/\partial T)$ in small fields, at $T \lesssim T_c$. It is too early to give the precise order of this transition (first, second or third order). Our results at $T < T_c$ (Eqs. (8), (9)) giving the variation of $H_c(T)$ do not disagree with the « mean field » predictions. For $T > T_c$, our estimation of critical exponent gives rise to a value of γ_2 , different from that of « mean field » theory. The critical region around T_c requires more careful investigation (in progress) in order to have an accurate estimate of the exponents.

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