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Critical measurements in the spin glass CuMn

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Résumé. — Nous avons mesuré l'aimantation $M(H)$ du verre de spins CuMn 1 at. % à des températures comprises entre T_c et 4 T_c et dans des champs allant jusqu'à 7 teslas. Un effort expérimental particulier a été fourni afin d'éliminer au maximum les causes d'erreurs systématiques et de gagner en précision relative. Il a été alors possible d'étudier la dépendance en température des trois premiers coefficients A_1, A_3, A_5 du développement de $M(H)$ en puissances impaires du champ $\left(M = \sum_{n=0}^{\infty} A_{2n+1} \left(\frac{H}{T} \right)^{2n+1} \right)$. A_3 et A_5 augmentent respectivement de 3 et 6 ordres de grandeur quand T varie de 4 T_c à 1,1 T_c. Nous avons pu exprimer ces variations à l'aide d'exposants critiques ($\gamma \simeq 3{,}25$, $\beta \simeq 0{,}75 \pm 0{,}25$) que l'on peut utiliser pour décrire l'ensemble de nos résultats dans des diagrammes universels. Le succès de ces lois d'échelle pèse fortement en faveur de l'hypothèse d'une transition de phase verre de spins à trois dimensions dans les systèmes RKKY.

Abstract. — We present experimental data on the magnetization $M(H)$ of a CuMn 1 at. % in the range $T_c \leqslant T \leqslant 4 T_c$, $0 < H < 7$ teslas. We took special precautions in order to eliminate systematic errors and improve the reliability of the data. It was then possible to study the temperature dependence of the first coefficients A_1, A_3, A_5 in the expansion of the low field magnetization data in terms of odd powers of $H/T \left(M = \sum_{n=0}^{\infty} A_{2n+1} \left(\frac{H}{T} \right)^{2n+1} \right)$, in the range 1.1 $T_c \leqslant T \leqslant 4 T_c$. From the divergence of A_3 and A_5 (which vary over 3 and 6 orders of magnitude respectively in this range), we derive two exponents ($\gamma = 3.25$, $\beta = 0.75 \pm 0.25$) which allow the rescaling of all our data points onto a universal function. The success of the scaling argument is strong evidence in favour of the existence of a phase transition in three dimensions for RKKY spin glasses.

1. Introduction.

The problem of the spin glass transition [1] arose when a pathological cusp was observed in the a.c. susceptibility data at a temperature $T_c(\omega)$ when measuring at frequency ω [2]. For a long time, experimental efforts focused upon the description of this anomaly which indicates the onset, at $T < T_c(\omega)$, of irreversibilities which hamper the determination of an equilibrium response [3]. These irreversibilities scale in T/T_G with

$$ kT_G \sim W_{max}/\text{Ln} \frac{t}{\tau_0} \tag{1} $$

which implies an Arrhenius law and activated processes [4] (W_{max} is an activation energy, t is the time of measurement and τ_0^{-1} is an attempt frequency of the order of 10^{12} s^{-1}). $T_c(\omega)$ however varies much less rapidly than would be expected from equation 1 and its dependence on ω has been interpreted in terms of a Fulcher law (5) :

$$ k(T_c(\omega) - T_F) \sim W_{max}/\text{Ln} \frac{t}{\tau_0}. \tag{2} $$

The present point of view seems to be that the Fulcher law describes the frequency dependence of the temperature of the susceptibility cusp in the high temperature (i.e. high frequency) range only. For low frequencies, the temperature of the cusp reaches a stable limit T_c larger than the limit T_F predicted by the Fulcher law. This temperature T_c is thought to be associated with the spin glass transition. In CuMn, it differs by less than 3×10^{-3} [6] of the value of $T_c(\omega)$ measured at 10 Hz.

The theory has been of a little help for these dynamical studies. With great conceptual and mathematical difficulties a mean field theory (S.K.) was developed [7]. This theory is faced with unexpected complications (such as a negative entropy) in the relevant domain $T < T_c$. At about the same time when these problems were apparently overcome [8] through the introduction of a new concept (an order parameter which is a function), some authors [9] attracted the experimentalists' attention to the fact that the mean field result was in principle correct in the domain $T > T_c(H)$ ($T_c(H)$ is the De Almeida-Thouless instability line [10]). In this range the theory predicts non-trivial results which can be checked in a regime where the experimentalists are not bothered with the irreversibilities and have therefore access to the true thermodynamical response. According to this theory a usual development of the magnetization in terms of odd powers of the field :

$$M = A_1(T) H - A_3(T) H^3 + A_5(T) H^5 ... \quad (T > T_c),$$

(3)

yields in spin glasses a first order susceptibility A_1 which follows the standard Curie law ($A_1 = C/T$).

But all the higher order susceptibilities A_3, A_5, ... diverge at T_c where the magnetization becomes very anomalous, its development involving both odd and even powers of the field H :

$$M = A_1(T_c) H - A_2' H^2 + A_3' H^3 ... \quad (T = T_c). \quad (4)$$

More general expressions can be phenomenologically derived [11] which allow the possibility for the exponents which control the behaviour of M to differ from those expected for the mean field theory, while obeying the usual sum rules :

$$M - A_1(T) H \sim H(T - T_c)^\beta F\left[\frac{H^2}{(T - T_c)^{\gamma + \beta}}\right] \quad (5a)$$

$$\sim H^{1 + 2/\delta} \Phi\left[\frac{H^{2 - 2/\delta}}{(T - T_c)^\gamma}\right] \quad (5b)$$

where $\Phi(x = \infty) = 1$ (i.e. for $T = T_c$). In the S.K. model, $\beta = \gamma = 1$ and $\delta = 2$. The comparison of these predictions with the experimental data has been very interesting. In the insulating spin glass $Ti_{1-x}V_xO_3$, Chikazawa et al. [12] observed an anomalous enhancement of the second and third order susceptibilities (A_3 and A_5) on approaching T_c by

Table I. — *The values of the critical exponents in* $CuMn$ *(columns 2 to 6) as determined experimentally are compared with expectation from the S.K. theory (column 7). In the lines 3 and 4, we specify the actual range of* H/T_c *and* T/T_c *values utilizable in this determination. The numbers which are underlined have been determined either directly (Nagata et al., our work) or through a best fit to the scaling law. The other numbers are deduced from the application of the sum rules which relate the different exponents. The values* δ' *and* δ'' *are derived as explained in the text. The maximum value of the field for which the relation* $M - \chi_0 H = H^{1 + 2/\delta'}$ *remains valid is indicated in brackets.*

Authors	Our work	Barbara et al.	Berton, Odin et al.	Odin	Nagata et al.	S.K.
Reference		15	16	26	20	7
System	$\underline{Cu}Mn$ 1 at. %	$\underline{Cu}Mn$ 4.6 %	$\underline{Cu}Mn$ 0.25 at. %		$\underline{Cu}Mn$ 2.02 %	theory
$\frac{H}{T_c}$ Oe/K	20-7 000	4-400	70-1 500	30-70	0.6	
$\frac{T}{T_c}$	1.10-4	1.03-2	1.04-2	1.007-1.04	0.90-0.99 ($T < T_c$)	
γ	$\underline{3.25 \pm 0.10}$	$\underline{3.4 \pm 0.4}$	$\underline{3.65 \pm 0.35}$			1
β	$\underline{0.5} - ①$	1.1 ± 0.2	$\underline{1}$		$\underline{1.05 \pm 0.07}$	1
δ' δ''	$\frac{5.7 \pm 0.5}{(20 \text{ kOe})}$ $\underline{4.4 \pm 0.25}$	$\underline{4.15 \pm 0.15}$	4.65 ± 0.35			2
α	$- 2.75 \pm 0.7$	$- 4.08 \pm 0.25$	$- 3.65 \pm 0.35$			$- 1$

studying at 3 ω and 5 ω the amplitude of the magnetic signal associated with a field monitored at frequency ω. In AgMn, Bouchiat and Monod [13, 14] showed, using an M/H vs. H diagram, that the magnetization is not described by the same analytic development at T_c and above T_c. Barbara et al. [15] using a squid magnetometer and Berton et al. [16] studying the magnetocaloric effect have derived scaling laws for the CuMn and the AlGd systems involving critical exponents which are compared in table I with the values predicted for the mean field S.K. model. The difficulties which explain the scarcity of quantitative results in such a controversial field are seen at a glance in figures 1, 3 and 5. The M vs. H data in CuMn 1 at. % shown in figure 1 appear rather trivial to the eye and suggest paramagnetism. But high field deviations from strict paramagnetic behaviour are visible on the M vs. H/T plot of figure 3. These deviations themselves do not suggest any pathological behaviour, but a plot of $M/\chi H$ vs. H^2 of the same data (see Fig. 5) directly shows the divergence of the coefficient of H^3 (the initial slope in this diagram, which is A_3 (Eq. 3), varies by several orders of magnitude between 4 T_c and 1.1 T_c). Notice on this figure that, in order to determine with confidence the amplitude of the coefficient of the H^3 term, we need to analyse deviations which represent a fraction of $\sim 10^{-2}$ of the total magnetization. A very good control of the magnetization, the field, and the temperature is thus necessary in traditional magnetization measurements [17].
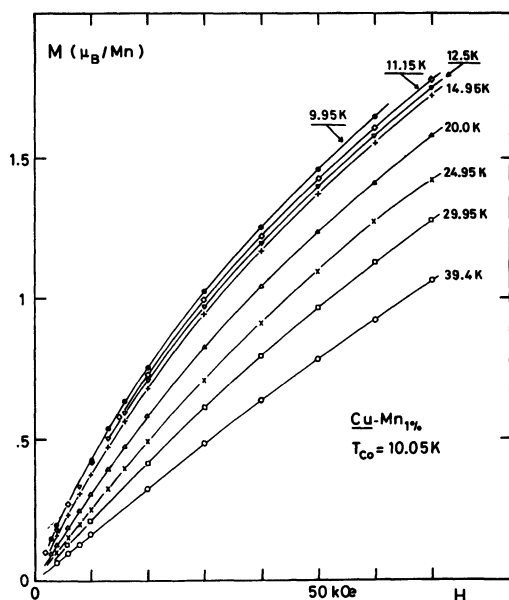

Fig. 1. — The magnetization M of a CuMn$_{1\,at.\,\%}$ vs. the field H at different temperature $T \geqslant T_c$.

We will first describe the experimental technique (chapter 2) which we have developed using our classical apparatus (extraction technique) in order to obtain data sufficiently accurate to investigate the character of the deviations from the Curie law. The experimental data are presented in chapter 3 to illustrate in a pedagogical way the main features of the transition. A scaling law is derived and critical exponents are determined in chapter 4. The data are discussed in chapter 5.

2. Technical aspects.

2.1 SAMPLE PREPARATION. — Three ingots of CuMn (1 at. %, 5 at. %, 8 at. %) were prepared by melting the constituants in a semilevitation induction furnace and by quenching the melt in a rotating cylindrical copper mold 7 mm in diameter.

Samples 20 mm long were then machined out of the ingots. They were annealed one hour at 1 200 K in Ar + H_2 atmosphere and then cooled slowly to room temperature.

2.2 MAGNETIZATION (AND SUSCEPTIBILITY) MEASUREMENT CIRCUIT. — The magnetization was measured by an extraction method : the sample is moved between the centres of two counter-wound copper coils ($\sim$ 20 000 turns) in a constant magnetic field. The magnetization is obtained by integrating the amplified emf induced by the flux variation during the extraction. As we use a very reliable galvanometric amplifier (gain 10^3-10^4) and a high resolution integrating digital voltmeter (IDVN 521) we only have to deal with the problems associated with any measurement of low level d.c. voltages. An electrical and thermal shield protects the room temperature circuit : this circuit is composed of a voltage divider made with a few precise ($\pm 10^{-4}$) and stable (5 ppm/K) low noise resistances, a Tinsley switch, and the galvanometric amplifier ; a low thermal emf solder was used for the contacts. The resistance of the pick-up coils and of the leads were measured for each data point : this allows to accurately determine the gain of the circuit in the conditions (external field, temperature, He level...) of the experiment. 20 to 30 measurements were performed for each data point in order to correct for stray inputs and eliminate statistical errors. The absolute residual error due to the effect of the mechanical vibrations in the presence of the field and to the effect of the thermal e.m.f., is less than 10^{-4} e.m.u. The resolution is of the order of 10^{-4}.

The pick up coils can also be used as the secondary signal of an a.c. susceptibility circuit. In this circuit, the primary signal is produced by an additional copper coil wound inside the superconducting coil which gives the main static field. χ_{ac} is then determined (with the same conditions of thermometry as for the magnetization measurement), as the difference between the two signals which are observed at the two positions of the extraction.

2.3 MAGNETIC FIELD. — The external field (0-80 kOe) is produced by a multifilamentary Nb-Ti super-conducting coil. This device is very stable in the persistent mode and shows only small hysteresis. However, in order to avoid distortion of the spatial dependence of the field due to the hysteresis, the present experiments have all been performed in the first magnetization regime of the coil. A Hall generator is used to measure the lower values of the field and to control its stability over the whole range of field values. The absolute error is less than 0.3 Oe.

We have taken into account the effective local residual magnetic field due to the metallic surroundings and which includes the earth field component along the z-axis of our magnetometer. Its value is 0.7 Oe. It is easily determined in the low field, high temperature, paramagnetic limit when the magnetization of our sample is very accurately proportional to H (to better than 10^{-4}). A plot of $\frac{H}{M}$ vs. $1/M$ then yields a straight line whose intercept with the H/M axis gives χ^{-1} and whose slope gives the effective value of the residual field along the z-axis.

2.4 CRYOGENICS. — The sample together with a capacitance, a carbon resistor and a heater is tightened by a braid made of copper wires which is connected by a heat leak to the helium bath. The capacitance is the field independent reference of our regulation which ensures temperature stability within 1 mK.

All these devices are contained in an epoxy high vacuum cell which is slowly extracted pneumatically. The contribution of the cell and of the addenda, which in low fields can represent as much as 1 % of the sample contribution, was determined in a separate experiment in the same conditions of field and temperature, and was subtracted from the data.

3. Experimental results.

With the precautions described in chapter 2, it has been possible to match the difficult conditions defined in chapter 1 on a properly chosen sample in an appropriate range of temperature, and to obtain data which compete with those obtained with more sophisticated techniques. Most of these data refer to a 1 at. % CuMn sample. This is a typical spin glass system and the concentration is small enough to avoid most of the complications which would be associated with clustering or with the occurrence of a too large chemical short range order. The concentration is high enough, though, to ensure a sizeable signal in a range of normalized T/T_c and H/T_c values larger than those used in previous experiments (see Table I).

We have chosen to identify and measure the temperature dependence of the three first terms A_1, A_3 and A_5 of the expansion of the magnetization in terms of the field. In general two exponents determine com-

pletely the behaviour of thermodynamical quantities near a transition. In our case, no information can be obtained from A_1 which is trivial, hence the necessity to measure A_3 and A_5. With two exponents known it is in principle possible to directly check a scaling law. Our method permits a total control of what is being done since we compare terms which *have to* correspond to each other in the scaling. This is not totally obvious when, as in previous work, a best fit involving many coefficients is matched to a scaling law which does not present very dramatic variations in terms of the scaling variable.

The price which we have to pay is a restriction of the range of the measurement : our accuracy did not allow us to perform this analysis (i.e. the identification and measurement of A_3 and A_5) for T/T_c ratios smaller than 1.1. Admittedly, we thus loose the range which would have been the most promising with a traditional phase transition. However a surprising feature of the spin glass transition which is evident from the data now available is the unexpectedly large range of fields and temperature over which a scaling behaviour is observed.

All the results which we will discuss in the following correspond to the magnetic contribution of the Mn atoms. This contribution was deduced by subtracting the diamagnetic contribution of the Cu matrix from the total magnetization ($M_{\text{matrix}}/\text{e.m.u.}/g = -0.88 \times 10^{-7}$ H/Oe [8]).

3.1 PRESENTATION OF THE DATA (QUALITATIVE APPROACH). — The a.c. susceptibility cusp occurs at $T_{c0} = 10.05 \pm 0.05$ K in our 1 at. % sample (see Fig. 2). The initial d.c. susceptibility $\chi_0 = M/H$, measured between T_c and 4 T_c, follows quite accurately a Curie Weiss law in this range. We have $\chi_0 = C/(T + T_0)$. The value of the effective moment μ which we can deduce from the Curie constant ($C = N\mu^2/3\,k_B$) is equal to 5.45 μ_B/Mn, i.e. slightly larger than usually accepted values [19-21]. The presence of a negative Curie temperature T_0 ($T_0 \simeq -0.8$ K for our sample) is not predicted by the
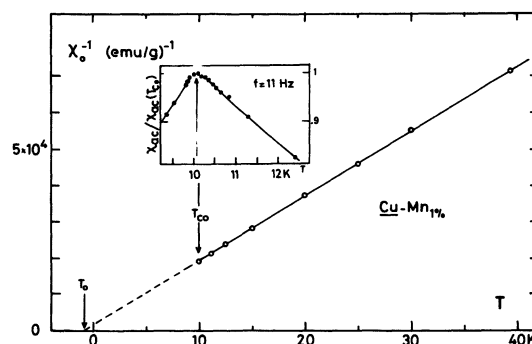

Fig. 2. — The reverse initial susceptibility χ_0^{-1} is a linear function of the temperature in the T_c to 4 T_c range. The a.c. susceptibility data $\chi_{ac}(T)/\chi_{ac}(T_c)$ shown in the insert, exhibit the well known cusp at the temperature T_c.

mean field theory when positive and negative inter-actions are present with exactly the same probability $(J_0/J = 0)$. This, by the way, indicates a defect of the theory and not of our sample. Experiments on very dilute systems interacting through the RKKY inter-action show that, in the limit of vanishing concentra-tions (i.e. when we can insure an exact balance between positive and negative interactions), T_0 is negative and of the order of T_{co}, both quantities being proportional to the concentration [21].

The origin of the negative value of T_0 is quantum mechanical [22] and associated with the fact that μ_{eff}^2 changes from $S(S + 1)$ to S^2 for increasing values of the moments of the clusters which are responsible for the paramagnetic susceptibility (see chapter 5). It is therefore not surprising that the classical S.K. theory does not account for it. Indeed T_0 depends to some extent upon the range of temperature used for its determination [21]; i.e. the Curie Weiss law is itself an approximation valid over a restricted range of temperature. However, for increasing concentrations, an unbalance between positive and negative inter-actions occurs when the average distance between impurities becomes of the order of the interatomic distance. This effect favours the type of order which ultimately exists in the magnetic material (i.e. ferro-magnetism in Fe, antiferromagnetism in Mn) and occurs at about the percolation threshold ($\sim 15 \%$ in AuFe). However, at much lower concentrations, short range order creates an unbalance between the statis-tical occupation of the 1st and 2nd neighbour shells which favours the ferromagnetic 2nd neighbour inter-actions in the alloys of Mn with the usual noble metals [14, 23]. As a result T_0 changes from $- \alpha c$ to $+ \alpha c$ in those systems with an accidental cancellation at a concentration which is of the order of 1 at. % in CuMn (and much lower in AgMn and AuMn). This is the reason for the small magnitude of $\overline{T_0}$ with respect to T_c in our sample. As far as we know, the magnitude and the sign of T_0 have no influence on the spin glass transition itself.

We will in the following take account of these facts by assuming that the Curie Weiss law experimentally determined in our sample describes the paramagnetic regime. In practice, therefore, the reference paramagnet to which we compare our data follows a $B_s(\mu H/kT^*)$ law with $T^* = T_0 + T$ (B_s is the Brillouin function).

The logic of our argument therefore leads us to use diagrams where our magnetization data enter always in combination with the reduced variable H/T^*. We therefore introduce instead of (3) the following deve-lopment of the magnetization :

$$\frac{M}{M_{sat}} = \sum_{n=0}^{\infty} a_{2n+1} L_{2n+1} x^{2n+1} \qquad (6)$$

or alternatively $M/\chi_0 H = a_1 - \frac{1}{15} a_3 x^2 + \frac{2}{305} a_5 x^4 + \cdots$

where $M_{sat} = N\mu$ (N is the total number of Mn atoms),

$$x = \frac{\mu H}{kT^*}, \quad \chi_0 = \frac{N\mu^2}{3 kT^*} \text{ and } L_1 = \frac{1}{3}, \quad L_3 = -\frac{1}{45},$$

$L_5 = \frac{2}{915}, \cdots, L_{2n+1}$ are the numerical coefficients which enter in the series expansion of the Langevin function (in the case of a paramagnet : $a_1 = a_3 = a_5 = a_{2n+1} = 1$). Since, in such reduced diagram, the refe-rence paramagnet would appear as a unique curve, all the deviations apparent for example in the figures 3, 4
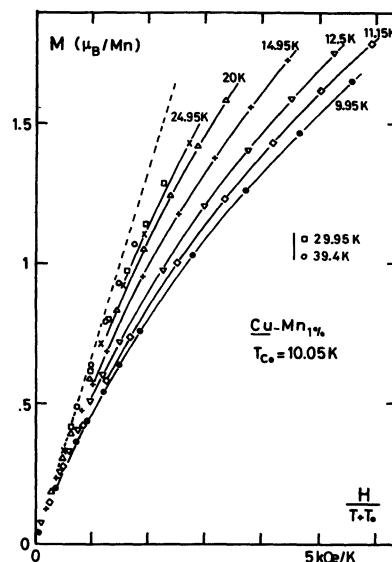

Fig. 3. — Plot of the magnetization M vs. $\frac{H}{T^*}$ showing the deviations to a simple paramagnetic behaviour (dashed line).
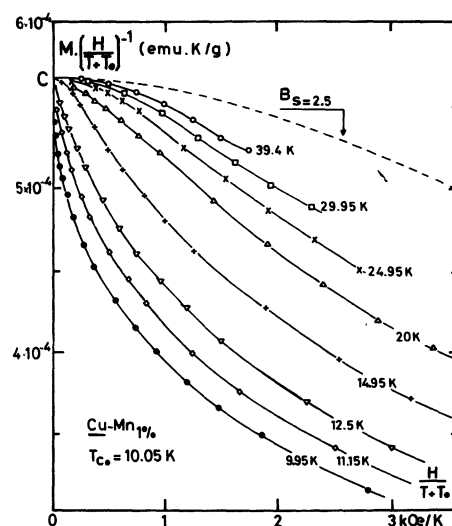

Fig. 4. — Plot of $M \left/ \left(\frac{H}{T^*}\right)\right.$ vs. $\frac{H}{T^*}$. The data depart from the Curie constant C which is obtained in the limit of small H/T^* values. The dashed curve represents the Brillouin behaviour expected for a paramagnet. Notice the change in initial curvatures which is observed on lowering T towards T_c.

and 5 are to be attributed to the spin glass effect. It is already obvious on the M vs. H/T^* plot of figure 3 that deviations from paramagnetic behaviour become more and more apparent for a given H/T^*, when T approaches T_{co}. The singular character of these deviations is emphazised in the $M(H/T^*)^{-1}$ vs. H/T^* plot of figure 4. The isothermal curves, there, sweep the plane from a high temperature limit fixed by the theoretical Brillouin function to the measured limit at T_c. Well above T_c the curves exhibit initially the quadratic behaviour which is expected for a magnetization which can be developed in terms of odd powers of the field. But the range in H/T^* over which this initial quadratic behaviour is dominant shrinks on lowering the temperature, and ultimately vanishes when T reaches T_c. The dramatic character of the variation of the coefficient a_3 of the $(H/T^*)^3$ term in the development of $M(H)$ is best seen in the figure 5 where $M(H/T^*)^{-1}$ is plotted vs. $(H/T^*)^2$. This coefficient is thus the slope of the initial straight line which is seen in our scale to vary from quasi-horizontal to quasi-vertical near T_c. At the same time the strong curvatures which appear stress the necessity to include H^5 terms and higher powers of H in the expansion of M in order to account for the magnetization in a given H/T^* range when T approaches T_{co}.
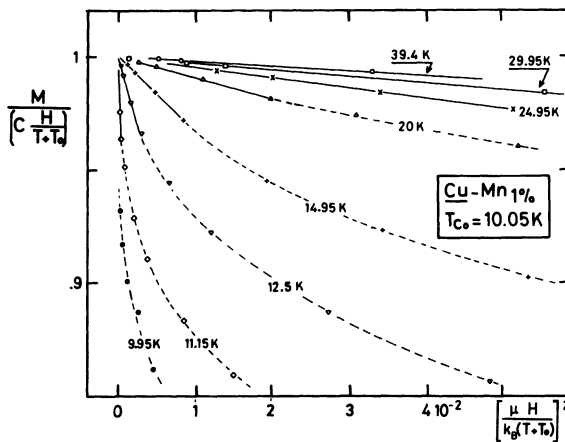

Fig. 5. — The same data as in figure 4 are represented in a $M\left(\dfrac{CH}{T^*}\right)^{-1}$ vs. $\left(\dfrac{\mu H}{kT^*}\right)^2$ diagram. The slope of the initial straight line is the coefficient of $(H/T^*)^3$ in the development of the magnetization. It is seen to vary by orders of magnitude in our range of temperature.

3.2 COMMENTS ABOUT THE ACCURACY. — The sequence of figures 3, 4 and 5 illustrates the somewhat obvious fact that the lower order terms may be determined more precisely if higher order terms are taken into account. For example, figure 3, which directly presents the initial susceptibility, allows a less accurate determination of this quantity (the slope of the envelope) than the other two figures. In a M/H vs. H^2

plot (Fig. 5), we benefit from a linear extrapolation from measurements in high fields (where the relative error is less important) which allows the determination of χ_0 to within a relative error of 10^{-3}. This admittedly could be done with a linear regression using a computor. But, unless we are very cautious in this procedure, we can easily lose knowledge of the range over which a given limited development is valid and of the way in which this range varies on approaching T_c. Thus the term in H^3 is dominant up to 40 kOe at 40 K and only up to 400 Oe at 11.15 K. Thus, also, figures 3, 5 and 7 show, in a pedagogical way, how more and more terms in H/T^* are requested to fit the magnetization in a given window of H/T^* values when we approach T_c. This suggests that, at T_c, it may be necessary to use the total infinite series with diverging coefficients in order to fit the $M(H)$ curve over any finite range of H. This is an essential result of the S.K. equations which state that *both* even *and* odd terms in H are requested in the development of the magnetization *at* T_c.

Benefiting from the accuracy gained on χ_0 we can go a step further and plot $(1 - M/\chi_0 H)/H^2$ vs. H^2 to obtain a straight line of intercept a_3 and slope a_5 (the coefficient of the $(H/T^*)^5$ term). Small adjustments of χ_0 (within a fraction of 10^{-3}) will permit us to reach the best linearity. a_3 is known within a few percent and a_5 within 30 % (see Table II). A normalization to a_3 of these data in a $(1 - M/\chi_0 H)/a_3(H/T^*)^2$ vs. $a_3(H/T^*)^2$ plot allows us to present these straight lines (Fig. 6) with intercept 1 and slope $a_5/(a_3)^2$ by making use of the fact that a_3 varies by 3 orders of magnitude when a_5 varies by 6 orders of magnitude.

3.3 QUANTITATIVE ANALYSIS. — The values of the parameters a_1, a_3 and a_5 determined for the different temperatures of measurement have been compiled in table II and their variations with temperature are shown in figure 7 (right hand side). On this figure it is seen that a_1 keeps a constant value (equal to one by definition of the effective moment μ (5.45 μ_B)) as expected for paramagnetic behaviour. On the contrary a_3 and a_5 deviate very rapidly from their paramagnetic value of one on decreasing the temperature. They vary by 3 and 6 orders of magnitude respectively between 4 T_c and 1.1 T_c, and seem to diverge on approaching T_c at a rate which cannot be accounted for by any power law of the temperature in this range (see insert). Notice on the same figure (left-hand-side) that a_5 can be expressed as a power law of a_3 : $a_5 = a_3^{2.22 \pm 0.05}$ suggesting that the same variable governs the divergences of both a_5 and a_3.

The log a_3 vs. log a_5 line extrapolates to a point where *both* a_3 and $a_5 = 1$ on the high temperature side. This is consistent with the fact that the high temperature limit of the magnetization should be that of a paramagnet, i.e. it should be represented by the Langevin function for which $a_1 = a_3 = a_5 = \cdots 1$ (see Eq. 6).

Table II. — *Temperature dependence of the first coefficients* a_1, a_3, a_5 *involved in the series expansion of the magnetization M in terms of odd powers of* H/T^* :

$$\frac{M}{M_{sat}} = \sum_{n=0}^{\infty} a_{2n+1} \; L_{2n+1} \left(\frac{\mu H}{kT^*}\right)^{2n+1}.$$

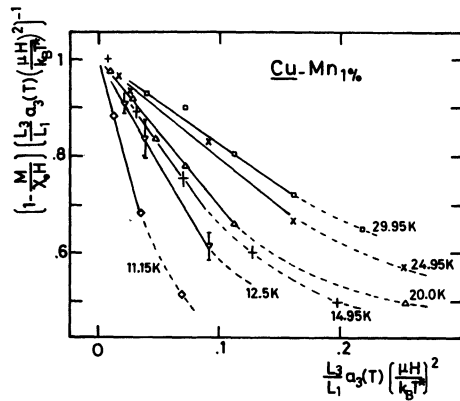| T	$\dfrac{T}{T_c}$	a_1	a_3	a_5
11.15 K	1.11	1	1 970 ± 300	$(0.38 \pm 0.22) \times 10^8$
12.5 K	1.24	1	210 ± 20	$(1.44 \pm 0.22) \times 10^5$
14.95 K	1.49	1	58 ± 3	5 700 ± 200
20.0 K	1.99	1	13.6 ± 0.6	235 ± 15
24.95 K	2.48	1	7.8 ± 0.2	100 ± 10
29.95 K	2.98	1	4.75 ± 0.15	28 ± 3
39.4 K	3.92	1	3.50 ± 0.10	

Fig. 6. — $(1 - M/\chi_0 \, H) \left[\dfrac{L_3 \, a_3}{L_1}\left(\dfrac{\mu H}{kT^*}\right)^2\right]^{-1}$ vs. $\dfrac{L_3}{L_1} a_3 \left(\dfrac{\mu H}{kT}\right)^2$.

On this plot, the slope of the initial straight line is proportional to $a_5.(L_3 \, a_3)^{-2}$. The use of $L_3 \, a_3$ units allows us to present on a single figure data which vary by 6 orders of magnitude.

4. Comparison with theories at $T > T_c$

4.1 CRITICAL EXPONENTS FOR THE FIRST THREE TERMS OF THE DEVELOPMENT.

— Our data illustrate in a spectacular way many of the predictions of the S.K. model (Eq. 3). Namely, while the first order susceptibility is that of a paramagnet, the terms of higher order in H in the development of the magnetization (a_3 and a_5) seem to strongly diverge (see Fig. 7) at the same temperature T_c. The high temperature limit of these coefficients is the paramagnetic value as predicted. However the observed divergence of a_3, can obviously not be accounted for by the $(T - T_c)^{-1}$ term predicted by the mean field theory which would also predict a_5 to vary like $(a_3)^3$ near T_c, in disagreement with the observed $a_3^{2.22+0.05}$ variation.
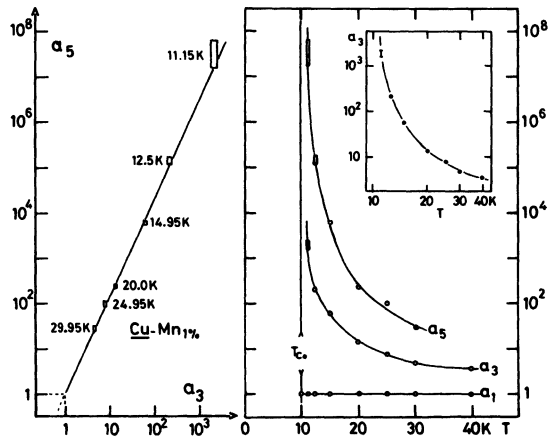

Fig. 7. — The coefficients a_1, a_3, a_5 involved in the series expansion of the magnetization in terms of $x = \dfrac{\mu H}{kT^*}$

$$\left(\frac{M}{M_{sat}} = L_1 \, a_1 \, x + L_3 \, a_3 \, x^3 + L_5 \, a_5 \, x_5 \ldots \right)$$

are represented vs. the temperature in a semi-log plot on the right-hand-side of the figure. On the left hand side, Log a_5 is represented in terms of Log a_3 at the same temperature. Notice that the data extrapolate to a point where a_3 and a_5 are both equal to 1 and which should be associated to the high temperature paramagnetic limit. For $T \geqslant 1.1 \, T_c$, we have $a_5 = a_3^{2.25 \pm 0.05}$. The log a_3 vs. Log T plot shown in the insert stresses the failure of any attempt to represent the data which a power law of the temperature.

As suggested by Suzuki and Chalupa [11], we are tempted to search for empirical exponents able to fit the a_3 variation. We then seek a function $a_3(T/T_c)$ such that $a_3(T/T_c) \sim \left(\dfrac{T_c}{T - T_c}\right)^{\gamma}$ in the vicinity of T_c. Unlike previous authors we are not very close to T_c. Figure 7, where the paramagnetic limit $a_3 = a_5 = a_1 = 1$ appears very close to the data points, suggests that we might, even, be in a situation more appropriate to

check the high temperature behaviour of the function $a_3(T/T_c)$ rather than its divergence at T_c. We request in this limit $a_3\left(\dfrac{T}{T_c}\right) \sim \left(1 + \gamma' \dfrac{T_c}{T} - \cdots\right)$. Restricting the expansion to its first two terms we write :

$$a_3(T/T_c) \sim \left(1 - \gamma' \frac{T_c}{T}\right)^{-1} \sim \left(\frac{T}{T - T_c}\right)^{\gamma'} \quad (7)$$

which suggests a plot of $\mathrm{Log}\, a_3$ (and of $\mathrm{Log}\, a_5$) in terms of $\mathrm{Log}\, \dfrac{T - T_c}{T}$. The figure 8 shows the linearity which is obtained for both our a_3 and a_5 data if we take for T_c the measured temperature $T_{c0} = 10.05$ K of the susceptibility cusp. We find $\gamma' = 3.25 \pm 0.10$, and the exponent derived for the $a_5\left(\dfrac{T}{T - T_{c0}}\right)$ dependence is 2.22 γ' (in agreement with Fig. 7). The fact that the truncated expansion [7] can fit the data reasonably well from $T \to \infty$ down to 1.1 T_c is already surprising. More surprising even is the fact that the value which we derive for γ' is the same, within the experimental error, as the value γ that Odin *et al.* have derived in the same system using the usual linear scaling variables $(T - T_c)/T_c$ and H/T_c (rather than $(T - T_c)/T$ and H/T) in the close vicinity of T_c (down to 1.007 T_c) [26].
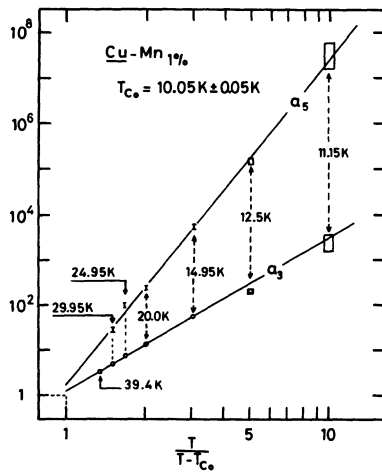

Fig. 8. — Log-Log plot of a_3 and a_5 vs. $\dfrac{T}{T - T_c}$. We have

$$a_3 = \left(\frac{T}{T - T_c}\right)^{3.25 \pm 0.05}, a_5 = \left(\frac{T}{T - T_c}\right)^{7.25 \pm 0.05}.$$

Of course near and below 1.1 T_c the non-linear variable $\dfrac{T - T_c}{T}$ cannot be distinguished from the usual linear variable $\dfrac{T - T_c}{T_c}$. The use of the former however apparently allows the extension of the validity of the simple scaling law from temperatures within 7/1 000 of T_c to the highest temperatures.

This behaviour contrasts markedly with that observed in ordinary phase transitions where scaling arguments are valid only in a very restricted range near T_c. In any case, from our data only, the divergence of well-identified coefficients of the expansion of the magnetization in terms of H is well represented by a single power law over several orders of magnitudes (3 for a_3, 6 for a_5). Inasmuch as this fact can be considered as a sufficient proof for the existence of a phase transition, we surmise the existence of a phase transition in spin glasses.

4.2 HIGHER ORDER TERMS IN THE EXPANSION OF M. TOWARDS A SCALING LAW. — We have written the Taylor expansion of the magnetization in the form [6]

$$\frac{M}{M_{sat}} = \sum_{n=0}^{\infty} a_{2n+1}\, L_{2n+1}\left(\frac{\mu H}{kT^*}\right)^{2n+1} \quad (6)$$

where L_{2n+1} is the coefficient of order n of the Taylor expansion of the Langevin function. We have found that the multiplicative coefficients a_{2n+1}, which are all equal to 1 in the limit of high temperatures, diverge (at least for $n = 0$, 1 and 2) like $\left(\dfrac{T}{T - T_c}\right)^{\gamma_{2n+1}}$ with $\gamma_1 = 0$, $\gamma_3 = 3.25$, $\gamma_5 = 7.5 = 2.22\,\gamma_3$. These coefficients were derived in a restricted domain of field and temperature but we may wonder whether it is possible from the knowledge of these three terms to guess something about the behaviour of those which follow. The logic of our argument suggests that we should iterate the procedure developed in 4.1 in order to write the higher order terms in the form :

$$a_{2n+1} = \left(\frac{T}{T - T_c}\right)^{\gamma_{2n+1}} = \frac{1}{|t|^{\gamma_{2n+1}}}$$

where

$$t = \frac{T - T_c}{T}$$

and the magnetization therefore as

$$\mathfrak{R}(H^2) = 1 - \frac{M}{M_{sat}\, L_1 \dfrac{\mu H}{kT^*}} = 1 - \frac{M}{\chi_0\, H} =$$

$$= - \sum_{n=1}^{\infty} \frac{L_{2n+1}}{L_1} \frac{1}{t^{\gamma_{2n+1}}}\left(\frac{\mu H}{kT^*}\right)^{2n}. \quad (8)$$

We have defined $\chi_0 = M_{sat}\, L_1 \dfrac{\mu}{kT^*} = \dfrac{1}{3}\dfrac{N\mu^2}{kT^*}$ and have introduced the non-linear scaling variable $t = \dfrac{T - T_c}{T}$. We have also introduced the quantity $\mathfrak{R}(h) = \dfrac{\chi_0\, H - M}{\chi_0\, H}$. In the scaling argument of Susuki and Chalupa, this quantity plays the role that the magnetization plays in the standard argument of

Widom-Kadanoff [21] for usual phase transition while $h = H^2$ plays the rôle of the field associated with the order parameter $q = \langle \mu^2 \rangle$. In other words, we have for $\Re(h)$ the classical expression of Widom-Kadanoff :

$$\Re(h) = |\varepsilon|^\beta \, f\left(\frac{h}{\varepsilon^{\gamma+\beta}}\right) \qquad (9)$$

yielding

$$\Re(H^2) = |\varepsilon|^\beta \, f\left(\frac{H^2}{\varepsilon^{\gamma+\beta}}\right)$$

with $\varepsilon = \dfrac{T - T_c}{T_c}$ and $f(x) = \alpha_1 x + \alpha_2 x^2 + \cdots$, i.e. as in equation 5. The exponents α, β, γ, δ which govern the behaviour of the thermodynamic quantities near T_c $(C_H = \dfrac{1}{|\varepsilon|^\alpha}; \Re = (-\varepsilon)^\beta (T < T_c); \dfrac{d\Re(h)}{dh} = \dfrac{1}{|\varepsilon|^\gamma} = a_3 (T > T_c); H^2 = h = |\Re(h)|^\delta \text{ sign } \Re(T = T_c))$ stay related by the classical equalities :

$$\alpha + 2\beta + \gamma = 2$$
$$\alpha + \beta(1 + \delta) = 2.$$

We can develop equation 9 as :

$$\Re(h) = 1 - \frac{M}{\chi_0 H} = \sum_{n=1}^{\infty} \alpha_n \left(\frac{T - T_c}{T_c}\right)^{\beta - n(\beta + \gamma)} \times$$
$$\times \left(\frac{\mu H}{kT_c}\right)^{2n}. \qquad (10)$$

The success of the expansion of a_3 and a_5 in terms of

$\dfrac{T - T_c}{T}$ suggests as a working hypothesis the possibility that equation 10 $\left(\text{with its linear variables } \dfrac{T - T_c}{T_c}\right.$ and $H/T_c)$ could be an approximation valid near T_c of our expression 8 in terms of the non linear variable $\dfrac{T - T_c}{T}$ and H/T^*. Equating 10 and 8 near T_c imposes :

$$\gamma_{2n+1} = n(\beta + \gamma) - \beta \qquad (11a)$$
$$\alpha_n = -\frac{L_{2n+1}}{L_1}. \qquad (11b)$$

The two exponents β and γ which completely determine the magnetization can be deduced from the knowledge of γ_3 and γ_5 using equation 11. We have

$$\gamma = \gamma_3 = 3.25 \pm 0.10$$
$$\beta = \gamma_5 - 2\gamma = 0.75 \pm 0.25 \qquad (12)$$
$$\text{hence } \alpha = -2.7 \pm 0.6 \text{ and } \delta = 5.8 \pm 1.5.$$

Within our hypothesis, i.e. on substituting the variables H/T^* and $\dfrac{T - T_c}{T}$ for the variables $\dfrac{H}{T_c}$ and $\dfrac{T - T_c}{T_c}$, our version of equations 5a and 5b becomes respectively :

$$\frac{\Re}{t^\beta} = \left(1 - \frac{M}{\chi_0 H}\right)\left(\frac{T}{T - T_c}\right)^\beta = f(X) \qquad (13a)$$

with $X = \left(\dfrac{T}{T - T_c}\right)^{\frac{\gamma+\beta}{2}} \dfrac{\mu H}{kT^*}$, $\chi_0 = M_{sat} L_1 \dfrac{\mu}{kT^*}$
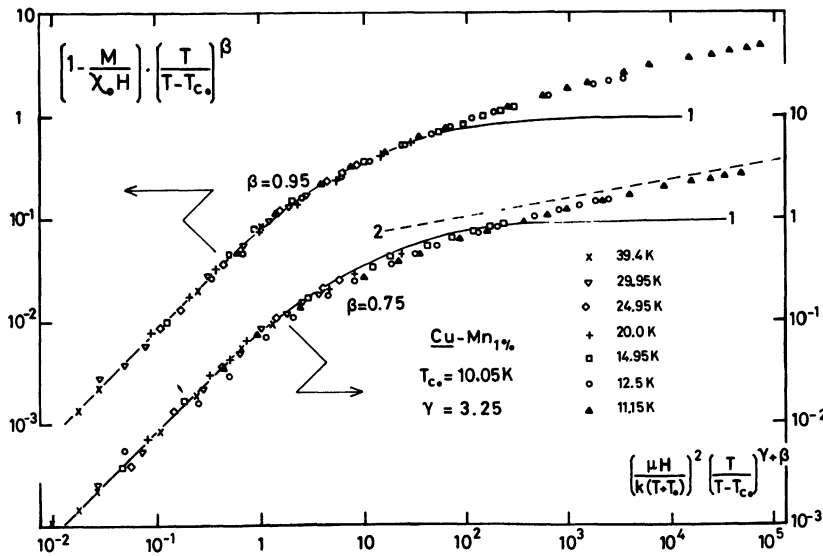

Fig. 9. — Universal plot of $[1 - M/\chi_0 H] \left(\dfrac{T}{T - T_c}\right)^\beta$ vs. $\left(\dfrac{\mu H}{kT^*}\right)^2 \left(\dfrac{T}{T - T_c}\right)^{\gamma+\beta}$ showing the success of the scaling argument for all our data points (H up to 70 kOe, T up to 4 T_c) with $\gamma = 3.25$ and $\beta = 0.75$ as deduced from the temperature dependence of a_3 and a_5. The continuous curve 1 and the dashed curve 2 give the asymptotic behaviours predicted by equations 14 and 16 respectively. For $\beta \simeq 1$, we obtain a better fit to the expression 14 and an improved scaling.
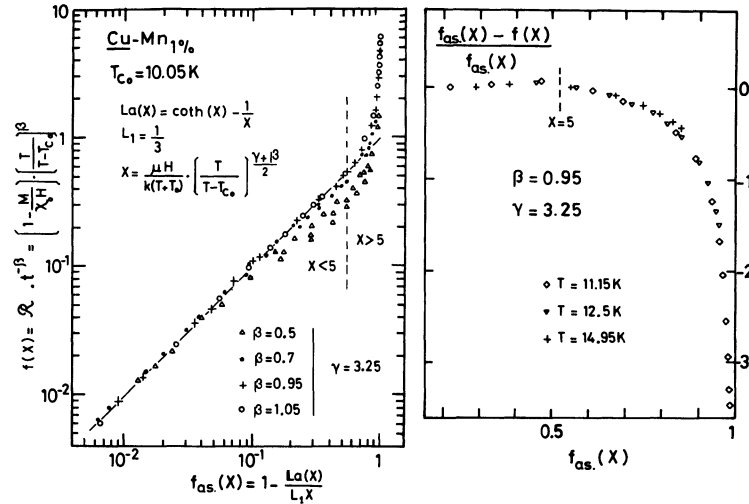
Fig. 10. — On the left hand side, $\Re t^{-\beta}$ from the experiments is compared to its theoretical value $f_{as}(X) = 1 - \dfrac{La(X)}{L_1 X}$ (as derived from the high temperature regime $X = 0$) in a Log-Log plot for different values of β. Despite a relative insensitivity of our data to the values of β (for $0.5 < \beta < 1.05$), we observe on this figure a degradation of the validity of the scaling and, at the same time, of the fit to the asymptotic expression if we try to decrease β below 0.7. It is in fact near T_c (for $X > 5$ where our asymptotic expression is no more valid) that we may hope to reach a more accurate determination of β. Further details are seen on the error curve $-\dfrac{\Re t^{-\beta} - f_{as}(X)}{f_{as}(X)}$ vs. $f_{as}(X)$ which is shown on the right-hand side for $\beta = 0.95$.

or otherwise

$$\frac{\Re}{(\mu H/kT^*)^{2/\delta}} = \frac{1 - \dfrac{M}{\chi_0 H}}{(\mu H/kT^*)^{2/\delta}} = \frac{f(X)}{X^{2/\delta}} = g(X). \quad (13b)$$

The success of the scaling argument is impressive : all our data ($1.1\ T_c < T < 4\ T_c$, $0 < H < 7$ teslas) superimpose to fit a unique function of the scaling variable X which we derived from the two exponents determined in the $(H/T) \rightarrow 0$ limit as may be verified from the figure 9.

Moreover the method which we used implies that equations 13 hold in the high temperature limit $\left(\dfrac{T - T_c}{T} \rightarrow 1 \text{ and } X \rightarrow 0\right)$ where the magnetization obeyed a Langevin function. In this limit, we can therefore make use of equation 11b which, together with equation 10, provides us with an asymptotic explicit expression of the function $f(X)$. We have

$$f_{as}(X) = 1 - \frac{La(X)}{L_1 X}. \quad (14)$$

This high temperature expansion accounts for the data in the entire range $X < 5$ as may be seen from figure 9.

On figure 10, we have plotted $\text{Log}\left(\dfrac{\Re}{t^\beta}\right)_{exp}$ vs. $f_{as}(X)$ as given by equation 14, for several values of β within the limits of errors in equation 12. This figure allows us to improve the accuracy for the values of β determined from data with $X < 1.5$ by making use of the data in the range $1.5 < X < 5$. We observe on this figure that both the agreement with the theoretical expression 14

and the dispersion are improved if we choose values of β closer to the upper limit $\beta \simeq 1$ of our previous error estimate. Compare also the scaling of the plots for $\beta = 0.75$ and $\beta \simeq 1$ in figure 9.

4.3 THE MAGNETIZATION AT T_c. — The version 13b of the scaling equation imposes :

$$\Re(X) = 1 - \frac{M}{M_{sat}\ L_1\ \dfrac{\mu H}{k(T_c + T_0)}} \sim A\left(\frac{H}{T}\right)^{2/\delta} + \cdots \quad (15)$$

where $H \rightarrow 0$ at T_c (i.e. in the limit $t \rightarrow 0$ or $X \rightarrow \infty$). This in turn requires

$$f(X) \sim AX^{2/\delta} + \cdots \quad (16)$$

in the same limits.

From the magnetization at $T = T_c$, it is thus possible in principle to obtain δ and hence a relation ($\delta^{-1} = \beta/(\gamma + \beta)$) between β and γ independent of our previous determination at $T > T_c$. Experimentally we have all the necessary accuracy to obtain a precise value of δ from which a precise value of β could be deduced. Unfortunately, this determination seems sensitive to the higher order terms in H which may follow the initial term in $H^{2/\delta}$. This can be seen on figures 11 and 12 where we have represented the same data in plots of $M(H, T_c)/H$ vs. $H^{2/\delta'}$ and $\text{Log}\ (M(H, T_c)/H)$ vs. $H^{2/\delta''}$ respectively for the values δ' and δ'' which correspond to the best linearity in each case. In these figures M/H is expressed in units of the initial susceptibility at T_c (χ_0' and χ_0'' respectively)
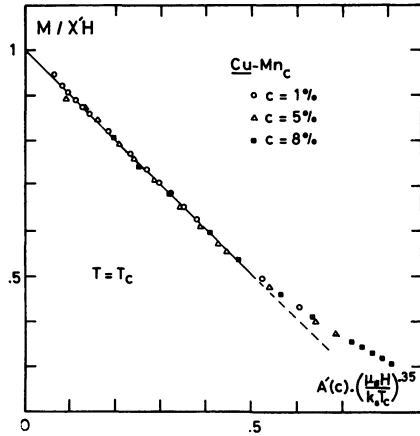
Fig. 11. — $M(T_c)/\chi'_0\ H$ vs. $A'(c).(\mu_B\ H/kT_c)^{0.35}$ for 3 CuMn alloys with $A'(c)$ as given in table III. A power law $1 - M/\chi'_0\ H \sim H^{2/\delta'}$ can describe the magnetization at T_c over a large range of field values as predicted by the theory. Notice the systematic deviations which appear for the three samples in large fields.
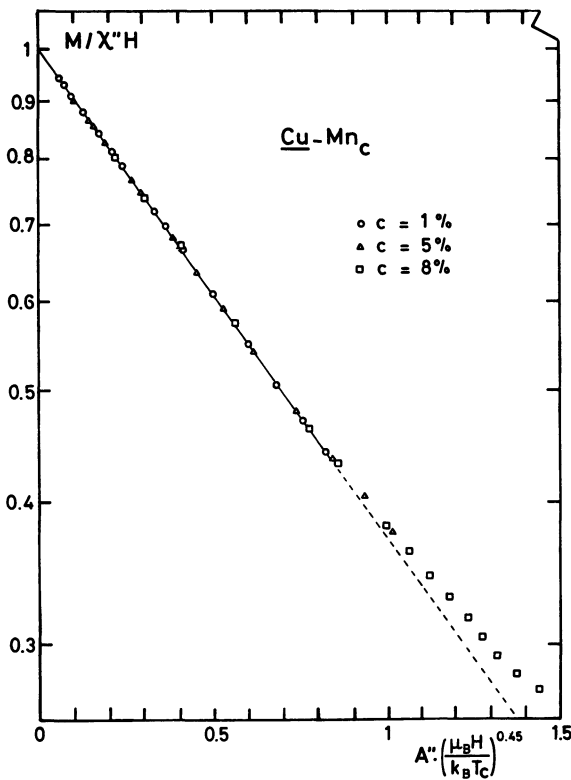

Fig. 12. — The same data as in figure 11 in a

$$\text{Ln}\ [M(T_c)/\chi''_0\ H]\ \text{vs.}\ A''(c)\ (\mu_B\ H/kT_c)^{0.45}$$

plot with $A''(c)$ given in table III. An exponential law $M/\chi''_0\ H = \exp(- A''\ H^{2/\delta''})$ can describe the magnetization at T_c in all the range of field values for the 1 %. Deviations to this law are observed in the highest fields for the two other samples.

which is obtained by extrapolating the initial linear part of the plots to the $H = 0$ limit. Note that in both cases, the same values δ' and δ'' fit not only the data for our 1 at. % sample but also for two higher concentrations ($c = 5$ and 8 at. %), with the values $\chi'_0(c)$ and $\chi''(c)$ reported in table III. The deviations from the linearity (for $H > 20$ kOe in 1 at. %) in the $M/\chi'_0\ H$ vs. $H^{2/\delta'}$ plot are quite small; it is thus not surprising if an exponential function produces a good fit in still higher fields (up to 70 kOe for the 1 at. %). The bad point is that the values we obtain for δ' and δ'' are somewhat different. With the accurate determination of $\gamma = 3.25$ being given, we calculated $\beta' = 0.7$ and $\beta'' = 0.95$ i.e. two values which lie within our previous error for β (see Eq. 12). In fact we have no real reason to prefer one development to the other in the absence of a theoretical prediction. A point in favour of the plot of figure 12, which assumes $M/H \sim \exp(- H^{2/\delta''})$ for small H, is that it extrapolates in zero field to a value χ''_0 closer to that deduced from the high temperature determination of the Curie constant (i.e. $\chi''_0 = 0.579 \times 10^{-6}$ emu/g rather than $\chi'_0 = 0.592 \times 10^{-6}$ emu/g as compared with the value 0.568×10^{-6} emu/g which was determined from the $T > T_c$ data for our 1 at. % sample).

The scaling diagram 9 has been calculated with $\gamma = 3.25$ and β fixed somewhat arbitrarly to 0.75. The asymptotic form (16) of the function $f(X)$ for the $t \to 0$ ($X \to \infty$) limit is represented by a dashed line on the figure 9.

Table III. — Values of the parameters χ_0 (initial susceptibility) and A involved in the development of the magnetization at T_c :

$$M = \chi'_0\ H\left[1 - A'\left(\frac{\mu_B\ H}{kT_c}\right)^{2/\delta'}\right] \quad \text{(see Fig. 12)}$$

$$M = \chi''_0\ H \exp\left[- A''\left(\frac{\mu_B\ H}{kT_c}\right)^{2/\delta''}\right] \quad \text{(see Fig. 13)}$$

for three different Mn concentration c in CuMn system.

	c	1 %	5 %	8 %
$\delta' = 0.35$	$\chi'_0(c)$ 10^{-4} emu/g	0.543	2.33	4.20
	$A'(c)$	0.82	1.38	1.73
$\delta'' = 0.45$	$\chi''_0(c)$ 10^{-4} emu/g	0.533	2.29	4.13
	$A''(c)$	1.21	2.30	3.85

4.4 DISCUSSION OF THE METHOD. — Barbara et al. in reference 15 search for the phenomenological exponent $\alpha(T)$ which best fits their data (in a fixed and

relatively broad window of field values $H < H_0$) according to an expression of the form

$$\Re(H < H_0) = 1 - \frac{M}{\chi_0 H} \sim a_\alpha(T) H^{\alpha(T)}. \quad (17)$$

In the framework of the scaling theory this procedure makes sense at $T = T_c$ where $\alpha(T) \rightarrow 2/\delta$ and at high temperatures where $\alpha(T)$ should tend towards 2. This is precisely what the data show. Between these two limits equation 17 is more qualitative since as we saw in chapter 3 more and more higher order terms become necessary to account for data in a given window of fields on approaching T_c. Consequently, no conclusion can be drawn from the temperature dependence of $a_\alpha(T)$. This appears clearly in reference 25 where Binder and Kinzel analysing their numerical data for the 2-dimensional short range Ising model in the same way find a coefficient $a_\alpha(T)$ which apparently diverges at $T_c = J$. One could be mislead to a wrong conclusion in this system which *does not* present a phase transition. To be fair, one should realize, though, that the claim about the existence of a phase transition in reference 15 is *not* based upon the temperature dependence of $a_\alpha(T)$; it is based upon the success of the scaling argument. It would certainly be very instructive to everybody to know whether such a scaling can also be reached (within the same experimental uncertainty) using the numerical data of Binder and Kinzel [25].

Our method has been different and, we believe, more convincing since two exponents are derived from the study of the divergence of two well identified coefficients of the expansion of $M(H)$. The scaling law is then constructed without further fitting or assumptions. This imposed a gain of more than one order of magnitude in the relative accuracy with respect to previous experimental or numerical data (compare our Fig. 13 to Fig. 4 of Ref. 25) and, as we already stressed, a restriction to the $T > 1.1\ T_c$ range. Notice that the range where the scaling was found to be valid within our accuracy has been considerably extended through the use of $\frac{H}{T}$ and $\frac{T - T_c}{T}$ (rather than $\frac{H}{T_c}, \frac{T - T_c}{T_c}$) as the variables of the scaling argument.

Another method, still, is used in reference 16. The advantage of the thermal analysis technique which is used is that it permits a more direct access to the singular magnetization : γ is accurately determined in the close vicinity of T_c. Then the scaling law is reached using reasonable assumptions for β.

It should be stressed that all experiments seem to agree in their conclusions. A puzzling feature of this transition (if it is accepted that the observation of a scaling is a sufficient proof) is the large domain where this scaling is observed with exponents which differ from the mean field predictions.
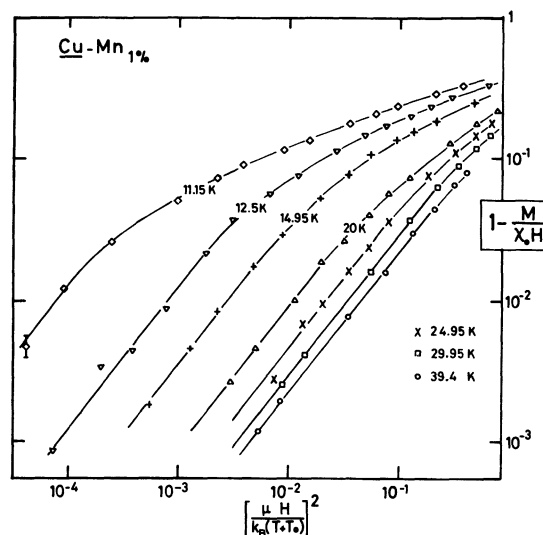

Fig. 13. — $\mathrm{Log}\,(1 - M/\chi_0 H)$ vs. $\mathrm{Log}\left(\frac{\mu H}{kT^*}\right)^2$. This diagram shows that an accuracy better than 10^{-3} on M/H is necessary in order to conclude on the temperature dependence of the H^3 term of the magnetization. Compare this plot with the figure 4 of reference 25.

We have stressed, on several occasions, the importance of using non linear scaling variables in order to improve, as we did, the success of the scaling plots (compare Fig. 9 to the equivalent plots in Ref. 15). The critical exponent for the function $f(\varepsilon)$ is λ if $f(\varepsilon) \sim A\varepsilon^\lambda$ when the dimensionless variable $(T - T_c)/T_c = \varepsilon$ tends to zero. It is natural to use ε in the spirit of the scaling procedure which aims at describing the dominant divergence only. The same exponent would be obtained with any other non linear variable which like $t = \varepsilon/(1 + \varepsilon)$ tends to ε when ε tends to zero. For example equation 13a which is illustrated by the figure 9 can be written in terms of the usual variables

$$\frac{\Re(h)}{A\varepsilon^\beta} = f(\alpha X')$$

with $X' = \varepsilon^{\frac{\gamma + \beta}{2}} \frac{\mu H}{kT_c}$.

The prefactors $A = (1 - \varepsilon)^\beta$ and $\alpha = (1 - \varepsilon)^{1 - (\gamma + \beta)/2}$ account for non pathological temperature dependences which do not affect the scaling result. They however permitted to extend the validity of the scaling in a range between $1.1\ T_c$ and $4\ T_c$ where, among other advantages, the results are less dependent on an accurate choice of T_c. Notice that this does raise a serious question about the previous evaluations of the range of validity of the scaling which obviously should not depend on the choice of the variable.

We have several arguments, none of them totally convincing, to support our choice of the non linear variables

$$t = \frac{T - T_c}{T} = \frac{\varepsilon}{1 + \varepsilon} \quad \text{and} \quad \frac{H}{T} = \frac{H}{T_c}\frac{1}{1 + \varepsilon}.$$

i) It seems natural, in the spirit of the Boltzman factors, to weigh any field (H or $T - T_c$) in units of the actual temperature T and not of the fixed temperature T_c.

ii) We have, on this basis, developed our experimental analysis from figure 3 to figure 6. These plots conserve the paramagnetic response which appears, not as an additional contribution of a different nature from the spin glass anomaly, but as the natural $T \to \infty$ limit of this anomaly. Now the well known scaling equation for the magnetization (our Eq. 9) can be written :

$$\Re\left(h\left(\frac{H}{T_c}\right)\right) = \frac{|\varepsilon|^{-(\gamma+\beta)}}{|\varepsilon|^{-(\gamma+2\beta)}} f\left[|\varepsilon|^{-(\gamma+\beta)} h\left(\frac{H}{T_c}\right)\right]$$

or otherwise

$$= \frac{\mu(\varepsilon)}{\text{vol}(\varepsilon)} f\left[\mu(\varepsilon) h\left(\frac{H}{T_c}\right)\right],$$

$\mu(\varepsilon) = |\varepsilon|^{-(\gamma+\beta)}$ has the dimensions of the moment associated to the field h (a real moment μ if $h\left(\dfrac{H}{T_c}\right) = \dfrac{H}{T_c}$ as in usual transitions, and the square of a moment if $h\left(\dfrac{H}{T_c}\right) = \left(\dfrac{H}{T_c}\right)^2$ as in the spin glass transition); $\text{vol}(\varepsilon) \sim |\varepsilon|^{-(\gamma+2\beta)}$ is the correlation volume (ξ^d in dimension d if ξ is the correlation length). Observe that this expression fails to provide the paramagnetic limit when $T \to \infty$ while with ε and H replaced by $\dfrac{\varepsilon}{1+\varepsilon}$ and $\dfrac{H}{1+\varepsilon}$ respectively this limit is automatically obtained. We have

$$\Re(H) = \frac{\mu(t = 1)}{\text{vol}(t = 1)} f\left(\frac{\mu(t = 1)H}{kT}\right)$$

for usual transitions (or Eq. 13 in the spin glass case) with finite values $\mu(t = 1)$ and $\text{vol}(t = 1)$ which can be ascribed to the atomic limits of the corresponding quantities.

iii) The remarkable success that we had when we identified $f(X)$ in its asymptotic limit with the corresponding expression deduced from the Langevin function suggests that we have made more than a simple interpolation (see Eq. 14 and Fig. 9).

iv) Finally notice that the high temperature limit is reached with a law which to first order in h can be written as

$$\Re(h) \sim \left(\frac{T - T_c}{T}\right)^{-\gamma} h\left(\frac{H}{T}\right)$$

which tends to

$$\Re(h) \sim \frac{hT}{T - \gamma T_c} \quad \text{when} \quad T \to \infty.$$

For usual phase transitions when $h = H/T$, we obtain the mean field result

$$\Re(H) = M(H) \simeq \frac{H}{T - \gamma T_c}.$$

For the spin glass transition where $h = (H/T)^2$ we obtain the S.K. results

$$\Re\left(\left(\frac{H}{T}\right)^2\right) = \frac{M(H) - \chi_0 H}{\chi_0 H} = \frac{H^2}{T(T - \gamma T_c)}.$$

In both cases the high temperature Curie temperature (presumably the mean field limit) $T_c^{\text{M.F.}}$ is equal to γT_c.

Table IV shows some cases where this relation accounts for the calculated results with a surprising success. We are not supporting the conjecture that $T_c^{\text{M.F.}} = \gamma T_c$ is an exact result but simply suggestions that our choice of variables might be very pertinent in cases other than the spin glass transition.

Table IV. — *Comparison of γ to the $T_c^{\text{M.F.}}/T_c$ ratio for the Ising model. The data can be gathered from Stanley H.E. in « Introduction to phase transitions and critical phenomena ». Clarendon Press Oxford 1971 and the references therein (notice that in the case $d = 1$ we have $\chi = \dfrac{C}{T}\left(\dfrac{T}{T-0}\right)^\gamma$ with our variables and γ therefore is undetermined).*

Dimension	1	2 square lattice	3 f.c.c. lattice	M.F.
$T_c^{\text{M.F.}}/T_c$	∞	1.763	1.23	1
γ	undetermined	1.75	1.25	1

5. Discussion of the results.

The main point of this paper is the confirmation of the validity of the particular scaling that equation 12 implies : the quantity which is coupled to the field is $\sqrt{\varepsilon^{\gamma+\beta}}$ rather than $\varepsilon^{\gamma+\beta}$ as in usual phase transitions.

We find a similar idea in Neel's description of the superparamagnetism of antiferromagnetic particles [27]. In the superparamagnetic regime, Neel writes the magnetization of N/n grains, containing an average of n individual moments each, as

$$M = \frac{N}{n} S_n La\left(\frac{S_n H}{T}\right). \tag{18}$$

If vacancies or defects affect the internal order of each sublattice in the grain, Neel proposes to estimate the giant moment S_n of the grain through a random walk

calculation, yielding $S_n \sim \sqrt{n\mu}$. Equation 18 can then be developed as

$$\frac{M}{M_{sat}} = L_1 \frac{\mu H}{T} + L_3 \, n \frac{\mu^3 H^3}{T^3} + L_5 \, n^2 \frac{\mu^5 H^5}{T^5} + \cdots .$$

(19)

The first order susceptibility is then the same in the high temperature paramagnetic regime as in the superparamagnetic regime, when the individual spins have ordered to form a giant moment, and does not depend on the size of this giant moment which can be estimated from the higher order terms in the development of the magnetization. The argument would then of course remain valid if the size of the moments was temperature dependent : the argument is a statistical one, and comes from the fact that the field is coupled to a quantity which increases as the square root of the correlation volume (and not like the total volume as would be the case in a ferromagnet).

Similarly, in the scaling argument of Suzuki and Chalupa, the field is coupled with $|\varepsilon|^{-\frac{\beta+\gamma}{2}}$ rather than $|\varepsilon|^{-(\beta+\gamma)}$, while the correlation volume increases in principle like $|\varepsilon|^{-(\gamma+2\beta)}$. With large γ and small β values, the condition is not far from being realized. In fact equation 19 is strictly the $\beta = 0$ approximation of this model. Since the β parameter enters only at the level of the term of order H^5 in the expansion of M, this « critical superparamagnetic » approach in terms of non interacting moments diverging like $(T - T_c)^{-\gamma}$ is bound to be valid in the low field high temperature limit anyway.

The similarities between the two types of argument seem interesting to us. They call attention upon the fact that our parameter a_3 is closely connected with the extension of the correlations in the system. It directly measures the number n of individual moments which are correlated at this temperature (see Eq. 19). Notice that this same argument of Neel has also been used by Cyrot to justify the Fulcher law (28).

6. Conclusion.

The main result of this paper is the confirmation in CuMn 1 at. % of the validity of a scaling equation of the form

$$1 - \frac{M}{\chi H} = \Re(h = H^2) = t^\beta \, f(X)$$

with

$$X = \frac{\mu H}{kT} \cdot t^{-\frac{\gamma+\beta}{2}} , \qquad t = (T - T_c)/T ,$$

$$\gamma = 3.25 \pm 0.10 , \qquad \beta = 0.75 \pm 0.25 .$$

We have drawn in the (H, T) plane (see Fig. 14) two lines which each correspond to a particular value of the scaling variable X. The most important is the $X = 5$ line which indicates a cross-over between two
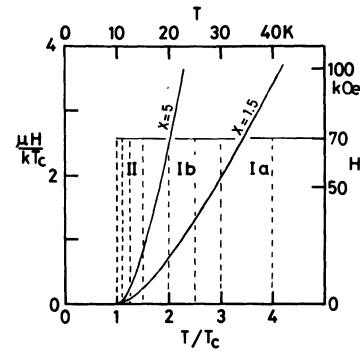

Fig. 14. — Cross-over in the (H, T) plane. In the region I $(\chi < 5)$ the magnetization can be interpreted in terms of a Langevin function of magnetic moments which increase on lowering the temperature and diverge at T_c. In the region II $(\chi > 5)$ the behaviour of the singular magnetization is dominated by its limits at T_c where it has the form $\chi^{2/\delta}$ in low fields. Our isothermal measurements explore the range of fields $(0 < H < 70 \text{ kOe})$ which is schematized by the vertical dashed lines at the different temperatures $T_c < T < 4\,T_c$. All the data in this range superimposed on a single universal function which was entirely determined with the data of the $X < 1.5$ range (Ia).

regimes. For $X < 5$, we may use an explicit approximation for $f(X)$ which comes from the high temperature limit when $X \to H/T$ and the magnetization tends to a Langevin function. We can use, in this regime, a physical picture in terms of superparamagnetic clouds, increasing in size on lowering the temperature and *tending to diverge at T_c* (region I). For $X > 5$, in contrast, the behaviour of the singular magnetization $\Re(H^2)$ is governed by the limit of $f(X)$ at $T = T_c$ where it has the form of a power law $X^{2/\delta}$ in low fields : this traduces possibly the deformation of the diverging clusters under the effect of the field in the saturated regime (region II). In the high temperature regime, the line $X = 1.5$ limits the range of the (H, T) plane (region Ia) where $M(H)$ is sufficiently defined by the first three terms of its series expansion in terms of H/T. We have determined the exponents β and γ of our scaling equation in this range. With the values found, it was possible to rescale all our data $(T_c < T < 4\,T_c, \ 0 < H < 7$ teslas) onto a universal function.

We believe that this is strong evidence in favour of a phase transition occurring at finite temperature in three dimensional RKKY spin glasses. The coefficient a_3 of the $(H/T)^3$ term in the expansion of the magnetization is a measure of the number of correlated Mn atoms in the $\beta \to 0$ model which we discussed in chapter 5. In this approximation, the effective cluster radius varies by a factor 10, increasing from $3a$ to $20a$ (where a is the dimension of the atomic cell in the fcc lattice) in our range of measurements. If we include the data of Odin et al. [26], this number is increased to $400a$ when we approach $1.007\,T_c$.

Our conclusion is not in contradiction with the

evidence which may be obtained from the low temperature data. We are aware of the fact that many of the effects which dominate the properties in the $T < T_c$ phase (such as the field, temperature and time dependence of the remanences and the TLnt scaling which prevails at low temperature [4]) can be described in the model of independent particles. These properties are not characteristic of a phase transition, but they do not disprove the possibility of a phase transition (see the discussion in Ref. 4). On examining the detailed aspects of these properties, we even find features such as the presence of energy relaxations [29] at $T < T_c$, or the existence of a Fulcher law [5] (and the discontinuity that it suggests between two regimes with different dynamics) which do not fit easily into a description in terms of paramagnetism.

It is however in the $T > T_c$ regime that we reach more easily reliable time independent data showing a behaviour which is definitely singular. It is worthwile to recall in this respect that there exists a good quantitative agreement between the experimental determinations of critical exponents which have been performed in this range and rely upon the verification of the validity of a scaling equation.

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