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# Static and dynamic evidence for a transition at $T_c = 0$ in the Ising spin glass $Fe_{0.3}Mg_{0.7}Cl_2$

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**Résumé.** — L'étude du comportement critique du verre de spin Ising Fe $_{0.3}$ Mg $_{0.7}$ Cl $_2$  a été réalisée à l'aide de mesures d'aimantation, de susceptibilité alternative et de chaleur spécifique. Les mesures de susceptibilité alternative ont été effectuées jusqu'à de très basses fréquences ( $0.02\,\mathrm{Hz} < f < 3000\,\mathrm{Hz}$ ) grâce à la mise au point d'un dispositif expérimental utilisant un convertisseur analogique numérique 16 bits rapide. Le temps de relaxation  $\tau$  comme les coefficients  $a_3$  et  $a_5$  de la susceptibilité non linéaire sont puissances l'un de l'autre et divergent comme des exponentielles de  $\theta/T$ . L'analyse des résultats confirme donc les conclusions d'une première étude du comportement critique dynamique : Fe $_{0.30}\,\mathrm{Mg}_{0.70}\,\mathrm{Cl}_2$  appartient à une classe d'universalité différente des systèmes 3d et apparaît comme exemple de verre de spin 2d Ising caractérisé par une transition de phase à  $T_c = 0$ . Nous affirmons que les singularités observées apparaissent comme la limite naturelle à  $T_c = 0$  des lois habituelles de "scaling". Nous nous différencions sur ce point des analyses présentées récemment pour d'autres systèmes, où un comportement semblable est interprété par une théorie ad hoc pour  $T_c = 0$ .

Abstract. — The critical behaviour of the Ising spin glass  $Fe_{0.3}Mg_{0.7}Cl_2$  has been studied by d.c. magnetisation, a.c. susceptibility and specific heat measurements. We have extended the range of our previous a.c. susceptibility measurements down to 0.02 Hz by using a new set up which utilises the possibilities of a 16 bit analog to digital converter. The relaxation time  $\tau$  and the coefficients  $a_3$  and  $a_5$  of the non linear susceptibilities are powers of each other and diverge like exponential functions of  $\theta/T$ . This confirms our former statement that  $Fe_{0.3}Mg_{0.7}Cl_2$  is of a class different from that of traditional 3d systems and appears as an example of a real 2d Ising spin glass with  $T_c = 0$ . We claim that the essential singularities are obtained as the natural  $T_c = 0$  limit of the usual assumption of the scaling theory. We differ at this point from recent analyses in other systems, where similar evidence is interpreted in the framework of a different theory ad hoc for the  $T_c = 0$  case.

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#### 1. Introduction.

Theory predicts a transition at finite temperature for 3d Ising spin glasses only. On the other hand, theoreticians would rather favor a transition at  $T_c = 0$  for 2d Ising spin glasses and no transition at all in cases of lower dimension or Heisenberg systems. However, experimentally it is now accepted that the spin glass transition occurs at finite temperature at d = 3 with Heisenberg spins. Typical exponents are  $\gamma \sim 3$ ,  $\beta \sim 1$ ,  $z\nu \sim 6 - 10$ .

Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub> spin glass has properties which differ from the standard behaviour. For example the field cooled magnetisation does not reach as flat a plateau as in CuMn but it conserves a slight negative slope in terms of the temperature and remains strongly dependent on the cooling rate [1]. Also, our first dynamic data, obtained from Faraday rotation experiments in the frequency range 1.5-80000 Hz [2,3], showed that the low temperature slope of  $\chi''$  depends more on frequency than is usual. Furthermore we pointed out that the dynamic critical behaviour was unconventional, not characteristic of a 3d Ising spin glass, but more relevant of 2d Ising spin glasses and we were able to conclude from this study that we had with this system the first example of a spin glass transition occurring at  $T_c = 0$ .

Recently other unconventional critical behaviours were reported in a number of systems and discussions arise as whether they can be taken as examples of a transition at  $T_{\rm c}=0$  or whether they call for a different interpretation [4-6]. Because of the fundamental interest that there is in the discussion from the point of view of the spin glass transition as well as from the point of view of transitions in general, we have completed our former dynamic study of the Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub> system. We have developed an original method based upon the acquisition of the signal by means of a 16 bit analog to digital converter and computer data processing which allowed us to measure a.c. susceptibilities down to 0.02 Hz with a good accuracy. Thus the range of our previous dynamic study was extended significantly by two decades at low frequencies.

In the following we briefly describe the new low frequency a.c. susceptibility technique and report our new data in a crystal of  $Fe_{0.3}Mg_{0.7}Cl_2$  over the 0.02-3000 Hz frequency range. We then recall our non linear static susceptibility results [1] and give new low temperature specific heat measurements on this compound. We show that the temperature dependence of the characteristic relaxation time over the whole range between 0.02 and 80000 Hz is well described by an Arrhenius law and that the non-linear susceptibilities are described by essential singularities [which are, as we show, the natural  $T_c = 0$  limit of the power laws], confirming our former conclusion on the 2d-Ising like character of the  $Fe_{0.3}Mg_{0.7}Cl_2$  spin glass. We discuss our data in connection with recently published results on other systems [4-6].

# 2. Experimental procedures and results.

All the samples were issued from a Fe<sub>0.30</sub>Mg<sub>0.70</sub>Cl<sub>2</sub> single crystal grown by the Bridgman method. They were cleaved in a glove box filled with an <sup>4</sup>He atmosphere to avoid hydration during preparation.

2.1 DYNAMIC SUSCEPTIBILITY. — The sample used in our new low frequency experiments was composed of three identical disks, 6 mm in diameter and 3 mm in thickness, the c-axis of the crystal being perpendicular to the platelets. These three disks were stacked in a sample holder to get a 9 mm thick sample.

The a.c. magnetic field was supplied by a small superconducting coil; its amplitude was about 2 Oe for all experiments and it was applied along the c-axis of the crystal. The pick up coil, composed of two opposite windings of 5000 turns of copper wire, was coaxial with the primary coil.

It could be weakly shifted inside the primary magnetic field at the beginning of the measurements to tune the compensation of the two opposite windings, the induced voltage being zero in the absence of sample.

The a.c. susceptibility was measured by two different methods. At high frequencies  $(f = \omega/2\pi > 100 \, \text{Hz})$  a conventional lock-in amplifier was used. At the low frequencies  $(0.02 \, \text{Hz} < f < 100 \, \text{Hz})$  we used a 16 bit analog to digital converter (ADC). At frequencies smaller than 3 Hz, 1700 values of a signal e(t) proportional to the pick up coil voltage, and of a signal h(t) proportional to the primary field were recorded for one period as shown in figure 1 for 0.02 and 0.1 Hz. From these data we deduce precisely the amplitude of e(t) and h(t) and their phase shift allowing to calculate the real  $\chi'_{\omega}$  and the imaginary part  $\chi''_{\omega}$  of the susceptibility. We have checked that this method gives the same results as lock-in amplifier measurements at a typical frequency of 100 Hz.

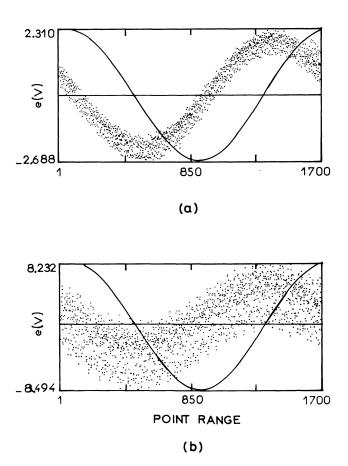


Fig. 1. — The pick up coil voltage e(+) and a voltage u(+) proportional to the primary magnetic field, recorded versus the point range over a period for two working frequencies 0.1 Hz (a) and 0.02 Hz (b).

Figures 2 and 3 show the temperature dependence of the longitudinal susceptibilities  $\chi'_{\omega}(T)$  and  $\chi''_{\omega}(T)$  at different frequencies. On decreasing the frequency, the cusp in  $\chi'_{\omega}(T)$  shifts continuously towards lower temperature,  $\Delta T(\chi'_{\max})/\Delta \log f$  being roughly equal to 0.25 K/decade in

the high frequency range and to 0.10 K/decade in the low frequency range. This cusp is strongly sharpening at lowest frequencies: at 0.02 Hz it is similar in width and in position to the cusp we have observed in static ZFC susceptibility measurements by using a heating rate of 0.2 mK.s<sup>-1</sup> [1]. The amplitude of the dissipative term  $\chi_{\omega}''(T)$  decreases by lowering the frequency; the value of the ratio  $\chi_{\omega}''(T)/\chi_{\omega}'(T)$  at maximum is roughly equal to  $3\times 10^{-2}$  for our lowest frequency at 0.02 Hz. The signal to noise ratio on  $\chi_{\omega}''$  is rather poor at low frequency since it is measured from the pick-up coil voltage which is proportional to  $\omega$ .

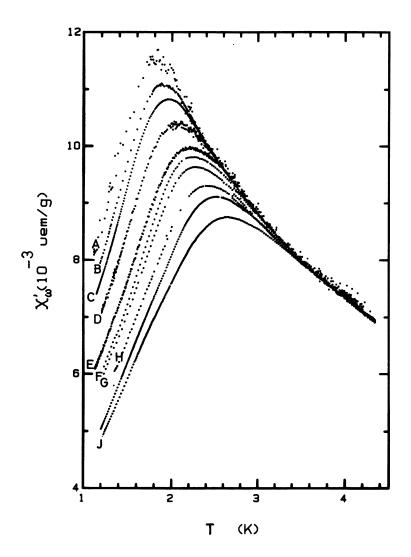


Fig. 2. — The real part  $\chi'_{\omega}(T)$  of the complex susceptibility versus temperature for various frequencies: 0.02 Hz (A), 0.1 Hz (B), 0.3 Hz (C), 3 Hz (D), 30 Hz (E), 113 Hz (F), 211 Hz (G), 1 kHz (H), 3 kHz (I) and 10 kHz (J).

Our previous a.c. susceptibility data in the range 1.5 - 80000 Hz were obtained by a method derived from Faraday rotation: experimental details are given in reference [3].

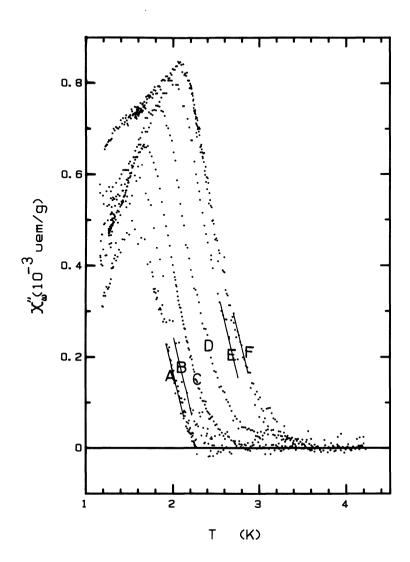


Fig. 3. — The imaginary part  $\chi''_{\omega}(T)$  of the complex susceptibility versus temperature for various frequencies: 0.1 Hz (A), 1 Hz (B), 10 Hz (C), 113 Hz (D), 1 kHz (E) and 3 kHz (F).

2.2 STATIC MAGNETISATION. — The magnetisation measurements were performed for several values of the magnetic field between 10 and 400 Oe, the sample being cooled at a constant rate from 4.2 to 1 K. This field cooled procedure is known to provide data which remain close to the equilibrium response even in the non ergodic regime where the relaxation time becomes of the order of laboratory times. However, in this system, the dependence of the magnetisation on the cooling rate at the lowest temperatures constrained us to reduce the temperature range under study to the range 1.7 - 4.2 K. The data acquisition was controlled by computer, allowing measurements at well defined temperatures: the isothermal magnetisation curves were easily deduced from the field cooled magnetisation data. The main results are presented in a previous paper [1].

2.3 SPECIFIC HEAT. — The measurements have been performed with a dilution refrigerator using a transient heat-pulse technique [7]. The sample (a single crystal piece of 0.832 g in weight) was glued to the silicon sample holder with a controlled amount (10 mg) of Apiezon N grease which protected the sample from moisture during the preparation of the experiment. Due to the large heat capacity of the sample, the time constant of the exponential decay of the temperature transients varies from 10 minutes at 7 K to several hours at 0.1 K, which corresponds to the quasi-adiabatic conditions. Also the contribution of addenda was generally negligible (less than 1 % below 5 K). The curve of the specific heat measured in the range 0.1 - 7 K is shown in figure 4.

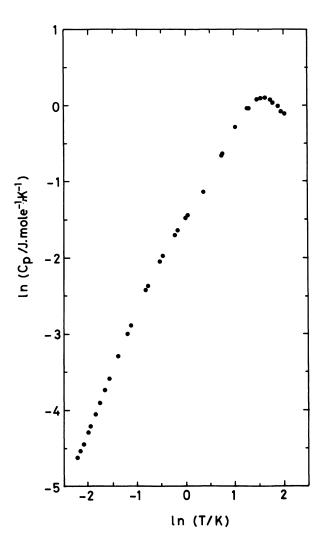


Fig. 4. — In-ln plot of the specific heat temperature dependence of  $Fe_{0.3}Mg_{0.7}Cl_2$ . The data are corrected from the lattice contribution (see text).

### 3. Discussion.

3.1 DYNAMIC SUSCEPTIBILITY COLE-COLE ANALYSIS. — Following Saito et al. [8], several authors have analysed the susceptibility of spin glasses with an empirical law which was first introduced for dielectrics [9]:

$$\chi(\omega) = \chi_{\rm o} / \left( 1 + (i\omega \tau_{\rm cc})^{\alpha(T)} \right) \tag{1}$$

For  $\omega \tau << 1$  the out of phase susceptibility reduces to :

$$\chi''(\omega) \sim (\omega \tau_{\rm cc})^{\alpha(T)} \tag{2}$$

For  $\alpha(T)=1$  we recover the usual Debye result for a single relaxation time. A smaller  $\alpha(T)$  results from a distribution of relaxation times which becomes broader as  $\alpha$  decreases to zero.

Figures 5a and 5b represent our data plot of  $\log(\chi''_\omega)$  vs.  $\log(\omega)$  at different temperatures between 1 and 4 K, measured either by the Faraday rotation technique or with the new set up. The different isotherms correspond to vertical sections of the different curves shown in figure 3. As we decrease the temperature the low frequency tail of  $\chi''(\omega)$  shifts up and flattens out. It is asymptotically well described by a straight line of slope  $\alpha(T)$ , which is consistent with a power law of the type described by equation (2). The figure 6 shows how  $\alpha(T)$  depends on the temperature. We notice some mismatch between the data obtained at T=2.8 K by the two methods; this may reflect a slight difference in the concentrations of the two different samples and in the thermometer calibrations which have been used for these two sets of experiments.

In usual spin glasses the parameter  $\alpha(T)$  varies abruptly near the phase transition temperature  $T_c$ , the existence of a transition at finite temperature being associated to an abrupt broadening at  $T_c$  of the relaxation time spectrum. For example,  $\alpha(T) \sim 1$  at 1.1  $T_c$  and 0.1 at 0.9  $T_c$  in  $(\text{Ti}_x \text{V}_{1-x})_2 \text{O}_3$  [8] . Similar results have been also reported in  $\text{Eu}_{0.4} \text{Sr}_{0.6} \text{S}$  [10] or in  $\text{CdCr}_{2-x} \text{In}_{2-2x} \text{S}_4[11]$  . In  $\text{Fe}_{0.30} \text{Mg}_{0.70} \text{Cl}_2$ , in contrast,  $\alpha(T)$  smoothly varies, approximately like  $T^2$  in the experimental range 1.8 - 4 K where it reaches a value close to one (figure 6). Dekker et al. [4] have observed a similar behaviour in the layered compound  $\text{Rb}_2 \text{Cu}_{0.78} \text{Co}_{0.22} \text{F}_4$  ( $\alpha$  varies like T in this case) which is also interesting to test the 2d-Ising behaviour. The progressive broadening of the relaxation time spectrum indicates that there is no transition at finite temperature.

Like Dekker et~al we have determined  $\tau_{cc}(T)$  which appears in the Cole Cole law (equation 1). In the high temperature regime ( $T \geq 3.5~{\rm K}$ ) where the width of the distribution collapses to zero, we find that  $\tau_{cc}(T)$  is equivalent to the value of the relaxation time  $\tau$  determined by others methods (see later). As the temperature decreases and the distribution broadens  $\tau_{cc}$  becomes much smaller than  $\tau$  (for example we observe  $\tau_{cc} \sim 10^{-7}~{\rm s}$  and  $\tau \sim 1~{\rm s}$  for  $T=2~{\rm K}$ ). This is consistent with the analysis of Saito et al. [8] and Alba et al. [11]: the relaxation time  $\tau_{cc}$  depends strongly on the width of the relaxation time spectrum and has no clear physical meaning when the temperature is decreased; it represents some typical relaxation time which is far from the averaged relaxation time (notice the logarithmic scale in the curve giving the distribution of  $\tau$  associated with the Cole Cole law [9]).

3.2 STATIC AND DYNAMIC CRITICAL BEHAVIOUR. — In a spin glass, expanding the static magnetisation in terms of H/T [12]:

$$M = a_1(H/T) - a_3(H/T)^3 + a_5(H/T)^5 - \dots$$
 (3)

the first order Curie constant  $a_1$  is a constant but  $a_3$ ,  $a_5$ ,... diverges like finite powers of the coherence length  $\xi$ . Assuming that for a transition at finite temperature  $T_c$  the coherence length diverges as the power of reduced temperature  $t = 1 - T_c/T$ :

$$\xi = \xi_0 (1 - T_c/T)^{-\nu} = t^{-\nu} \tag{4}$$

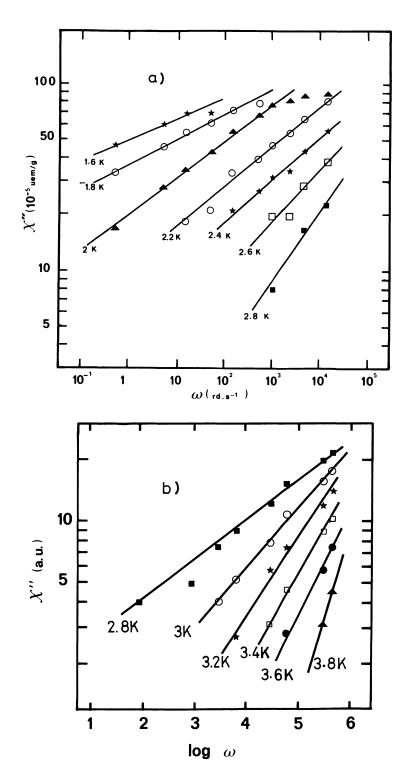


Fig. 5. — Plots of the imaginary part  $\chi''(\omega)$  of the susceptibility versus  $\omega$  for various temperatures: a) from our nex measurements; b) from reference [1]. The straight lines are fits with a power law:  $\chi'' \sim \omega^{\alpha}$ .

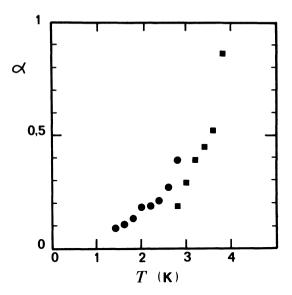


Fig. 6. — Temperature dependence of the exponent  $\alpha$  of the Cole-Cole law obtained from the fits of  $\chi$ "( $\omega$ ) described in the text. Circles ( $\bullet$ ) correspond to the present experimental a.c. measurements, squares ( $\blacksquare$ ) correspond to Faraday rotation measurements.

we have:

$$a_{2n+1} \sim (1 - T_c/T)^{-n(\beta + \gamma) + \beta}$$
 for  $n \ge 1$  (5)

On another hand, the dynamic scaling hypothesis states that the relaxation time is also a power of the coherence length and hence of the reduced temperature t:

$$\tau/\tau_0 = (\xi/\xi_0)^z = (1 - T_c/T)^{-z\nu} \tag{6}$$

For the specific heat, one has:

$$C_{\rm p}T^2 \sim (1 - T_{\rm c}/T)^{-\alpha}$$
 (7)

(the temperature independent critical exponent  $\alpha$  should not be mistaken for the temperature dependent  $\alpha(T)$  of the Cole-Cole law).

In a previous paper [1], we have presented the results of the analysis of the magnetisation in terms of odd powers of H/T as suggested in reference [12]. We have shown that the third order  $a_3$  and the fifth order  $a_5$  Curie constants vary respectively by one and two orders of magnitude between 2.5 and 1.7 K. Such a divergence was considered as a sign for a phase transition but we had difficulties to determine precisely a transition temperature. We shall examine again these results in connection with the analysis of the relaxation time of the system deduced from a.c. susceptibility measurements.

In our former a.c. susceptibility study [2,3], we mainly considered the temperature  $T_{\rm f}$ , related to the onset of irreversibilities, where  ${\rm tg}\varepsilon=|\chi_\omega''/\chi_\omega'|$  had a constant small value (10<sup>-2</sup>), and assuming that  ${\rm tg}\varepsilon=\omega\tau/2\pi$  [13], we deduced the correspondence between  $T_{\rm f}$  and the characteristic relaxation time  $\tau$  of the system:  $\tau(T_{\rm f})=0.01/\omega$ . Because of the scatter on the measurement of  $\chi''$  at the lowest frequencies, we have used in the present paper the  $\tau(T)$  dependence obtained

from the analysis of the breakaway point between  $\chi'_{\omega}$  and  $\chi_{\rm eq}$ , as suggested by Carré et al. [14]. They write that  $\tau_{\rm max}(T_{\rm i}) = \Delta\chi/\omega$  at the temperature  $T_{\rm i}$  where:

$$\Delta \chi = (\chi_{eq} - \chi'_{\omega})/\chi_{eq} = 2 \times 10^{-2}$$
 (8)

Figure 7 presents the temperature dependence of  $\Delta \chi$  in Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub>. In a recent paper, Bontemps *et al.* [15] have shown that in the limit  $\omega \tau << 1$  these two criteria used to determine the  $\tau(T)$  law are equivalent.

In most accepted spin glasses, fixing successively  $T_c$  at different finite values, the exponents  $\beta$  and  $\gamma$ , on one hand, the exponent  $z\nu$  and the characteristic time  $\tau_0$ , on the other hand, can be calculated by fitting  $a_3(T)$ ,  $a_5(T)$  and  $\tau(T)$  with equations (5) and (6) respectively. It is generally observed that there is only one value of  $T_c$  which minimises the variances; the exponent  $\gamma$  is generally of the order of 3,  $\beta$  of the order of 1 and the exponent  $z\nu$  ranges between 6 and 10 [16]. As figure 8 a shows, with our new dynamic data in Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub>, the smaller the  $T_c$  we chose the smaller the error. When  $T_c$  decreases the corresponding exponent  $z\nu$  steadily increases like  $\theta/T_c$  where  $\theta \sim 50$  K (Fig. 8 b). This is consistent with a transition at  $T_c = 0$ . Indeed in the high temperature "paramagnetic" regime,  $\xi$  ought to remain an analytic function of  $T^{-1}$  ( $\tau/\tau_0 \sim 1 + (z\nu T_c)/T + ... = 1 + \theta/T + ...$ ). This constraint which imposes that  $z\nu$  diverges like  $\theta/T_c$  leads to an Arrhenius law for the dynamic (and to essential singularities for all static properties) [17]:

$$\tau = \tau_{\rm o}(1 - T_{\rm c}/T)^{-\theta/T_{\rm c}} \xrightarrow{T_{\rm c} \to 0} \tau_{\rm o} \exp(\theta/T)$$
 (9)

Consistently with the above discussion we find (fig. 9) that if we try to represent  $\ln(\tau_{\rm max})$  vs.  $\ln(1-T_{\rm c}/T)$  for different attempt values of  $T_{\rm c}$  (0.5 K, 1 K or 1.5 K), the larger the  $T_{\rm c}$  we chose, the larger the curvature. The best linearity is obtained in the Arrhenius plot of  $\ln(\tau)$  vs. 1/T (fig. 10) which yields  $\theta = 57$  K and  $\tau_0 \sim 1.5 \times 10^{-12}$  s.

The same result follows from figure 11 which is the differential form of the equation (6).

$$P_{\tau}(T) = -\frac{\partial \ln T}{\partial \ln \tau} = \frac{T - T_{\rm c}}{z\nu T_{\rm c}} = \frac{T - T_{\rm c}}{\theta}$$
 (10)

As  $\tau_0$  has been eliminated in the differentiation, it is possible to obtain in one step the best value of the two other parameters  $T_c$  and  $z\nu$  (or  $\theta$ ). According to equation (6)  $P_{\tau}(T)$  should give a straight line. The intercept of this line with the y=0 axis is  $T_c$  and with the y=1 axis  $T_c+\theta$ . The data reported on figure 11 lead to:  $T_c=0\pm0.5$  K which implies an Arrhenius behaviour with  $\theta\sim63\pm7$  K.

The static data agree well with this conclusion. Figure 12 shows that  $a_5$  is indeed a power of  $a_3$  as equation (5) tells us and from this plot of  $\ln(a_5)$  versus  $\ln(a_3)$ , whose slope is  $2+\beta/\gamma$ , we obtain  $\beta/\gamma=0.2$ . We also find that, in an attempt to fit the temperature dependence of  $a_3$  with the power law, the best variance is obtained for  $T_c=0$ , although a satisfactory fit is noticed in the range 0-1.4 K, due to the uncertainty on the static data. We find thus that  $a_3$  and therefore  $a_5$ , like  $\tau$ , can be described by an essential singularity of the form  $a_{2n+1}(T)=a_{2n+1}\exp(\theta_{2n+1}/T)$  where  $\theta_{2n+1}=n\theta_{\gamma}-(n-1)\theta_{\beta}$  [ $\theta_{\gamma}=\gamma T_c$  and  $\theta_{\beta}=\beta T_c$ ] follows from equation (5). From the data of figure 13, we find  $\theta_{\gamma}=8.3$  K so that  $\theta_{\beta}=1.7$  K.

In the spirit of the previous analysis we would expect that  $\alpha$  becomes infinite when  $T_c \longrightarrow 0$  and that the anomalous contribution reduces to a Schottky anomaly:

$$C_{\mathbf{p}}T^{2} = A \exp\left[-(|\theta_{\alpha}|/T)\right] \tag{11}$$

which would become exponentially small on approaching  $T_c = 0$  and presents a bump at  $|\theta_{\alpha}|/2$  where, we would expect [17]:

$$\theta_{\alpha} = -\theta_{\gamma} - 2\theta_{\beta} = -11.7 \text{ K} \tag{12}$$

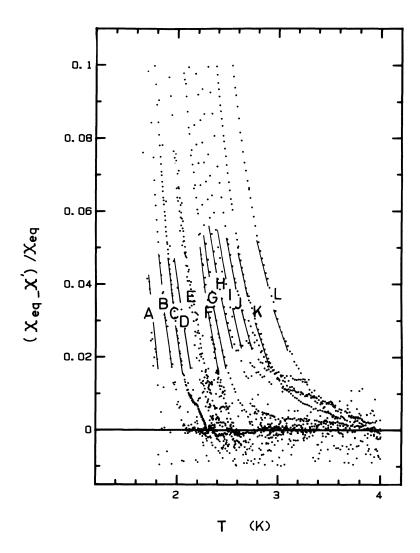


Fig. 7. — Values of the ratio  $(\chi_{eq} - \chi'_{\omega})/\chi_{eq}$  versus temperature for various frequencies: 0.02 Hz (A), 0.1 Hz (B), 0.3 Hz, (C), 3 Hz (D), 10 Hz (E), 30 Hz (F), 113 Hz (G), 211 Hz (H), 500 Hz (I), 1 kHz (J), 3 kHz (K) and 10 kHz (L). We arbitrary fix the irreversibility onset at the value  $\Delta \chi = 10^{-2}$ .

(this relation is the  $T_c \longrightarrow 0$  limit of  $\alpha = 2 - \gamma - 2\beta$  which can be written  $\theta_\alpha/T_c = 2 - \theta_\gamma/T_c - 2\theta_\beta/T_c$ ). We have measured the specific heat of Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub> down to 0.1 K to verify this equation. Figure 4 shows the experimental data corrected from the lattice contribution: the curve exhibits a rounded maximum at  $T \sim 4.5$  K and also a hump at about 1 K. This last small anomaly probably corresponds to a Schottky anomaly which is also observed in similar compounds and has been attributed to the presence of some impurities [18]. Our data extend to lower concentrations the measurements of Wong *et al.* [19] and they agree qualitatively. At lower temperature, we do not observe the expected exponential behaviour but we notice that there is a dominant contribution which varies like  $T^{1.6}$ . This does not make the situation better nor worst than in any other

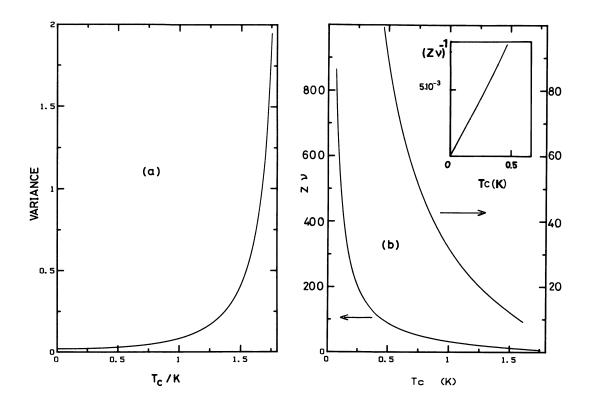


Fig. 8. — For different attempt values of  $T_c$  we calculate (a) the variance and (b) the exponent  $z\nu$  of the ajustment of  $\tau_{\max}(T)$  by a power law of  $t=1-T_c/T$ . The best values of exponent  $z\nu$  tend to increase like  $\theta/T_c$  with  $\theta=50$  K when  $T_c$  approaches zero for which the variance is minimum (inset).

spin glass where very little entropy is contained in the "critical contribution" which is controlled by a large exponent  $\alpha \sim 3$  and where the dominant contribution which accounts for most of the entropy is also described by a small power of the temperature.

The analysis of the static and dynamic critical data in Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub> comforts our previous conclusions that this system is representative of a class of universality different from that of usual spin glasses and presumably the same as for 2d Ising systems where  $T_c = 0$ . More recently untraditional behaviours have been reported in other spin glasses, e.g. by Dekker *et al.* [4] or Geschwind *et al.* [5]. The same dependence of the f.c. magnetisation on the cooling rate is reported in Rb<sub>2</sub>Cu<sub>0.78</sub>Co<sub>0.22</sub>F<sub>4</sub> and Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub>; it limits to temperature over 3.2 K and 1.7 K respectively the window where reliable static measurements can be obtained. They however exhibit definite differences which are more easily recognised and analysed in the figure 14. We have presented  $P_{\tau}(T)$  versus the dimensionless temperature  $T/\theta$  normalised by  $\theta$  which is a measure of the interaction for several spin glasses. The usual scaling hypothesis (Eq. (4)) leads to parallel straight lines which intersect the  $T/\theta$  axis at  $T/\theta = (z\nu)^{-1}$ . So that we have one line for a given universality class. Accepted spin glasses including simulations or 3d Ising spin glasses are reputed to have their  $z\nu$  between 6 and 10, i.e. they lie between CuMn 4.0% and aluminosilicate on the plot in figure 14. In practice we find in this range many RKKY systems and a number of classical systems as Eu<sub>x</sub>Sr<sub>1-x</sub>S. Our data in Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub> appear as the mere continuation to  $T_c = 0$ 

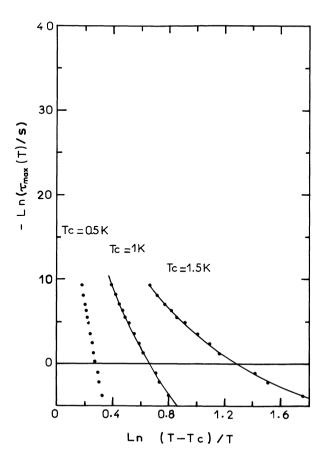


Fig. 9. — Plots of  $-\text{Ln}\tau_{\text{max}}(T)$  versus  $\text{Ln}(T-T_c)/T$  for three values of  $T_c$ : 0.5 K, 1 K and 1.5 K. The solid lines are simple guides to the eyes.

of the scaling hypothesis: this implies, as explained in equation 9, that  $z\nu$  diverges like  $\theta/T_c$  so that  $(1-T_c/T)^{-\theta/T_c} \longrightarrow \exp(\theta/T)$  and we find accordingly that  $a_3$ ,  $a_5$  and  $\tau$  are all power of each other and described by essential singularities as we expect when we reach a lower critical dimension.

The same plot shows that other systems such as  $Rb_2Cu_{0.78}Co_{0.22}F_4$  or  $Cd_{0.6}Mn_{0.4}$  Te are different and we would be prompted by this plot to interpret them with untraditional exponents ( $z\nu \sim 30$  and  $z\nu \sim 15$  in  $Rb_2Cu_{0.78}Co_{0.22}F_4$  and  $Cd_{0.6}Mn_{0.4}$  Te respectively). The authors reject these values as unphysically large on the basis of a tradition which is enforced by the theory and practice of ferromagnets. Following Fisher and Huse [20], they argue that for this transition  $\tau \sim \tau_0 \exp(W/T)$  where W, i.e.  $\ln(\tau)$ , rather  $\tau$  is a power of  $\xi$ . This leads to a family of solutions of the form  $\tau = \tau_0 \exp\left(\frac{B}{T-T_0}\right)^{\sigma}$  with among them the Fulcher law for  $\sigma=1$  and the solution that Binder and Young [21] promoted for  $T_0=0$ . Within this framework  $P_{\tau}(T)$  is a power law as we have:  $P_{\tau}(T) = B/\sigma T \left[(T-T_0)/B\right]^{\sigma+1}$  [17]. It is easy to understand that for a given experimental window there is always a choice of  $T_0$  and  $\sigma$  values for which the power law cannot be distinguished

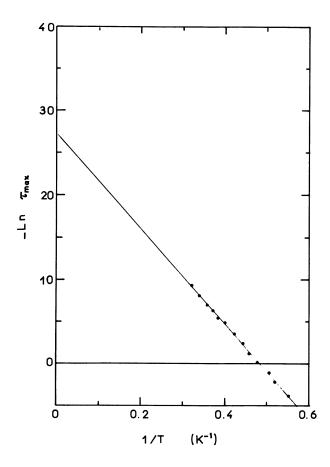


Fig. 10. — Plot of  $-\text{Ln}\tau_{\text{max}}(T)$  versus 1/T. The straight line corresponds to the fit with an Arrhenius law and  $\tau_0 = 1.5 \times 10^{-12} \text{s}$   $\theta = 57 \text{ K}$ .

from its tangent and the Fisher and Huse theory therefore will provide a family of solutions alternative to the one that provided by the critical slowing down: for example, rather than with  $T_c = 2.8$  K and  $z\nu = 32$ , as suggested by figure 14, Dekker *et al.* prefer to fit their data with the equation of Binder and Young ( $T_0 = 0$ ) and find  $\sigma = 3.2$ . The question is not to deny that alternatives exist to the slowing down equation which provide a comparable fit to the data. However we do not want a different theory for each of the different data shown in figure 14. The disturbing point finally with Fe<sub>0.30</sub>Mg<sub>0.70</sub>Cl<sub>2</sub> and a number of other systems is that we are confronted with different values of the exponents which we do not know how to justify by an obvious difference in either the spin or the space dimension. Perhaps, the reason for our discomfort is that we are willing to extend to spin glasses an argument (essentially the Harris criterion) which is valid for ferromagnets but may be wrong with spin glasses when frustration is present. What is, in the first place, the space dimension of a spin glass? In a disordered system with short range interactions we know from the percolation theory that the space dimension is different at short range and at long range with respect to  $\xi_s \sim (p - p_c)^{-\nu'}$  the correlation length of the percolation problem. If the magnetic correlation length is smaller than  $\xi_s$  the magnetic problem is at the dimension

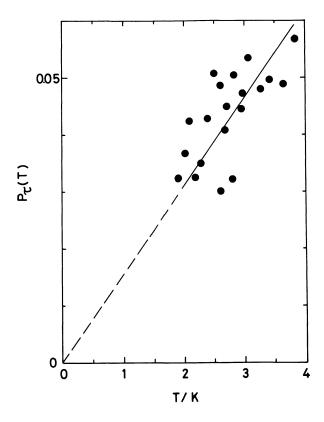


Fig. 11. — Plot of  $P_{\tau}(T) = -\delta \text{Ln} T / \delta \text{Ln} \tau$  versus T. The straight line is a simple guide to the eyes.

of the percolation cluster. If  $\xi > \xi_s$  it has the dimension of space. In a spin glass the magnetic problem makes averages at all ranges, smaller than a maximum one, which is  $\xi$ , so that the short range and the long range are mixed for each  $\xi$  in a proportion which may depend on how close to the percolation threshold we are. Perhaps rather than the notion of space dimension it would be appropriate to introduce some sort of dimension of the disorder which would evolve with the proximity of the percolation threshold.

The situation is particularly tricky in the bidimensional systems such as  $Fe_{0.3}Mg_{0.7}CL_2$  or  $Rb_2Cu_{0.78}Co_{0.22}F_4$ . The two dimensional character of the antiferromagnetic  $FeCl_2$  system is well established. It results from the great ratio  $(J_1/-J_1'\sim 10)$  between the intraplane ferromagnetic exchange  $J_1$  and the interplane antiferromagnetic interaction  $J_1'$ . It is more dubious that this character persists in the range of concentrations near the 2d percolation threshold where the spin glass effect occurs. As in most disordered systems there is a clear difference between the short range order which prevails at high temperature and the order which finally sets in. The effect of  $J_1$  is felt mainly through the constitution of short range 2d ferromagnetic clusters which then may well interact in the third dimension also. We are presently checking the possibility of exponents varying continuously with concentration in insulating spin glasses: such a possibility seems to be supported by the experimental evidence since the compounds with lower concentration in Mg show a flat plateau, as is generally observed in systems with a transition at finite  $T_c$ . In RKKY systems or when the interaction is long range we would expect a unique universality class: but the

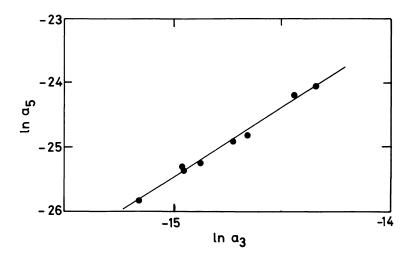


Fig. 12. — Plot of  $\ln a_5$  vs  $\ln a_3$ .

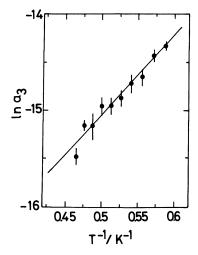


Fig. 13. — Arrhenius plot of ln a<sub>3</sub>

problem might depend on the way the interaction decreases with the distance due to the effect of a mean free path limitation or of exchange enhancement.

## Conclusion.

Our new dynamic data in the  $Fe_{0.3}Mg_{0.7}Cl_2$  spin glass have allowed us to enforce the evidence for a transition at  $T_c = 0$  in this Ising system. Its critical behaviour is different from that observed in typical 3d Ising systems as  $Fe_{0.35}Mg_{0.65}Br_2$  [22] or  $Fe_{0.25}Zn_{0.75}F_2$  [23] and we describe it by essential singularities considered as the limit of the traditional power law when  $T_c$  tends to zero.

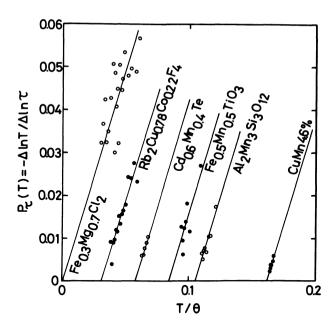


Fig. 14. — Plot of  $P_T(T)$  vs.  $T/\theta$  for Fe<sub>0.3</sub>Mg<sub>0.7</sub>Cl<sub>2</sub>, Rb<sub>2</sub>Cu<sub>0.78</sub>Co<sub>0.22</sub>F<sub>4</sub> [4], Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te [5], Fe<sub>0.5</sub>Mn<sub>0.5</sub>TiO<sub>3</sub> [24], amorphous aluminosilicate Al<sub>2</sub>Mn<sub>3</sub>Si<sub>3</sub>O<sub>12</sub> [25] and CuMn 4.6% [26].

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