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Short Communication

Dynamic evidence for a 2d - 3d crossover in the Ising spin glass $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$

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Résumé. — Nous présentons une étude du comportement critique dynamique du verre de spin de type Ising $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$. Les courbes de susceptibilités alternatives ont été déduites de mesures directes et de mesures par effet Faraday sur une large gamme de fréquence allant de 100 kHz à 0.05 Hz. La dépendance en température du temps de relaxation caractéristique du système ne peut être représentée ni par une loi de puissance ni par une loi d'Arrhénius sur toute la gamme de fréquence. Nous montrons que ce système présente un crossover de dimensionnalité 2d-3d quand la température décroît.

Abstract. — We report on magnetic dynamic properties in the Ising spin glass $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$. The ac susceptibility curves have been deduced from direct susceptibility and Faraday rotation measurements over a wide frequency range extending from 100 kHz down to 0.05 Hz. The temperature dependence of the characteristic relaxation time cannot be fitted by either a power or an Arrhenius law over the whole frequency range. We show that this system exhibits a dimensional 2d-3d crossover when lowering the temperature.

Introduction.

Theoretical studies, like Monte-Carlo simulations, high temperature developments and many experiments lead to a strong evidence for a nonzero transition temperature T_c in 3d Ising spin glasses [1-4]. On another hand, all theoreticians agree with a $T = 0$ transition temperature in 2d-Ising spin glasses. Recent static and dynamic magnetic studies on some disordered layered compounds like $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$ [5] and $\text{Rb}_2\text{Cu}_{0.8}\text{Co}_{0.2}\text{F}_4$ [6] suggest that they behave like 2d-Ising spin glasses. Up to now, only few studies have been devoted to test the influence on the dynamic critical behaviour of the spin glass dimensionality [7, 8]. $\text{Fe}_x\text{Mg}_{1-x}\text{X}_2$, ($\text{X} = \text{Cl}, \text{Br}$), stands as a convenient class of compound to perform such a systematic investigation since the interplane exchange interaction is reduced for the chloride and x can be varied over a large concentration range.

We report on a.c. susceptibility measurements in the Ising insulating spin glass $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$. Some results have been published earlier for this compound [4] but the too crude first analysis had not permitted to evidence a change in the critical behaviour with temperature. We will compare the magnetic dynamic properties of $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$ to those of $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$.

$\text{Fe}_x\text{Mg}_{1-x}\text{Br}_2$ is a layered compound characterised by a strong uniaxial anisotropy, $D \simeq 10$ K, conferring an Ising character to this system [9]. The in-plane NN interaction, $J_1 \simeq 10$ K, is ferromagnetic and the NN interplane, $J'_1 \simeq -3$ K, as well as the in plane NN interaction $J_2 \simeq -3$ K, are antiferromagnetic. Because of the competition between these interactions, the disordered compound $\text{Fe}_x\text{Mg}_{1-x}\text{Br}_2$ behaves like a spin glass for $x \leq 0.5$ [10]. We recall that, in $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$, $J_1 \simeq 10$ K and $J'_1 \simeq -1$ K, conferring an effective dimensionality lower than in $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$. Recently, we have observed a critical divergence of the coefficients of the non linear terms in the expansion of the static magnetisation of $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$ which accounts for a phase transition at a finite $T_c \simeq 1.95$ K [10].

Experimental.

The a.c. susceptibility and Faraday rotation measurements on $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$ have been done on a sample we used previously to test the static critical behaviour [10].

The temperature dependence of the longitudinal a.c. susceptibility over a wide frequency range, namely 0.05 Hz - 100 kHz, hence more than 6 decades, has been measured by using different experimental techniques. Figures 1a and 1b show some χ'_ω and χ''_ω experimental curves for various frequencies.

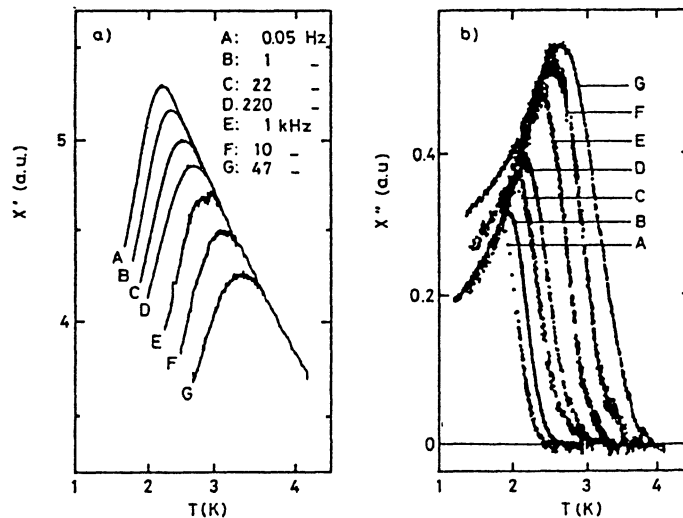


Fig. 1.— Plot of the real χ'_ω (a) and imaginary χ''_ω (b) part of the susceptibility vs. temperature for some frequencies. χ'_ω and χ''_ω are obtained from different methods depending on the frequency : A, B, C and D are obtained from direct susceptibility measurements ; E, F and G from Faraday effect measurements.

The first experimental apparatus, suitable for low frequency measurements, is composed of a mutual inductance device in a He^4 cryostat working down to 1 K. It is formed by a primary coil which provides the alternating magnetic field, and pick-up coils which collect the modulated signal.

The sample, made of small disc shaped pieces of single crystal and stacked in a tight container, is placed at the center of one of the two pick-up coils. Depending on the frequency, we have analysed the signal by different methods. In the lowest frequency range (0.05 Hz - 100 Hz), a 16 bit analog-to-digital converter associated to a PDP 11/73 computer allows to sample at 20 kHz both magnetic field and pick-up coil voltage and to calculate in real time the real χ'_ω and imaginary χ''_ω parts of the a.c. susceptibility. In the frequency range 100 Hz-4700 Hz, a double lock-in amplifier gave directly the modulus and the phase of the signal, from which we calculated χ'_ω and χ''_ω . In general, the experiments have been performed with an applied field of 1 Oe in order to remain in the linear response domain. At the lowest frequencies, we have also done measurements up to 20 Oe to increase the signal/noise ratio. The final conclusions of our study are unaffected by taking the data obtained at this field value into account. For the highest frequency, 4.7 kHz, the magnetic field has been kept weak enough (70 mOe) to reduce eddy currents induced in the cryostat.

The second series of experiments has been performed using a magneto-optical method derived from the Faraday effect [5, 11]. The sample used is one of the pieces constituting the stacked sample considered above. The sample has been cleaved to obtain a 2 mm thick platelet of good optical quality. The magnetic field ($H_{\text{eff}} = 1$ Oe) is provided by a 30 turns coil, having a small time constant, $\tau = 10 \mu\text{s}$, compatible with high frequency measurements. It is applied along the light beam ($\lambda = 6238 \text{ \AA}$) wave vector direction. The in-phase and out-of-phase signals related to the change in the Faraday rotation induced by the magnetic field are proportional to χ'_ω and χ''_ω , respectively.

The calibration of the a.c. susceptibility signal has been performed separately for the two sets of experiments. It has been realised by renormalising all χ'_ω curves in the paramagnetic region, i.e. for temperatures in the 3.5 K - 4.2 K range. For the highest frequency (100 kHz), a small error on the amplitude of χ'' is expected since irreversibilities occur at temperatures higher than 4.2 K. Note that the loss angle, $\varepsilon = \arctan(|\chi''|/|\chi'|)$, does not depend on the set-up calibration.

Discussion.

The aim of dynamic critical analysis is to know how the relaxation time of a system diverges at the transition temperature T_c . Assuming a magnetic transition at finite T_c , the relaxation time τ of a system is related to a power of the coherence length ξ , and therefore a power of the reduced temperature t :

$$\tau \sim \xi^z \sim t^{-z\nu} \text{ where } t = 1 - T_c/T$$

In order to determine the temperature dependence of τ , for each frequency ω , one generally assigns a temperature T_f for which τ is about $1/\omega$. Different criteria are used to define T_f and their respective validity have been extensively discussed [12-15]. Generally, because of the difficulty to measure χ'' , many authors define T_f from an empirical method by the temperature where χ'_ω is maximum. A second method consists in studying the break point between χ'_ω and χ_{eq} (the equilibrium susceptibility), defining T_f as the temperature where $(\chi'_\omega - \chi_{\text{eq}})/\chi_{\text{eq}}$ is set to a small constant. However, a finer criterium is to define T_f as $T(\arctan |\chi''_\omega|/|\chi'_\omega| = \text{constant})$ [12,13]. Our experimental facilities allow us to measure accurately χ' and χ'' over an extended frequency range and consequently, we have been able to determine T_f from these three criteria. Analysing our data according to these different criteria, we obtained similar conclusions, so that we shall expose here only the results obtained by the latest method. We have take a value of $\varepsilon = \arctan(|\chi''|/|\chi'|) = 0.01$

which is the smallest we can use to recover the $\tau(T)$ law over the whole experimental frequency range and warrant the validity of the condition $\omega\tau \ll 1$ allowing to write $\tau = \varepsilon/2\pi\omega$.

In order to search for a finite T_c , we have tried to get a good quality of the plot related to a power law, $\ln \tau$ vs. $\ln t$, for different values of T_c in the range 0 - 2.2 K ($T_f = 2.25$ K at 0.05 Hz). We have used the non linear variable $t = 1 - T_c/T$ because $T_f(47 \text{ kHz}) \simeq 3.5$ K and so, at high frequency, T_f will be greater than 1.1 T_c [16]. The best linear regression coefficient is obtained for $T_c = 1.9$ K associated with $z\nu = 17.5$ and $\omega_0 = 5 \times 10^{10}$ Hz. The Log-Log plot clearly exhibits a curvature (Fig. 2a) which proves that a power law is not suitable to fit the data over the whole

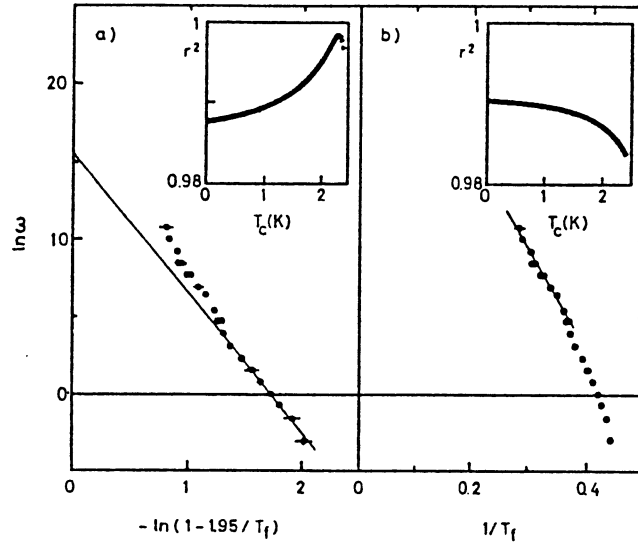


Fig. 2— Plots of $\ln \omega$ vs. $-\ln(1 - T_c/T_f)$ with $T_c = 1.95$ K (a) and $\ln \omega$ vs. $1/T_f$ (b) to check power and Arrhenius law, respectively. The insets in a) and b) show the linear regression coefficient of the power law for different attempts in value of T_c in low and high frequency range, respectively.

frequency range. Thus we restricted the analysis to low frequencies, where T_f is closer to T_c , and finally, the best regression coefficient is obtained in the range 0.05 Hz-22 Hz (inset Fig. 2a). The straightness of the Log-Log plot validates the power law, and leads to $T_c = 2.1$ K, $z\nu = 6$ and $\omega_0 = 10^6$ Hz. But reducing the frequency range implies a loss of accuracy on these values and we are more confident with the value of T_c previously determined from our static studies, 1.95 ± 0.05 K [10]. So, using this value of T_c we obtain $z\nu = 8.8 \pm 1.5$ and $\omega_0 = 10^{7 \pm 1}$ Hz. The corresponding fit is good within the experimental errors and gives parameters values expected for a 3d Ising spin glass[12,17,18].

In the high frequency range, 220 Hz - 47 kHz, the best power law fit is obtained for $T_c = 0$ (inset Fig. 2b) with a diverging $z\nu$ exponent. The characteristic relaxation time thus obeys to the Arrhenius law, $\tau = \tau_0 \exp(E/kT)$ (Fig. 2b), which stands as the natural limit of the power law when $T_c \rightarrow 0$ [5, 19]. The plot yields the values $\tau_0 \simeq 10^{-12}$ s and $E/k \simeq 65$ K which are close to that found in $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$ [5]. This means that in the temperature range far from T_c , $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$, as previously observed in $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$, behaves like a 2d system. Such a change in the critical behaviour of our system is confirmed from our nonlinear static study [10]. This study clearly shows that $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$ presents a transition at the finite temperature $T_c = 1.95$ K. However the analysis of the temperature dependence of the non linear terms of the expansion of the magnetisation

in terms of H/T in a temperature range far enough from T_c can be accounted by an Arrhenius law. In this temperature range, the variation over several orders of magnitude of these terms dismisses a simple paramagnetic behaviour which should imply a temperature independent value of the non linear terms of the magnetisation as observed in the system $\text{Fe} - \text{Al}_2\text{O}_3$ [20] and reflects the presence of cooperative effects, consistent with a transition at $T_c = 0$, convenient for 2d-Ising systems. The same data are also been analysed with theoretical laws proposed to describe the transition at $T_c = 0$ in spin glasses : $\tau = \tau_0 \exp(A/T^2)$ [21] or $\tau = \tau_0 \exp(A/T^3)$ [22]. Though these analysis lead to acceptable fits, the unphysical values of the characteristic relaxation time, $\tau_0 \sim 10^{-5}$ s, eliminate such laws.

In conclusion, we have analysed the frequency dependence of $T_f(\omega)$ for $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$ over a wide range between 0.05 Hz and 10^5 Hz. We have deduced the temperature dependence of the relaxation time τ , and in the close vicinity of T_c we found that $\tau(T)$ is well described by a power law associated with a finite T_c , $T_c = 1.95$ K, implying a 3d spin glass character, while at higher temperature, the power law fails, and a better fit being obtained with an Arrhenius law, compatible with a $T_c = 0$ transition temperature, hence a 2d spin glass character. So, though the precision we dispose of does not permit to determine the crossover temperature, this paper points out an experimental evidence for a 2d - 3d crossover in $\text{Fe}_{0.35}\text{Mg}_{0.65}\text{Br}_2$ when the temperature is lowered.

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