



HAL
open science

Mössbauer investigation of the dynamic critical behaviour in the spin glass system Rh 5 % Fe

C. Meyer, F. Hartmann-Boutron, Y. Gros

► **To cite this version:**

C. Meyer, F. Hartmann-Boutron, Y. Gros. Mössbauer investigation of the dynamic critical behaviour in the spin glass system Rh 5 % Fe. *Journal de Physique*, 1986, 47 (8), pp.1395-1404. 10.1051/jphys:019860047080139500 . jpa-00210333

HAL Id: jpa-00210333

<https://hal.science/jpa-00210333v1>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



HAL Authorization

Classification
 Physics Abstracts
 76.80 — 75.40

Mössbauer investigation of the dynamic critical behaviour in the spin glass system Rh 5 % Fe

C. Meyer, F. Hartmann-Boutron and Y. Gros

Laboratoire de Spectrométrie Physique, U.S.T.M. Grenoble,
 B. P. 87, 38402 Saint Martin d'Hères Cedex, France

(Reçu le 13 mars 1986, accepté le 10 avril 1986)

Résumé. — Nous avons effectué une étude Mössbauer détaillée du verre de spin Rh 5 % Fe en mettant l'accent sur le domaine de température 12,5 K-16 K (au-dessus de $T_g = 12,5$ K) où une structure hyperfine commence à apparaître. Rh Fe est mieux adapté à une telle étude que Au Fe, du fait de l'absence d'effets quadrupolaires de paires. Nous trouvons qu'une tentative d'interprétation de la largeur de raie Mössbauer en termes d'effets critiques dynamiques n'est possible que jusqu'à $T = 15$ K et conduit à un exposant critique $w \sim 4-5$. Au-dessous de 15 K la meilleure analyse des spectres s'obtient en supposant qu'il s'agit de spectres Zeeman statiques (avec élargissement inhomogène) associés à des valeurs moyennes $\langle S_i \rangle$ en temps non nulles des spins électroniques. La nature exacte, statique ou quasi statique de ces $\langle S_i \rangle$ au-dessus de T_g reste à élucider.

Abstract. — A detailed Mössbauer study of the spin glass alloy Rh 5 % Fe has been performed with emphasis on the temperature zone 12.5 K-16 K (above $T_g = 12.5$ K), where a hyperfine structure begins to appear. For such a study Rh Fe is more convenient than Au Fe, due to the absence of pair quadrupole effects. It is found that a tentative interpretation of the Mössbauer linewidth in terms of dynamic critical effects is only possible down to $T = 15$ K, leading to a critical exponent $w \sim 4-5$. Below 15 K the spectra are best analysed as inhomogeneously broadened static Zeeman patterns associated with non-zero time average values of the electronic spins $\langle S_i \rangle$. The exact nature, static or quasi static, of these $\langle S_i \rangle$'s above T_g has yet to be elucidated.

1. Introduction.

In a recent Mössbauer reinvestigation of the spin glass compound Au 3 % Fe [1] we showed that two different characteristic temperatures could be defined: the temperature of the cusp of the susceptibility, T_g , and a Mössbauer temperature $T_M > T_g$. T_M was defined as the temperature of the sharp break in the thermal variation of the linewidth of the high temperature single line. Below T_M a Zeeman spectrum with broad lines progressively develops. Its evolution cannot be explained by relaxation models which assume a progressive slowing down of large amplitude fluctuations (as in superparamagnetic clusters for example). On the other hand, within the assumption of a static inhomogeneous broadening, it is possible to derive from the spectra an average modulus $\langle H_n \rangle$ of the hyperfine fields $H_{n,i}$ at the irons, whose thermal variation looks quite similar to a Brillouin curve corresponding to a transition temperature T_M . The distribution $P(H_n)$ of $|H_{n,i}|$ around $\langle H_n \rangle$ is then attributed to the effect, on the electronic spins S_i , of

the distribution of exchange fields, which influences their average values $\langle S_i \rangle$. There is however some ambiguity in the interpretation of the spectra between T_g and T_M , since they consist of a single line which is very broad but not yet resolved.

In reference [1] we tentatively attributed the line-broadening above T_M to paramagnetic relaxation phenomena in order to obtain some information about the dynamic behaviour of the system. No such attempt was made in the zone $T_g < T < T_M$. Indeed the high temperature spectra of Au 3 % Fe are complicated by the presence of quadrupole splittings due to pair effects. The determination of the lineshape and hence of the relaxation rate is therefore not precise enough for looking at possible critical phenomena.

A better candidate for such a study seems to be the rhodium-iron system. First it provides a much better signal than AuFe, in which gold is highly absorbing for ^{57}Fe Mössbauer γ rays. Second, according to Window *et al.* [2], the high temperature spectrum of Rh 5 % Fe is not disturbed by quadrupole effects,

this being probably due to equal electric charges on Rh and Fe atoms. It is therefore easier to follow the paramagnetic lineshape as a function of temperature, especially in the momentous region $T \sim T_M$, and to look for critical phenomena. That was the aim of the present work.

2. Experimental results.

A sample of Rh Fe (5 % atomic) was provided by T. Cranshaw, AERE Harwell Laboratories. It was the sample used by Window *et al.* [2] more than fifteen years ago. We have recovered their results as concerns T_M and $\langle H_n(0 \text{ K}) \rangle$, which means that the sample is unaltered. However the point of view of the present study is different from theirs, because at that time the problem of the exact nature of the spin glass transition was not set in the same terms.

The cusp temperature T_g of the Rh Fe sample was determined by measuring the d.c. susceptibility in a 20 G external field with a SQUID magnetometer. This yields $T_g = 12.8 \pm 0.2 \text{ K}$ from the zero field cooled curve while a maximum at $12.2 \pm 0.2 \text{ K}$ is obtained from the field cooled one.

Mössbauer spectra were taken at room temperature (as a reference for the paramagnetic linewidth) and between 40 and 1.3 K. Figure 1 displays most of these spectra, covering the whole range of interest. At room temperature, fitting with a single Lorentzian line is satisfying. Down to around 14.5 K the spectra can be fitted in the same way, with an increasing linewidth,

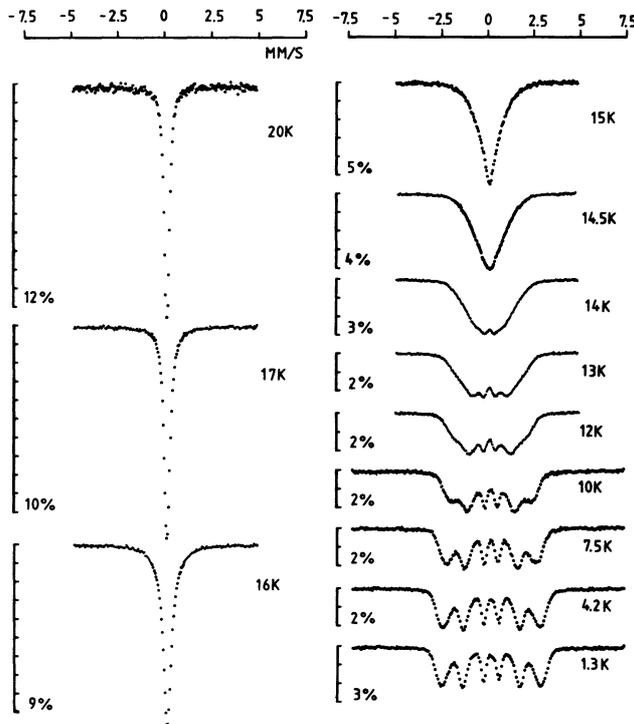


Fig. 1. — ^{57}Fe Mössbauer experimental spectra for Rh Fe (5 at %).

rather poorly however for $T = 14.5 \text{ K}$ (Fig. 2). In table I and figure 4 we report the evolution of the F.W.H.M. linewidth $2\Gamma_{\text{exp}}(T)$ as a function of temperature.

At 14 K small magnetic lines definitely appear in the centre of the spectrum. The Zeeman hyperfine splitting spreads out, becoming more and more resolved as the temperature goes down. These spectra at $T \leq 14 \text{ K}$ can be interpreted in terms of static hyperfine fields characterized by the distribution $P(H_n)$ of their moduli around an average value $\langle H_n \rangle = \int H_n P(H_n) dH_n$. Figure 3 and table II show the comparison between experimental spectra and theoretical spectra computed with the distribution $P(H_n)$ represented on the side. Note that this fit can be performed even above 14 K, when the spectrum reduces to a single broad line. In this range however ($14 \text{ K} < T < 16 \text{ K}$) the mean hyperfine field modulus $\langle H_n \rangle$ is small, the width of $P(H_n)$ is rather large and the accuracy is poor. In addition $P(H_n)$ has a finite value for $H_n = 0$.

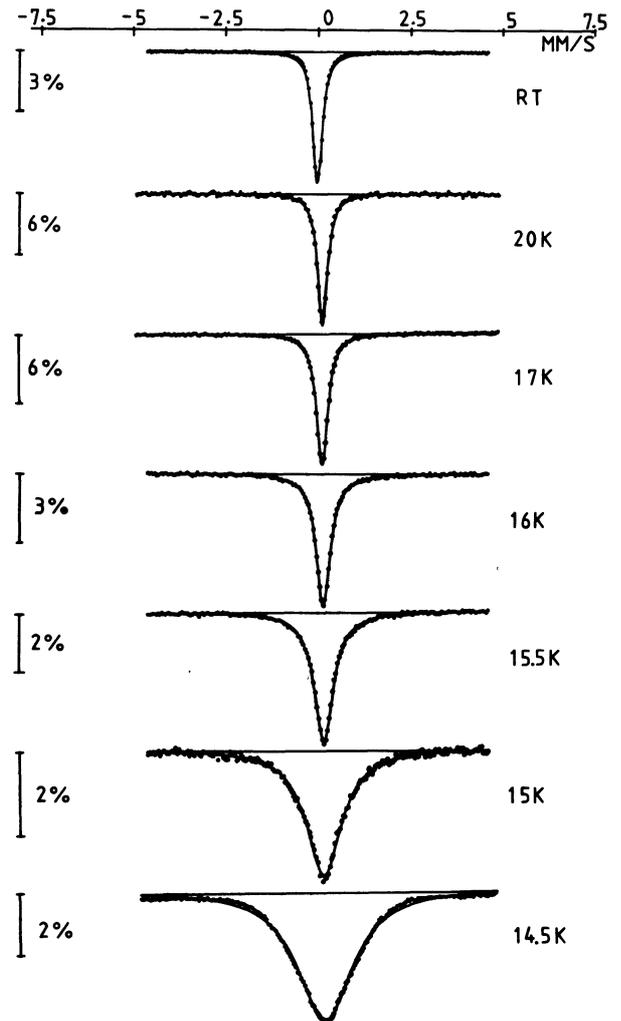


Fig. 2. — Spectra in the temperature range 295 K-14.5 K, as fitted with one Lorentzian line.

Table I. — *Temperature dependence between 295 K and 14.5 K of : the overall fitted Lorentzian linewidth : $2\Gamma_{\text{exp}}(T)$; the excess overall linewidth : $2\Delta\Gamma(T)$ with respect to room temperature; the correlation time τ_c derived from equation (3) for $S = 1/2$.*

T (K)	$2\Gamma_{\text{exp}}$ (mm/s)	$2\Delta\Gamma$ (mm/s)	τ_c (s)
295	0.318	0	
40	0.32	0.002	1.11×10^{-12}
20	0.322	0.004	2.22×10^{-12}
19	0.322	0.004	2.22×10^{-12}
<hr/>			
18	0.352	0.034	1.89×10^{-11}
17.5	0.357	0.039	2.17×10^{-11}
17.25	0.366	0.048	2.67×10^{-11}
17	0.367	0.049	2.726×10^{-11}
16.75	0.409	0.0914	5.085×10^{-11}
16.5	0.402	0.0846	4.707×10^{-11}
16.25	0.437	0.119	6.621×10^{-11}
16	0.462	0.144	8.012×10^{-11}
15.8	0.535	0.217	1.207×10^{-10}
15.7	0.576	0.258	1.435×10^{-10}
15.5	0.622	0.304	1.691×10^{-10}
15.4	0.738	0.42	2.337×10^{-10}
15.3	0.792	0.474	2.637×10^{-10}
15.2	0.896	0.578	3.216×10^{-10}
15.1	0.956	0.648	3.605×10^{-10}
15	1.167	0.849	4.724×10^{-10}
<hr/>			
14.5	1.796	1.478	8.224×10^{-10}
<hr/>			
accuracy	± 0.01	± 0.02	$\pm 10^{-11}$

In figure 4, $\langle H_n \rangle$ and $2\Gamma_{\text{exp}}(T)$ are plotted as functions of temperature together with Γ_H , the half width at half height of $P(H_n)$. As in Au 3% Fe we note a sharp coincidence, when T decreases, between the abrupt rise of the overall linewidth $2\Gamma_{\text{exp}}(T)$ and the appearance of the mean hyperfine field $\langle H_n \rangle$. Using the break in $2\Gamma_{\text{exp}}(T)$ (or more precisely the intersection of the tangents to the low slope and high slope parts of the curve) we obtain a Mössbauer transition point at $T_M = 15.8 \pm 0.2$ K $>$ $T_g \approx 12.5$ K. Figure 5 shows the fairly similar behaviour observed for the reduced hyperfine field $\langle H_n(T) \rangle / \langle H_n(0) \rangle$ and for the $S = 1/2$ Brillouin law curve as a function of the reduced temperature T/T_M . Indeed, according to the empirical rule $H_n = 147$ kG per μ_B , i.e. per half unit of spin, the value $\langle H_n(0) \rangle = 160$ kG suggests that $S = 1/2$ (rather than $S = 1$ as was admitted in Au Fe, in which $\langle H_n(0) \rangle = 250$ kOe). The flattening of the curve $\langle H_n(T) \rangle / \langle H_n(0) \rangle$ with respect to the Brillouin curve is reminiscent of what is observed in amorphous materials. Finally the presence of a maximum in the thermal variation of Γ_H , already observed in Au 3% Fe, was discussed in reference [1].

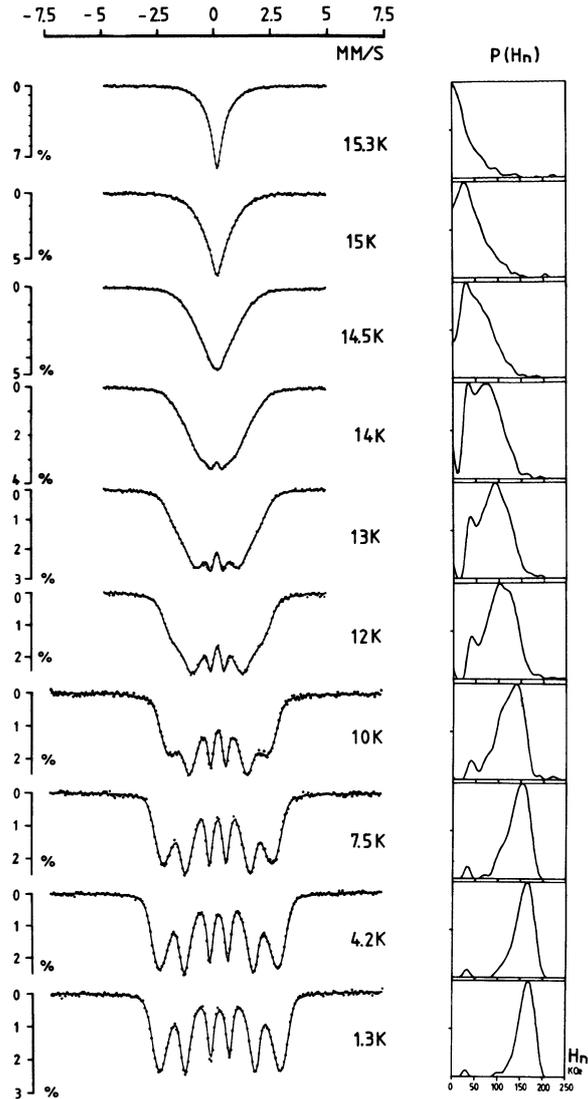


Fig. 3. — Spectra in the temperature range 15.3 K-1.3 K as fitted with a hyperfine field distribution $P(H_n)$ represented on the side.

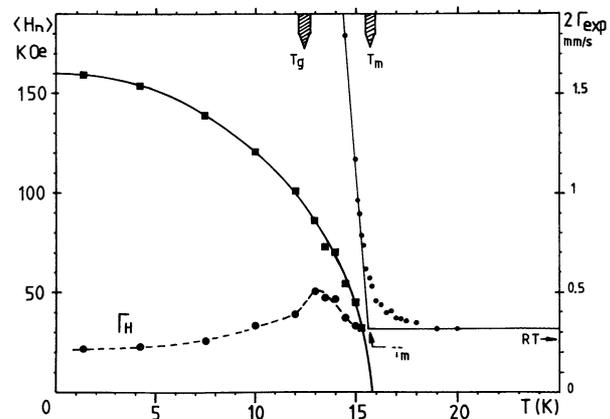


Fig. 4. — Thermal variations of the overall experimental linewidth $2\Gamma_{\text{exp}}$ (●), the average hyperfine field $\langle H_n \rangle$ (■) and the halfwidth Γ_H of the hyperfine field distribution (⊕). As in Au Fe, Γ_H exhibits a maximum at $T \sim T_g$.

Table II. — Temperature dependence between 15.3 K and 1.3 K of hyperfine field parameters (kOe) : the average hyperfine field : $\langle H_n \rangle = \Sigma H_n P(H_n) \delta H_n$ with $\Sigma P(H_n) \delta H_n = 1$; the standard deviation : $\sigma_H = \sqrt{\Sigma P(H_n) (H_n - \langle H_n \rangle)^2 \delta H_n}$; the hyperfine field H_{\max} defined as the position of the maximum of the main distribution peak, or as the centre of the half height width of the distribution when this one is double peaked; the half width at half height Γ_H of $P(H_n)$.

T (K)	$\langle H_n \rangle$	σ_H	H_{\max}	Γ_H
1.3	159.5	26.5	167	21
4.2	153.8	29.2	164	23
7.5	139.2	30.8	153	26
10	121.2	34.1	138	33.5
12	100.9	36.5	103	38.5
13	86.5	35	93	50.5
13.5	73.1	35.1	78	47.5
14	70.3	34.8	73	46.5
14.5	54.8	34	28	37.5
15	44.7	34.4	26	32.5
15.3	31.9	30.6	0	14

$\pm 0.5 \quad \pm 0.5 \quad \pm 1 \quad \pm 1$

To summarize : Mössbauer spectra obtained on the two compounds Au 3 % Fe and Rh 5 % Fe are quite similar. We will now examine in more detail the intermediate region $T_g < T < T_M$ where there is some uncertainty in the fitting process. Our aim is to try and clarify the meaning of T_M with respect to T_g .

3. Discussion of spectra at $T > 15$ K.

First of all we checked that the whole temperature evolution of the spectra between 300 K and 0 K cannot be explained by a continuous slowing down of spins with fixed length. For this, we compared the spectra with a number of relaxation spectra computed with the help of different models as described in [1]. None of these models was satisfying. The assumption of essentially static spectra below $T_g = 12.5$ K seems then rather reasonable. On the other hand dynamical effects are present above $T_M = 16$ K and could perhaps also play a role in the range ($T_g = 12.5$ K, $T_M = 16$ K). Indeed the meaning of T_M is not unam-

It leads to :

$$\Delta\Gamma = \frac{A_g^2}{\hbar^2} \left[\frac{15}{4} \frac{A_e^2}{A_g^2} - \frac{5}{2} \frac{A_e}{A_g} + \frac{3}{4} \right] \int_0^\infty dt \overline{S_z(0) S_z(t)} \quad (1)$$

where A_e, A_g are the hyperfine coupling constants in the excited and ground Mössbauer states ($\mathcal{H}_e = A_e \mathbf{I}_e \cdot \mathbf{S}$, $\mathcal{H}_g = A_g \mathbf{I}_g \cdot \mathbf{S}$). Neglecting the hyperfine anomaly, the ratio A_e/A_g is equal to that of the nuclear Lande factors, which is well known.

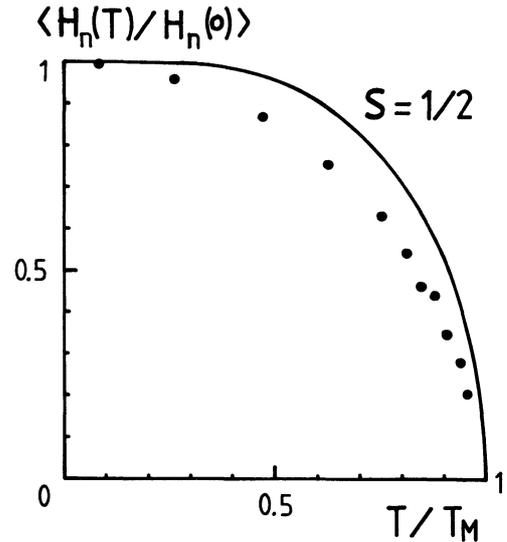


Fig. 5. — Reduced hyperfine field $\langle H_n(T) \rangle / \langle H_n(0) \rangle$ vs. the reduced temperature T/T_M as compared to the Brillouin law curve for $S = 1/2$ and $T_M = 16$ K.

biguous because, according to $P(H_n)$, down to 14.5 K some of the iron sites still have $|\mathbf{H}_{n_i}| = 0$ i.e. $|\langle \mathbf{S}_i \rangle| = 0$.

Because of this uncertainty between T_g and T_M , we may try another interpretation. Returning to the plot of the overall linewidth $2\Gamma_{\text{exp}}(T)$ in figure 4, we observe that although the large increase of $2\Gamma_{\text{exp}}$ starts around 16 K, the linewidth seems to be diverging towards a lower temperature, T_g for example. Therefore we can tentatively analyse the linewidth broadening in terms of a slowing down of critical fluctuations above a transition temperature T_g . The shape of the spectrum is a good Lorentzian line down to $T \sim 15$ K; when $T \geq 15$ K, its broadening $\Delta\Gamma = \Gamma_{\text{exp}}(T) - \Gamma_{\text{exp}}(300 \text{ K})$ can then be analysed with the help of the Bradford Marshall model [3] in which $\Delta\Gamma$ is proportional to the correlation time τ_c of the electronic spin \mathbf{S} .

This model applies to the relaxation of a nuclear spin by a paramagnetic spin \mathbf{S} with isotropic fluctuations. In the Mössbauer case there is a complication due to the existence of two nuclear states with two different nuclear spins \mathbf{I}_e and \mathbf{I}_g . The treatment uses second order perturbation and is valid only if :

$$\begin{aligned} \tau_c &\ll \tau_L \text{ (hyperfine Larmor period)} \\ \Delta\Gamma \tau_c &\ll 1. \end{aligned}$$

Let us define an average autocorrelation time τ_c by

$$\tau_c = \frac{1}{\langle S_z^2 \rangle} \int_0^\infty dt \overline{S_z(0) S_z(t)} = \frac{3}{S(S+1)} \int_0^\infty dt \overline{S_z(0) S_z(t)}. \quad (2)$$

Equation (1) then becomes

$$\Delta\Gamma = \frac{A_g^2}{\hbar^2} S^2 \frac{S+1}{3S} \left[\frac{15}{4} \frac{A_c^2}{A_g^2} - \frac{5}{2} \frac{A_c}{A_g} + \frac{3}{4} \right] \tau_c. \quad (3)$$

Note that this same form is obtained with the usual (but more restrictive) assumption of an exponential autocorrelation function :

$$\overline{S_z(0) S_z(t)} = \langle S_z^2 \rangle e^{-t/\tau_c} = \frac{S(S+1)}{3} e^{-t/\tau_c}. \quad (4)$$

Previous analyses of Mössbauer experiments (or similar experiments like PAC) performed in standard ferro or antiferromagnets near the magnetic transition point, T_{ord} , also use Bradford Marshall (or equivalent) formulas. It is admitted by the authors [4-8] that near T_{ord} , the average correlation time τ_c diverges according to a law

$$\tau_c = \left(\frac{T - T_{ord}}{T_{ord}} \right)^{-w} \quad (5)$$

where :

$$w = \nu(z + 2 - d - \eta) \quad (6)$$

in which d is the dimensionality, ν and η are static critical exponents and z is a dynamic critical exponent which depends both on d and on n , number of components of the order parameter (see Tab. I of [6], Tab. II of [7]).

The average correlation time τ_c defined above seems to be identical to the τ_{av} introduced by Ogielski [9] and w seems to coincide with his $z_{av} \nu$. There is however an important difference in that Ogielski writes that in spin glasses, due to the different nature of the order parameter :

$$z_{av} \nu = \frac{\nu}{2} (2z + 2 - d - \eta). \quad (7)$$

Here we will not concern ourselves about the difference between this expression and the preceding one for w . Our only interest will consist in trying to obtain experimental values for w . The numerical simulations of Ogielski [9] on an Ising spin glass ($d = 3, n = 1$) lead him to : $z_{av} \nu = 7.2 \pm 0.1$. However our experiments on Rh Fe are relative to a Heisenberg spin glass ($d = 3, n = 3$). The two situations are therefore different.

In figure 6 our experimental results for $2 \Delta\Gamma(\propto \tau_c)$ are plotted as a function of $t = (T - T_g)/T_g$ in a double logarithmic scale. Note that the error bars are reasonable only for 15 K $\lesssim T \lesssim$ 18 K corresponding to $0.2 \lesssim (T - T_g)/T_g \lesssim 0.5$ (while at 14.5 K an adjustment with a Lorentzian line is not very good). Allowing for the error bars, the experimental points are well aligned on a straight line, and the best fit corresponding to the law :

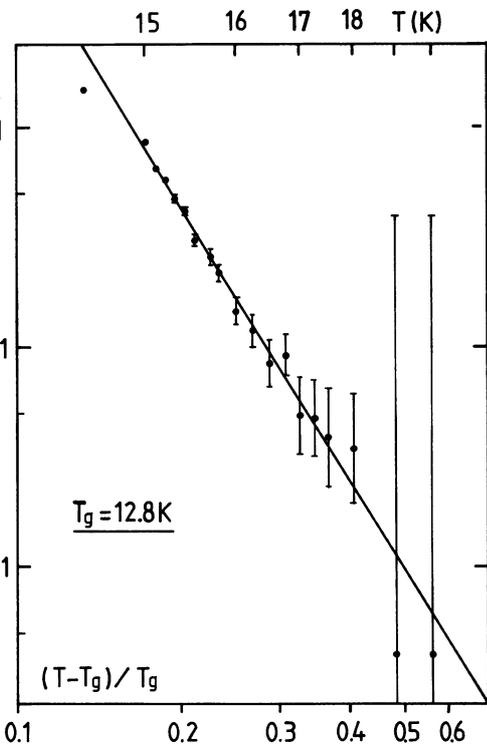


Fig. 6. — Plot of $2 \Delta\Gamma$ vs. $(T - T_g)/T_g$ on a double logarithmic scale.

$$\log(2 \Delta\Gamma) = \log(2 \Delta\Gamma)_0 - w \log t \quad (8)$$

yields

$$w = 4.12 (T_g = 12.8 \text{ K})$$

or

$$w = 4.94 (T_g = 12.2 \text{ K}).$$

As shown in figure 7, experimental data for $2 \Gamma_{\text{exp}}(T)$ (Tab. I) are well reproduced by computed curves based on the equations ($\Gamma_{\text{exp}}(T)$ in mm/s) :

$$2 \Gamma_{\text{exp}}(T) = 2 \Gamma_{\text{exp}}(300 \text{ K}) + 0.000569 t^{-4.12} \quad (T_g = 12.8 \text{ K})$$

$$2 \Gamma_{\text{exp}}(T) = 2 \Gamma_{\text{exp}}(300 \text{ K}) + 0.000548 t^{-4.94} \quad (T_g = 12.2 \text{ K}).$$

In order to extract the value of τ_c from $\Delta\Gamma$ we must know $A_g S$ and $(S + 1)/3 S$. The quantity $A_g S$ is deduced from the average hyperfine field at 0 K : $|\langle H_n(0) \rangle| = |A_g S/\hbar\gamma_g| = 160 \text{ kOe}$ and we assume that $S = 1/2$ whence $(S + 1)/3 S = 1$. We then get that (Tab. I) :

$$\tau_c = \tau_0 \left(\frac{T - T_g}{T_g} \right)^{-w} \quad (9)$$

with

$$\tau_0 = 3.2 \times 10^{-13} \text{ s}, \quad w = 4.12 \quad \text{for } T_g = 12.8 \text{ K}$$

$$\tau_0 = 3 \times 10^{-13} \text{ s}, \quad w = 4.94 \quad \text{for } T_g = 12.2 \text{ K}.$$

The use of these laws at $T = 14.5 \text{ K}$ leads to $\Delta\Gamma \tau_L \sim 10^{-1}$ i.e. we are at the limit of validity of the Bradford Marshall model, which might, at first sight, explain the failure of the Lorentzian fit.

Note that if we use the slightly different scaling law introduced by Souletie *et al.* [10]

$$\tau = \tau^* \left(\frac{T - T_g}{T} \right)^{-w} \quad (10)$$

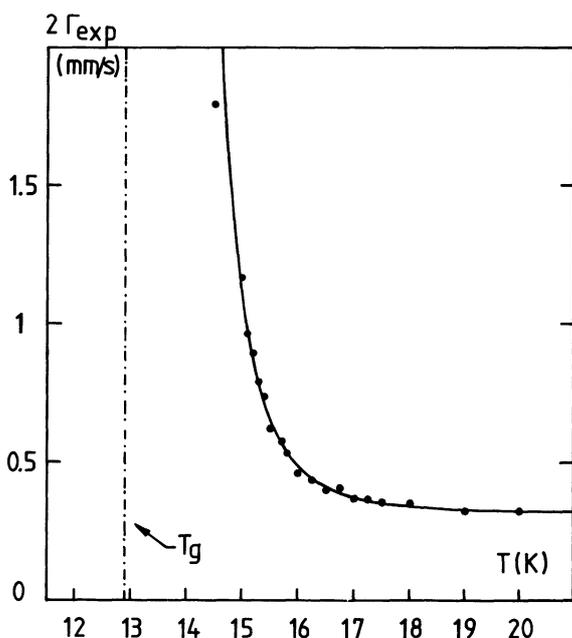


Fig. 7. — Comparison between the experimental thermal variation of the overall experimental linewidth $2 \Gamma_{\text{exp}}$ and the calculated curve deduced from equation (8) with $T_g = 12.8 \text{ K}$.

which can be used farther above T_g , we find that :

$$\tau^* = 2.5 \times 10^{-14} \text{ s} \quad w = 5.1, \quad \text{for } T_g = 12.8 \text{ K}$$

$$\tau^* = 9.1 \times 10^{-15} \text{ s} \quad w = 6.4, \quad \text{for } T_g = 12.2 \text{ K}.$$

Finally we have also checked that the temperature dependence of τ cannot be explained by an Arrhenius law, as is expected in the absence of a transition [11]. Indeed if we fit τ versus T with an expression :

$$\tau = \tau^* \exp[E_a/(k_B T)^\sigma] \quad (11)$$

we obtain unphysical values for τ^* and E_a ; for example :

$$\sigma = 1 : E_a/k_B = 340 \text{ K}, \quad \tau^* = 6.7 \times 10^{-20} \text{ s}$$

$$\sigma = 2 : E_a/k_B = 2700 \text{ K}, \quad \tau^* = 2.6 \times 10^{-15} \text{ s}.$$

We have now to consider the situation below 15 K where Lorentzian fits are no longer adequate.

4. Discussion of spectra between 15 K and T_g and below T_g .

Let us now consider the spectra between T_g ($\sim 12.5 \text{ K}$) and 15 K. Two interpretations can be thought of :

— *Interpretation 1* : Paramagnetic relaxation down to T_g . We found that at 14.5 K the Bradford Marshall model is no longer valid. However there exists a stochastic model due to Dattagupta and Blume [12-13] which is valid whatever the jump circular frequency ($1/\tau_c$) and which takes a form similar to the Bradford and Marshall one in the fast relaxation limit. If the Mössbauer data at 14.5 K, 14 K... can be fitted with this model, then the values of τ_c obtained should be in agreement with the thermal variation determined above for $T \geq 15 \text{ K}$. If this not so, we will have to turn to the second interpretation.

— *Interpretation 2* : Static broadening.

Below 15 K the broadening of the spectrum cannot be explained by critical dynamical effects, but seems to be essentially static, since a fit with a field distribution looks quite reasonable below 15 K.

Let us first examine interpretation 1, using the Dattagupta and Blume model [12-13]. This strong collision model describes the interaction between a nucleus and a classical field jumping randomly between all possible directions in space, the random process being characterized by a circular frequency λ .

If it is assumed that in the ground and excited states respectively :

$$\begin{aligned} \mathcal{H}_{\text{hf}}^g/\hbar &= -\mathbf{h}_0 \cdot \mathbf{I}_g \\ \mathcal{H}_{\text{hf}}^e/\hbar &= -\mathbf{h}_1 \cdot \mathbf{I}_e \end{aligned} \quad (12)$$

with $h_0 = \gamma_g H$, $h_1 = \gamma_e H$, then the Dattagupta-

Blume theory leads to a Mössbauer lineshape :

$$I(\omega) \sim G_1(p) = \frac{G_1^0(p + \lambda)}{1 - \lambda G_1^0(p + \lambda)} \quad (13)$$

where

$$G_1^0(p + \lambda) = \frac{1}{2} \frac{p + \lambda}{(p + \lambda)^2 + \frac{1}{4}(3h_1 - h_0)^2} + \frac{1}{3} \frac{p + \lambda}{(p + \lambda)^2 + \frac{1}{4}(h_1 - h_0)^2} + \frac{1}{6} \frac{p + \lambda}{(p + \lambda)^2 + \frac{1}{4}(h_1 + h_0)^2} \quad (14)$$

and $p = \Gamma/2 - i\omega$ for monochromatic irradiation, $p = \Gamma - i\omega$ for a real experiment ($\Gamma = 1/\tau_n$, inverse nuclear lifetime).

In the fast relaxation limit ($p + \lambda \gg h_1, h_0$), $G_1(p)$ reduces to :

$$G_1(p) = \left\{ p + \frac{h_0^2}{3\lambda} \left[\frac{15}{4} \left(\frac{h_1}{h_0} \right)^2 - \frac{5}{2} \frac{h_1}{h_0} + \frac{3}{4} \right] \right\}^{-1} \quad (15)$$

This formula has the same form as the Bradford-Marshall formula provided we make the identifications :

$$\lambda = 1/\tau_c \quad (16)$$

$$\frac{h_0^2}{3} = \frac{A_g^2 S(S+1)}{3\hbar^2} = \gamma_g^2 H_n(0)^2 \left(\frac{S+1}{3S} \right) \quad (17)$$

A whole set of spectra obtained with $S = 1/2$ and $H_n(0) = 160$ kOe, and with varying λ , is represented in figure 8; they clearly differ from those in figure 2. (Note that the Zeeman splitting provided by D-B model at 0 K is not quite equal to the experimental one because of the factor $(S+1)/S$ in Eq. (17)). In addition in figure 9A we tried to compare the spectra yielded by this calculation with the experimental spectra at 14.5 K and 14 K. For this, only the overall linewidth (i.e. λ) and the integrated area were adjusted : the line shape is not the same as the experimental one. On the other hand in figure 9B we computed the expected spectra at the same temperature, using the values of $\tau(T)$ derived from the results obtained above 15 K (Eq. (9) with $T_g = 12.8$ K) :

$$\tau(14.5 \text{ K}) = 1.3 \times 10^{-9} \text{ s}$$

$$\tau(14 \text{ K}) = 5.4 \times 10^{-9} \text{ s}.$$

Here too the disagreement is complete.

Consequently, if it makes sense, a dynamical analysis of the Mössbauer lineshape assuming a free fluctuating spin can only hold down to 15 K. Below this temperature and down to 0 K the spectra are very

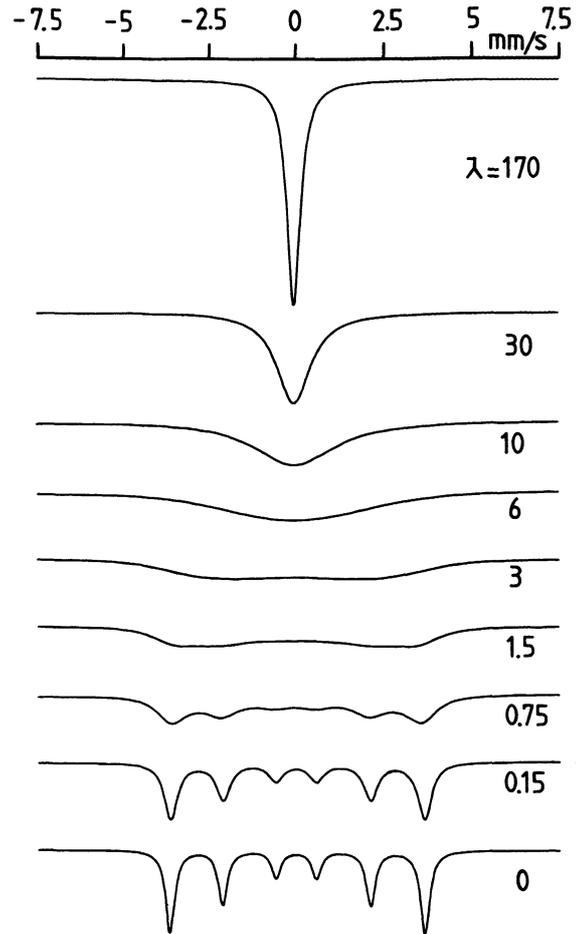


Fig. 8. — An example of some calculated spectra in the Dattagupta and Blume model assuming $S = 1/2$, $H(0) = 160$ kOe, $\Gamma = 0.159$ mm/s = $\Gamma_{exp}(RT)$ and different values of λ in mm/s.

well interpreted as static magnetic patterns in the effective field model, with an average hyperfine field $\langle H_n \rangle$ which follows (approximately) a Brillouin law as in conventional magnets. As for the broadening of the lines, it seems to be mainly static. As a matter of fact, as discussed in [1], one could think of a relaxation model (Dekker model [14]) in which the spin jumps between its states in a non zero molecular field following a Brillouin law. However if relaxation broadening is important, the spectrum is strongly distorted and there is no reason why it would be characterized by an apparent h.f. field following a Brillouin law. As an illustration of these considerations we report spectra at 14.5 K and 15.3 K computed with the Dekker model (with $S = 1/2$) under the assumption that the apparent transition temperature for the Mössbauer nucleus is $T_M = 16$ K (it cannot be taken equal to $T_g \approx 12.5$ K since a hyperfine structure begins to develop at 14 K). The molecular field only depends on T_M and once T is fixed, there remains a single unknown parameter which is the inverse average relaxation time Ω_c [14]. Figure 10 shows that even by varying Ω_c it is not possible to recover the experimental spectra.

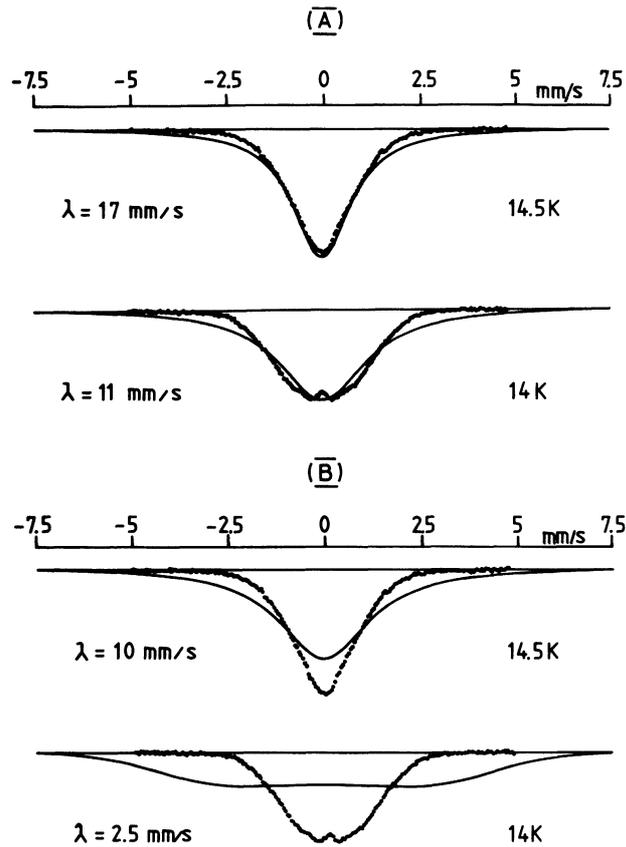


Fig. 9. — Comparison of the experimental spectra for 14.5 K and 14 K with calculated ones in the Dattagupta and Blume model within the same assumptions as in figure 8 : A) upon adjusting the overall linewidths; B) upon taking values of $\lambda = 1/\tau_c$ with τ_c obeying the law of equation (9).

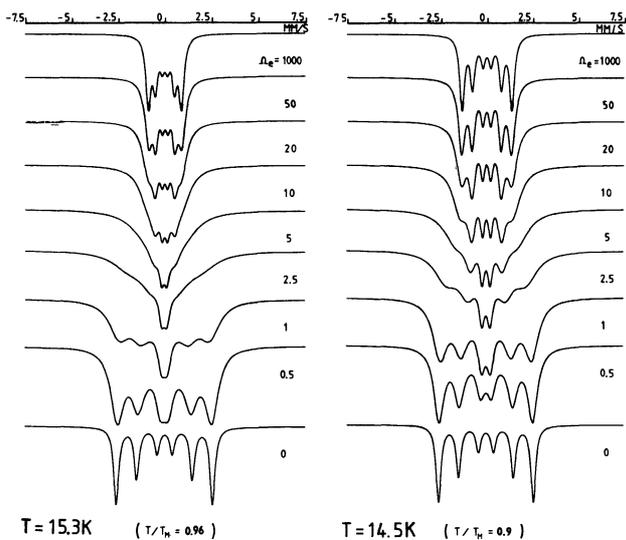


Fig. 10. — Examples of some calculated spectra in the Dekker model (relaxation in the presence of a non zero molecular field) assuming $S = 1/2$, $T_M = 16$ K, $\Gamma = 0.159$ mm/s, for 15.3 K ($T/T_M = 0.96$) and 14.5 K ($T/T_M = 0.9$). The different values of Ω_c are in mm/s.

5. Summary and comparison with other techniques

Let us first summarize the Mössbauer results. According to the present study, in Rh 5 % Fe it is possible to define a Mössbauer characteristic temperature $T_M = 16$ K, which is higher than the temperature of the cusp of the susceptibility : $T_g = 12.5$ K. Below T_M , the high temperature Mössbauer single line broadens very rapidly and a visible Zeeman structure begins to develop at 14 K. When T decreases further the Zeeman splitting steadily increases.

First, assuming that T_g represents the « true » transition temperature one may try to interpret the excess linewidth $2\Delta\Gamma$ down to 15 K in terms of dynamic critical effects with :

$$\Delta\Gamma \sim [(T - T_g)/T_g]^{-w} . \quad (18)$$

This leads to $w \sim 4.5$. However this picture breaks down below 15 K where the lineshape deviates from a simple Lorentzian line and cannot be explained by relaxation theories assuming zero or non zero molecular field.

Second, within a static description, it is possible to assume that below T_M a molecular field develops successively on the various sites and that the last paramagnetic spins disappear at 14 K (where $P(H_n = 0)$ vanishes). The average hyperfine field $\langle H_n \rangle$ deduced from the spectra, approximately follows a Brillouin curve corresponding to a « Mössbauer transition temperature » T_M , and the broadening of the Zeeman lines, which increases from the inner lines to the outer lines, can be attributed to a hyperfine field distribution around $\langle H_n \rangle$, itself related to the molecular field distribution produced by position disorder. This seems to imply that at the Mössbauer time scale ($\sim 10^{-8}$ s) some kind of instantaneous thermal equilibrium exists (or begins to exist) below T_M .

A third possibility is that around 15 K there is a crossing over from a « critical » behaviour to a « quasi static » behaviour characterized by a slowly modulated thermal quasi equilibrium, the characteristic time of the modulation going to infinity when $T \rightarrow T_g$.

Let us now compare with other compounds or techniques.

5.1 CsNiFeF₆. — In this insulating compound, Pappa *et al.* [15] have observed three different characteristic temperatures :

- $T = 4.75$ K for the linear susceptibility.
- $T = 5.37$ K for the non-linear susceptibility, assumed to correspond to the spin glass transition.
- $T = 6.1$ K for the appearance of a hyperfine field. This value also corresponds to the vanishing of the imaginary part $\chi''(\omega)$ of the linear susceptibility (at frequencies varying from 19 Hz to 50 kHz). Here too the thermal variation of the hyperfine field is below the Brillouin function relative to $S = 5/2$ (Fe^{3+} ions) and to $T_M = 6.1$ K (see Fig. 10 of [15]).

If there were no experimental problems, it would obviously be very interesting to have susceptibility data (especially for χ'') up to the Mössbauer frequency $\nu \sim 40$ MHz, in order to compare characteristic temperatures of different techniques at the same frequency.

5.2 MUONS. — The measuring time of this technique is of the order 10^{-7} - 10^{-6} s, much longer than the Mössbauer one, 10^{-9} - 10^{-8} s. There has been some evolution in the experimental results (better accuracy) and in their interpretation (dynamic \rightarrow static).

Uemura *et al.* [16] now admit that above T_g the muon sees a purely fluctuating field (a_d in their Eq. (17)), while below T_g it is submitted both to a static field ($a_s \propto \sqrt{Q}$, with Q the Edwards Anderson parameter) and to a fluctuating field ($a_d \propto \sqrt{1-Q}$). The existence of a_s below T_g is supported by decoupling experiments in an applied field ([16] : Fig. 9, [17], [18]). The temperature at which a_s vanishes is very near T_g ([16] Figs. 10 and 11). Above T_g Uemura *et al.* find that the correlation time τ_c of the electronic spins surrounding the muon obeys approximately a law :

$$\tau_c = \tau_0 \left(\frac{T}{T - T_g} \right)^w \quad (19)$$

with $w \sim 2.6$ - 2.9 for Cu Mn 3% and 5%. However deviations to this law occur below $1.1 T_g$. These results are compatible with the present Mössbauer study on Rh Fe.

Emmerich *et al.* [19] do not try to interpret their data with the formulas of Uemura *et al.* [16]. Instead, they assume that a fraction A of the sample relaxes so rapidly that its longitudinal relaxation function $G_1(t) = 1$ and that another fraction $(1-A)$ relaxes fairly rapidly so that its $G_2(t) = \exp(-\sqrt{\lambda}t)$ (this last expression is the one used by Uemura *et al.* to interpret their experiments above T_g). With this model Emmerich *et al.* find that the fraction A of spins with very fast relaxation ($\tau_c < 10^{-9}$ s) only vanishes at $0.8 T_g$, while they are not able to get precise information on the relaxation of the other part, as various expressions for $G_2(t)$ (Eqs. (2a), (4), (5) of Ref. [19]) give equally good fits. Emmerich *et al.* deduce from their results that the situation around T_g is inhomogeneous and characterized by the progressive development of magnetic correlated clusters. The comparison of these results with ours is not straightforward.

5.3 NMR EXPERIMENTS IN SPIN GLASSES. — Very short nuclear relaxation time T_2 have been observed at very low temperatures by Alloul and coworkers [20] in the spin glass Cu Mn. In the 4.7% Mn ($T_g = 27$ K) alloy : $T_2 T \sim 3 \mu\text{s K}$ for the ^{55}Mn nucleus ($\nu = 233$ MHz) [21], $T_2 T \sim 15 \mu\text{s K}$ for the first ^{63}Cu neighbour of the Mn ($\nu = 55$ MHz) and $T_2 T \sim 100 \mu\text{s K}$ for the fourth ^{63}Cu neighbour ($\nu = 13$ MHz) [20]. These values are much shorter than those observed in conventional ferro and antiferromagnets in which, at helium temperature, $T_2 \geq 10$ ms [22-24].

This shortness of T_2 , as well as the fact (checked on Cu neighbours) that $T_2 T \propto T_g$, seems to suggest some peculiar features of the dynamics in spin glasses (existence of very low energy magnetic excitations ?). On the other hand the « Korringa like » character of the relaxation, $T_2 T = \text{const.}$, might imply that the conduction electrons could be involved in the relaxation mechanism (perhaps as in the Weger [22] process ?).

Here we will only note that with the data relative to the first Cu neighbour in Cu 4.7% Mn (whose NMR frequency : $\nu = 55$ MHz, is comparable to the ^{57}Fe Mössbauer frequency) we get a line broadening which is of the order :

$$\Delta\nu = \frac{1}{\pi T_2} = 21.2 \text{ (kHz)} \times T \text{ (K)}.$$

It follows that even at $T = 10$ K, $\Delta\nu = 1/5$ MHz, which is much smaller than the natural Mössbauer linewidth (2.2 MHz) and all the more than the very large linewidths observed in Rh Fe or Au Fe Mössbauer spectra (in so far as these spin glasses can be compared to Cu Mn). This corroborates our interpretation of low temperature Mössbauer spectra in terms of static inhomogeneous broadening.

Note that at 0 K where $|S_i| = S$ whatever the exchange field, the residual broadening of M.E. spectra could arise from a distribution of transferred hyperfine fields.

6. Conclusion.

In Rh Fe there exist two different characteristic temperatures, the cusp temperature $T_g \simeq 12.5$ K and a Mössbauer temperature $T_M \simeq 16$ K. Below T_M there is a rapid increase of the Mössbauer linewidth and a hyperfine structure begins to develop at 14 K, i.e. above T_g .

A first interpretation consists in assuming that the spectra in the whole range 0-16 K are static Zeeman spectra with an inhomogeneous broadening around an average hyperfine field $\langle H_n \rangle$ ($\propto |\langle \overline{S_i} \rangle|$ averaged over i) which follows approximately a Brillouin's law. According to the hyperfine field distribution $P(H_n)$ obtained by fitting the spectra, hyperfine fields begin to appear at $T = 16$ K, but $P(H_n = 0)$ vanishes only at 14 K; this implies that between 16 K and 14 K only a fraction of the electronic spins are magnetized, this fraction reaching one at 14 K.

A second interpretation consists in noticing that since a dynamic broadening exists above T_M , it should still persist immediately below T_M and perhaps down to T_g . However a tentative explanation of the Mössbauer linewidth in terms of purely dynamic critical effects is only possible down to 15 K; data obtained between 18 K and 15 K then lead to an

average correlation time $\tau_c \propto \left(\frac{T - T_g}{T_g}\right)^{-w}$ with $w \sim 4.5$. Below 15 K, models assuming relaxation of the electronic spin around a zero average value are no longer satisfying; this could be due to a cross over to a static behaviour characterized by the appearance of non zero time average values $\langle S_i \rangle$ of the electronic spins, at least at the Mössbauer time scale.

It is not possible to choose between these two interpretations. Note however that the ambiguous region

only covers 1 K : the interval 15 K-16 K. On the other hand the nature : static or quasi static, of the non zero average values $\langle S_i \rangle$ observed above T_g remains to be elucidated.

Acknowledgments.

We are indebted to R. Rammal for enlightening suggestions, and to I. A. Campbell for useful discussions. We are grateful to J. L. Tholence, C. Jeandey and J. L. Oddou for the susceptibility measurements.

References

- [1] MEYER, C., HARTMANN-BOUFRON, F., GROS Y., CAMPBELL, I. A., *J. Magn. Magn. Mat.* **46** (1985) 254.
- [2] WINDOW, B., LONGWORTH, G., JOHNSON, C. E., *J. Phys. C Solid State Phys.* **3** (1970) 2156.
- [3] BRADFORD, E., MARSHALL W., *Proc. Phys. Soc.* **87** (1966) 831.
- [4] KOBEISSI, M. A., SUTER, R., GOTTLIEB, A. M., HOHENEMSER, C., *Phys. Rev.* **B 11** (1975) 2455.
- [5] KOBEISSI, M. A., HOHENEMSER, C., *Hyp. Int.* **4** (1978) 480.
- [6] HOHENEMSER, C., LEE CHOW, SUTER, R. M., *Phys. Rev.* **B 26** (1982) 5056.
- [7] KELLER, H., SAVIC, I. M., *Phys. Rev.* **B 28** (1983) 2638.
- [8] SLIVKA, J., KELLER, H., KUNDIG, W., WANKLYN, B. M., *Phys. Rev.* **B 30** (1984) 3649.
- [9] OGIELSKI, A. T., *Phys. Rev.* **B 32** (1985) 7384.
- [10] FAHNLE, M., SOULETIE, J., *Phys. Rev.* **B 32** (1985) 3328.
- [11] SOULETIE, J., THOLENCE, J. L., *Phys. Rev.* **B 32** (1985) 516.
- [12] DATTAGUPTA, S., BLUME M., *Phys. Rev.* **B 10** (1974) 4540.
- [13] DATTAGUPTA, S., *Hyperf. Int.* **11** (1981) 77 ; Eq. (4.36).
- [14] DEKKER, A. J., *Hyperfine Interactions*, ed. A. J. Freeman and R. B. Frankel (Academic Press, N. Y.) 1967, p. 679. In connection with this, see remark inserted in reference [21] of our previous paper (Ref. [1] of the present work).
- [15] PAPPA, C., HAMMAN, J., JEHANNO, G., JACOBONI, C., *J. Phys. C Solid State Phys.* **18** (1985) 2817.
- [16] UEMURA, Y. J., YAMAZAKI, T., HARSHMAN, D. R., SENBA, M., ANSALDO, E. J., *Phys. Rev.* **B 31** (1985) 546.
- [17] UEMURA, Y. J., NISHIYAMA, K., YAMAZAKI, T., NAKAI, R., *Solid State Comm.* **39** (1981) 461.
- [18] UEMURA, Y. J., YAMAZAKI, T., *Physica* **109 & 110B** (1982) 1915.
- [19] EMMERICH, K., LIPPELT, E., NEUHAUS, R., PINKVOS, H., SCHWINK, Ch., GYGAX, F. N., HINTERMANN, A., SCHENCK, A., STUDER, W., VAN DER WAL, A. J., *Phys. Rev.* **B 31** (1985) 7226.
- [20] ALLOUL, H., MURAYAMA, S., CHAPPELLIER, M., *J. Magn. Magn. Mat.* **31-34** (1983) 1353.
- [21] MENDELS, P., Third cycle thesis, Orsay 1984 (unpublished).
- [22] WEGER, M., *Phys. Rev.* **128** (1962) 1505.
- [23] ROBERT, C., Thesis, University of Paris 1962 (unpublished).
- [24] RADO, G. T., SUHL, H., *Magnetism II A* (Academic Press) 1965. See papers by JACCARINO, V. (*NMR in Antiferromagnets*) and by PORTIS, A. M., LINDQUIST, R. H. (*NMR in Ferromagnetic Materials*).