RECENT EXPERIMENTAL STUDIES OF DYNAMICS OF SPIN GLASSES

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Résumé.- On présente une revue des résultats expérimentaux sur la dynamique de spin des verres de spin en alliages binaires, obtenus par des techniques esr, nmr, precession de muons, effet Möss-bauer et de diffusion de neutrons. Les résultats peuvent être décrits de façon consistante en ter-mes de spectre de temps de relaxation evoluant continuellement avec la température et s'étendant de temps relativements courts ($\tau \sim 10^{-13}$s) à des temps très longs ($\tau\to\infty$) à des températures au voisignage du point de congélation.

Abstract.- A review of the experimental results of the spin dynamics of binary spin glass alloys obtained with the help of esr, nmr, muon precession. Mössbauer effect and neutron scattering techniques is given. The results may be consitently described in terms of a spectrum of relaxation times evolving continuously with temperature and extending from relatively short ($\tau \sim 10^{-13}$s) to very long times ($\tau\to\infty$) at temperatures around and below the freezing temperature.

1. INTRODUCTION.- Experimental studies of dynamics of spin glasses began about two decades ago with the first esr and ^{63}Cu nmr measurements on dilute Cu-Mn alloys by Owen et al. /1/ followed by further investigations of the more concentrated alloys where the effects of freezing of spins on the dynamical properties were studied through the temperature dependence of the esr signal /2-4/. Initially, host nmr measurements were aimed at the magnetic proper-ties associated with the Kondo effect in very di-lute alloys /5/ but more recently the nmr technique has also been applied to study the process of free-sing of spins in more concentrated alloys /6-9/. A closely similar probe is the muon precession technique /10/ which has also been sucessfully used in investigations of spin glasses /11/. A certain difference of feature from the nmr and because of the unexplained weaker influence on the muon's free precession decay rate due to rapid solute spin fluctuations at high temperatures, the method becomes effectively comparable to the Mössbauer effect technique which has been widely used to study the onset of magnetic "ordering" in spin glass alloys /12-15/. Neutron scattering techniques provide the most direct probes of spin dynamics and have been increasingly used in recent years to study dilute alloys as well as the more concentra-ted spin glasses /16-22/.

In the following we review the results of experi-mental studies of dynamics of spin glass alloys. We begin by considering the dynamics in the dilute limit and at high temperatures and then proceed to examine its temperature dependence across the spin glass freezing temperature, in order to gain some insight into the process of freezing of spins espe-cially in relation to the question of the sharp phase transition predicted theoretically /23-25/. Finally we review the spin dynamics below the freezing temperature and examine the low temperature excitations of the spin system.

2. DILUTE LIMIT/HIGH TEMPERATURE SPIN DYNAMICS.- The first esr measurements on dilute Cu-Mn alloys were carried out with the aim to study the s-d in-teraction between the conduction electrons and the localised Mn spins /1/. Observation of well defined resonance signal with a relatively small g-shift and a linear Korringa law temperature dependence of the line-width yield a coupling constant which is several orders of mangitude smaller than the J_{s-d} obtained independently from the ^{63}Cu nmr line-shape and width in the same system /26/. It was soon re-cognised however /27/ that because of the strong coupling of the localised spins with the conduction electrons and the relatively long relaxation time constant of the latter with the lattice, the esr is bottlenecked so that the full effect of the s-d interaction on the g-shift and the line-width and hence the spin dynamics cannot be observed. The longitudinal and the transverse relaxation times T_1 and T_2 of the host Cu-nuclei are, however, direc-tly influenced by the spin fluctuations. From the measurements of T_1 in dilute Cu-Mn alloys Alloul et

al. /5/ deduce Mn spin fluctuation time constant of
$\tau \sim 10^{-11}/T$.

Results of a recent neutron scattering investigation
of the dynamics of Cu-Mn alloys containing between
1 and 7 at %Mn /28/ show that the spectral function
in the dilute limit is a simple Lorentzian with a
relatively narrow width (half width $\Gamma \sim 2$ meV at
300 K) resulting principally from the Korringa re-
laxation of the spins. The effect of solute-solute
interaction with increasing Mn concentration is to
produce progressively larger departures from this
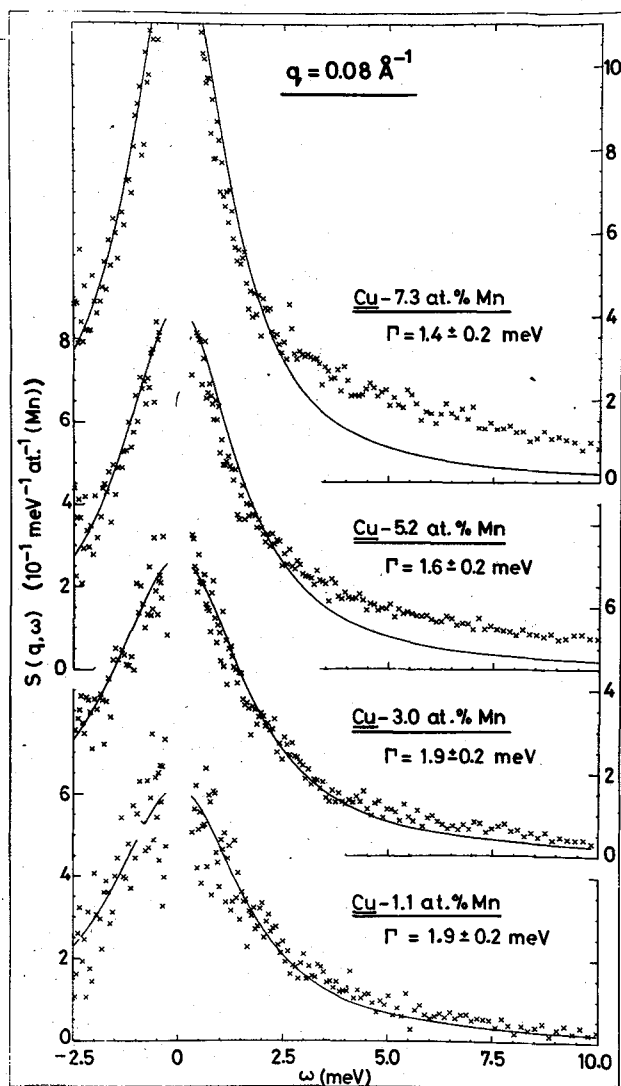simple form which are particularly noticeable at
low q's, figure 1.

Fig. 1 : $S(q,\omega)$ vs ω for $q = 0.08$ Å^{-1} at 300 K for
several Cu-Mn alloys. The continuous curves repre-
sent the best fits to the data using the Lorentzian
form of the spectral function. The data points for
elastic scattering are not shown in the diagram.
Results from ref. /28/.

The results show that the spectral function becomes
narrower in the central region with a shift of some
of the intensity into the high energy wings. At
higher q's we observe increasing broadening of the
spectra with increasing concentration. It should be
mentioned, however, that in the analysis of the data
the q-range has been effectively extended to lower
values and the energy range to much higher values
by a new method of analysis of the TOF data. In our
earlier measurements on a Cu-8 at % Mn /21/ and a
Au-10 at % Fe alloys /22/ a constant -q interpola-
tion technique was employed which severely limited
the lowest q-value and the energy range covered by
a constant -q cut through the q-ω plane, hence the
deviations from the simple Lorentzian form of the
spectral function at high energy transfers and low
q's could not be observed.

Lack of theoretical models prevent a quan-
titative analysis of the spectral shapes, although
some of the early measurements on Heisenberg para-
magnets were analysed using semi-empirical forms
of the spectral functions /29/. In the case of
spin glass alloys, the presence of two channels
of relaxation - the solute interactions and the
Korringa couplings - complicated the situation even
further. It should be mentioned, however, that the
measurements on Au-Fe alloys by Loewenhaupt et al
/18/ and by Scheuer et al /19, 20/ have been inter-
preted simply in terms of a sum of two Lorentzian
spectral functions of different widths which they
attribute to Korringa relaxation of single isolated
spins (a broad Lorentzian of half-width $\Gamma \sim 10$ meV
at 300 K) and of magnetic clusters (a narrower
Lorentzian with $\Gamma \sim 2$ meV at 300 K). The latter have
apparently been identified with statistical near-
neighbour clusters such as pairs, triplets and
higher agglomerates. Their analysis implies that
the relative intensity of the broad, high energy
part of the spectral function (associated with sin-
gle isolated spins- should decrease with increasing
solute concentration whereas the results for Cu -
Mn alloys show that the intensity in the high energy
wings increases with increasing solute concentration
and that the spectral function in the dilute limit
associated with the Korringa relaxation of isolated
spins is a simple Lorentzian of relatively narrow
half-width ($\Gamma \sim 2$ meV at 300 K for Cu - Mn alloys).
3. THE TEMPERATURE DEPENDENCE OF THE SPIN DYNAMICS.-
(Freezing of Spins). 3.1. Neutron Scattering Measu-
rements.- An important observation from the results
of the last section is that the paramagnetic

spectral function has contributions from two chan-
nels of relaxation, namely the Korringa mechanism
and the solute-solute interactions. The temperature
dependence of the dynamics is therefore expected
to be rather complex and reflect the temperature
dependence of both these processes.

The results of a recent detailed study on a Cu-3 at
% Mn alloy /29/ are shown in figure 2 where the
temperature dependence of the half-widths of the
spectra are shown for q = 1.0 Å^{-1} and for the limit
q→0.

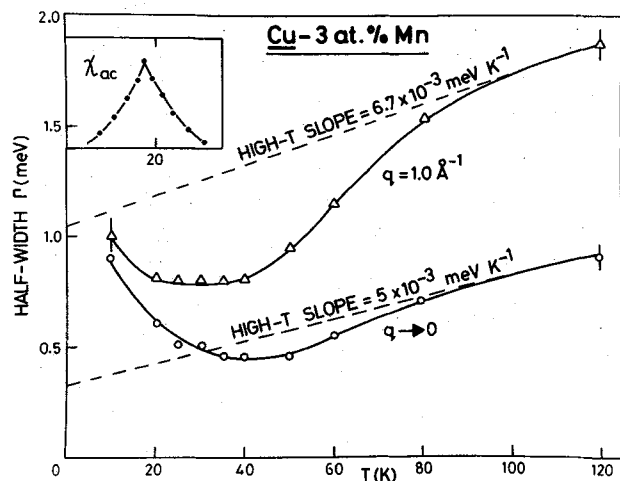
Fig. 2 : The half-widths at half-heights for the
broad part of the quasielastic spectra for a Cu-3
at % Mn sample for q = 1.0 Å^{-1} and in the limit
q→0. The inset shows the ac susceptibility χ on the
same temperature scale. The dashed lines show the
high temperature slopes. Results form ref. /30/.

The dashed line gives the high temperature extrapo-
lations for the two cases. In the same diagram, on
the same temperature scale the a.c susceptibility
χ is also shown, with a peak at 17 K. The results
indicate that the spectral width begins to narrow
at relatively high temperatures, reaches a certain
finite value and then increases again at lower
temperatures thus going through a shallow minimum
at a temperature well above the "freezing tempera-
ture" of 17 K. Similar results have been obtained
for a Cu-8 at % Mn and a Au-10 at % Fe alloys
/21, 22/. However, the results of measurements on a
series of AuFe alloys by Scheuer et al. /19, 20/
apparently show linear decrease to zero at T=0 of
the line-widths associated with the isolated spins
and the magnetic clusters mentioned earlier.
We recall that the neutron scattering cross-section
for a system of N spins is given by

$$\frac{d^2\sigma}{d\Omega d\omega} = N \left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{k'}{k_o} \{S_i(q,\omega) + S_e(q) \delta(\omega) \} \qquad (1)$$

where $S_i(q,\omega)$ is the dynamic structure factor repre-
senting the inelastic/quasi-elastic scattering and
$S_e(q)$ represents the elastic scattering.

In figure 3 the observed temperature variations of
the elastic cross-section, the integrated quai-
elastic cross-section as well as their sum are shown
for a Cu- 8 at % Mn sample /21/. The elastic scat-
tering is approximatly constant at high temperatures
where it is due to the nuclear incoherent cross-
section of the sample, but then begins to increase
with decreasing temperature around about the same
temperature where the quasi-elastic scattering starts
to decrease, these variations being continuous
across the spin glass freezing temperature determi-
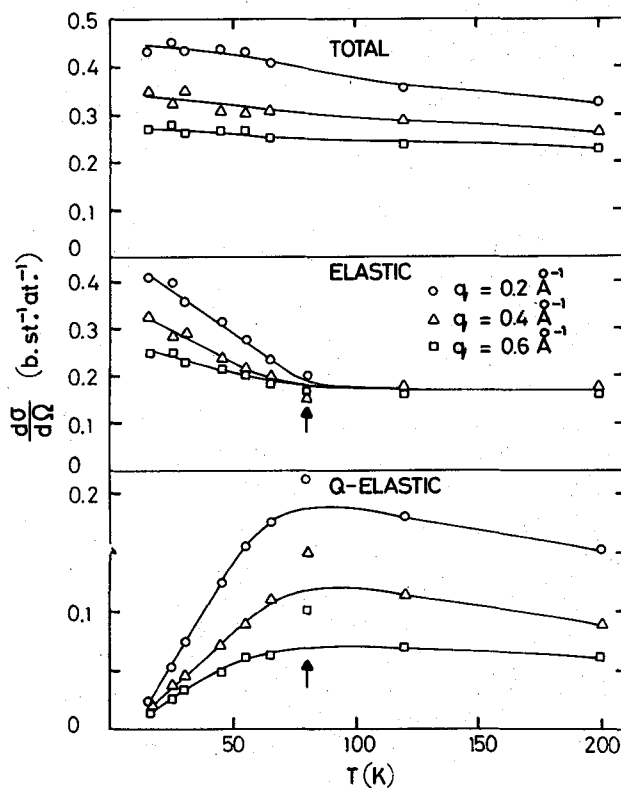ned from the ac susceptibility measurements.

Fig. 3 : The temperature dependence of the elastic
cross-section (energy resolution 230 μeV), the in-
tegrated quasi-elastic cross-section and their sum,
measured for a Cu-8 at % Mn sample. The dashed ver-
tical line marks the temperature of the maximum in
the ac susceptibility. Results from ref. /21/.

It should be remembered, however, that the separa-
tion of the elastic scattering from the inelastic/
quasi-elastic scattering contributions is limited
by the finite instrumental energy resolution. As
seen below this has special relevance for spin
glasses where with the evolution of correlations in

the spin system with decreasing temperature a whole spectrum of relaxation times τ develops. It therefore appears meaningful to express $S_i(q,\omega)$ as

$$S_i(q,\omega) = \sum_\alpha P(T,\Gamma_\alpha) \; \frac{1}{\pi} \; \frac{\Gamma_\alpha}{\Gamma_\alpha^2 + \omega^2} \qquad (2)$$

where $P(T, \Gamma_\alpha)$ is the probability distribution at temperature T of the relaxation times $\tau_\alpha (= \hbar/\Gamma_\alpha)$. Hence if Γ_α is small compared to the elastic energy resolution ΔE of a spectrometer the scattering would be indistinguishable from that due to a truly elastic process. It is obvious therefore that if a wide spectrum of relaxation times exists in spin glasses the result of measurement of the elastic scattering cross-section will depend strongly on the instrumental energy resolution i.e. the measurement time constant. This is clearly borne out by the results of measurements of the elastic scattering cross-section for the Cu - 8 at % Mn alloy /31/ employing energy resolutions of 1.5, 25 and 230 µeV, figure 4.
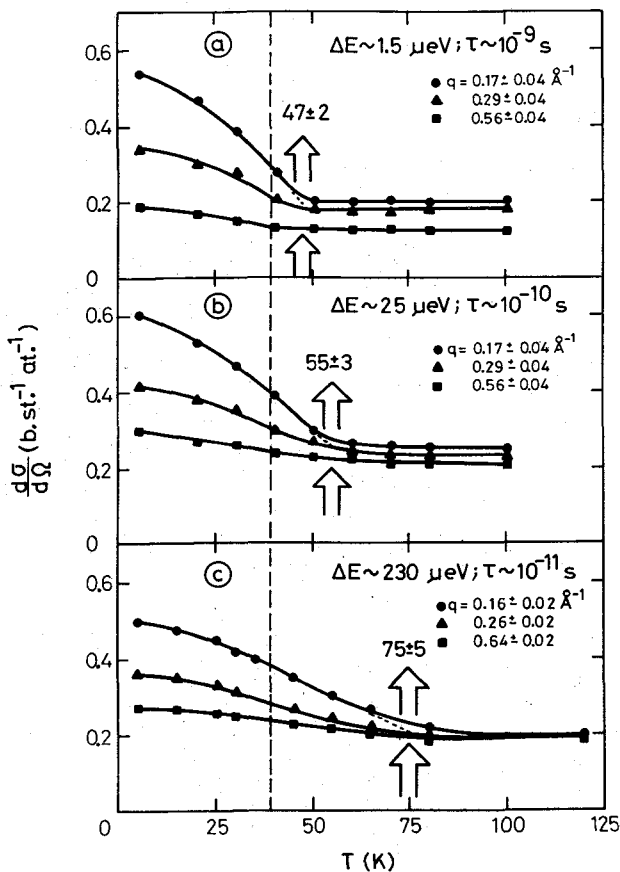

Fig. 4 : The temperature variation of the measured elastic cross-section $\frac{d\sigma}{d\Omega}$ using energy resolutions of a) 1.5 µeV, b) 25 µeV and c) 230 µeV, for several q-values. The arrows indicate the temperature where marked increase in the cross-section with decreasing temperature begin. Results from ref. /31/.

We see that the temperatures where the marked increase of the elastic scattering cross sections occur depend strongly on the energy resolutions employed and hence on the measurement time constant $\tau (= \hbar/\Delta E)$. It should be noted that these temperatures vary systematically with τ and are all higher than the temperature of the maximum in the ac susceptibility shown by the dashed vertical line in the figure.

The elastic magnetic scattering i.e. the static structure factor $S_e(q)$ has a special significance in spin glass theory, it being directly related to the Edwards-Anderson order parameter q (23), for

$$S_e(q) = \frac{2}{N} F^2(q) \sum_{ij} \left[\exp \; i\underline{q} \cdot (\underline{r}_i - \underline{r}_j)\right] <S_i> <S_j>$$

$$= 2 \; F^2(q) \sum_n \exp \left[-i\underline{q} \cdot \underline{r}_n\right] \overline{<S_{i+n}> <S_i>} \qquad (3)$$

where $\underline{r}_n = \underline{r}_j - \underline{r}_i$ and the bar represents the average over-all spins. Hence, taking the inverse Fourier transform we obtain the EA order parameter

$$q = \overline{<S_i>^2} = \frac{1}{N} \sum_q \frac{S_e(q)}{2F^2(q)} \qquad (4)$$

Although measurements of $S_e(q)$ over the whole q-range are not practicable, the results over a limited q-range can nevertheless reflect the actual behaviour of the order parameter q. As seen from the above results $S_e(q)$ is peaked in the forward direction (q=0) and for the q-range over which any temperature dependence is seen, the temperature of the marked increase is roughly the same. Hence each of the $S_e(q)$ vs T curves represents the temperature variation of the EA order parameter q_{τ_0} where the subscript τ_0 denotes the finite time constant τ_0 of the measurement. q_{τ_0} is defined as

$$q_{\tau_0} = \overline{<S_i>^2_{\tau_0}} = \frac{1}{N} \sum_{\tau_\alpha > \tau_0} <S_i>^2_{\tau_\alpha} \qquad (5)$$

where the bracket $< >_\tau$ represents the time average over the period τ.

Thus all quasi-elastic processes with time constants τ_α longer than $\tau_0 (= \hbar/\Delta E)$ contribute to the order parameter q_{τ_0}. As discussed below the Mössbauer effect which is another probe for the dynamics of spins does, in effect, measure the square root of the order parameter q_{τ_0} with a time constant $\tau_0 \sim 10^{-7}$ (for ^{57}Fe). These results also show a significantly higher temperature for the marked increase in the average nuclear hyperfine field (commonly referred to as the "ordering" temperature) compared with the temperature of the maximum in the ac susceptibility χ /14, 15/.

3.2. ESR measurements.- The observation of the esr of Mn solute spins in Cu-Mn alloys owes to the bottleneck effect in the combined resonance of the

local moment conduction electron spin systems due to the relatively long relaxation time constant of the latter to the lattice /27/. The combined resonance of the coupled systems nevertheless reflects the dynamics of the Mn spins which bear strong resemblence to the spin dynamics observed in neutron scattering measurements. Following Owen et al. /12/ the esr spin dynamics in concentrated Cu-Mn alloys were studied in detail by Griffiths /3/ and later by Okuda and Date /4/. In close similarity with the temperature dependence of the quasi-elastic spectral width in neutron scattering the esr line-width decreases with decreasing temperature and then goes through a shallow minimum at a temperature well above the spin glass freezing temperature, figure 5.
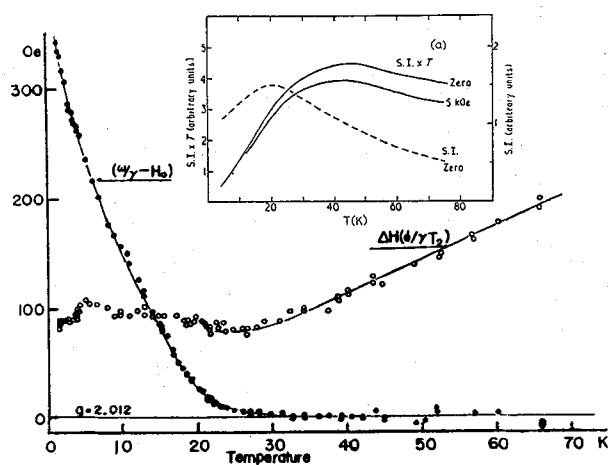

Fig. 5 : The esr line-width ΔH and the line-shift ($\omega/\gamma - H_o$) as a function of temperature for a Cu-2 at % Mn alloy. The temperature of the maximum in the static susceptibility for this concentration of Mn is expected to be around 13 - 14 K. Results from ref. /4/. The inset shows the esr intensity - proportional to the susceptibility χ - and the quantity S.IxT (signal intensity x temperature) plotted as a function of temperature for a Cu-4 at % Mn alloy. The results obtained are slightly different if a steady field of 5 kOe is applied when cooling the sample to low temperatures. Results from ref. /3/.

Another important result from the measurements is that the esr line position begins to shift to lower fields around about the same temperature as that of the minimum in the line-width. This temperature dependent field shift was first observed by Owen et al. /2/ who interpreted it in terms of an antiferromagnetic resonance. Recently, Malozemoff and Jamet /32/ have interpreted their measurements of the esr field-shifts in thin films of amorphous $Gd_{0.37}Al_{0.67}$ samples in terms of local demagnetizing fields due to inhomogeneous magnetisation regions

within the system and suggested that a similar interpretation could be applied to the esr results for Cu - Mn alloys also.

We recall that the resonance intensity of the esr signal in the paramagnetic phase is proportional to the susceptibility χ. This signal intensity and the quantity 'signal intensity x temperature' in Griffiths' results /3/ are shown in the inset in figure 5. The latter quantity, it should be pointed out, corresponds to the integral of the broad quasi-elastic scattering intensity in neutron scattering measurements. The similarity between the temperature variation of the two, particularly their continuous diminuation across the freezing temperature is therefore not surprising. Assuming that the susceptibility obeys approximately the Curie-law $\chi = C/T$, the broad quasi-elastic spectrum in neutron scattering measurements is then seen to be proportional to the number of spins with relatively rapid relaxation times. Similarly in the esr the quantity "resonance intensity x temperature" is proportional to the number of Mn spins which can "respond" with the resonance frequency. Both results therefore indicate a progressive and continuous diminution across the spin glass freezing temperature of the fraction of Mn spins with relatively rapid fluctuation times. Also, the integrated resonance intensity which should strictly be proportional to the static susceptibility $\chi(\omega=0)$ in effect measures an effective susceptibility at a frequency ω since the resonance signal only includes Mn spins which can "resonate " with the driving microwave field. Hence, as with the neutron scattering results /21/ the temperature of the maximum in the resonance signal should occur at a higher temperature than the maximum in the susceptibility measured with a slower frequency of the ac technique allowing, of course, for any field variation of the temperature of the maximum. Although there are indications in Griffiths' results /3/ that this may be so, the suggestion needs further verification.

3.3. NMR measurements.Host [63]Cu nmr measurements on Cu-Mn spin glass alloys containing between 1000 and 4000 ppm Mn were first reported by McLaughlin and Alloul /6/. The results show continuous diminution with decreasing temperature across the spin glass transition temperature of the longitudinal and the transverse relaxation times and of the resonance intensity. In these dilute samples the authors found no evidence of any static line-broadening effects due to frozen-in configurations of Mn spins,

whereas in subsequent measurements by Levitt and Walstedt /7/ on a more concentrated alloy containing 1 at % Mn and on alloys containing 0.6 and 1 at % Mn /8/ the line-width ΔH was found to increase rather slowly with field and to have a large finite value ΔH_o as $H \to 0$, giving definite evidence of static fields due to "frozen-in" Mn spins. In these latter measurements the resonance intensity was found to decrease with decreasing temperature from a temperature well above T_F, as in the more dilute alloys, and to go through a minimum at a temperature around $T_F/2$ increasing again at low temperatures, figure 6.
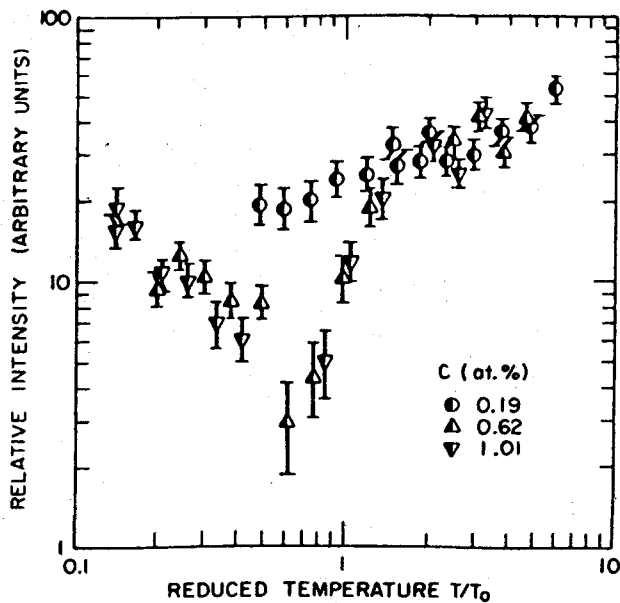

Fig. 6 : The temperature dependence of the nmr spin echo intensity for three Cu-Mn alloys of Mn concentrations 0.19, 0.62 and 1.01 at % Mn. The temperature scale is relative to T_o the spin glass freezing temperature observed in the static susceptibility measurements. Results from ref. /8/.

The transverse relaxation time T_1 was similarly found to go through a (shallower) minimum at a temperature below T_F, and increase by a small amount at low temperatures, figure 7.
The interpretation of solute spin dynamics from the host nmr measurements is more complicated than from the esr measurements for example, since the spin dynamics are observed indirectly through their influence on the host Cu-nuclei which experience a wide distribution of RKKY hyperfine fields. Presence of an external field further complicates the interpretation. For example, in recent measurements of the longitudinal relaxation times T_1 on extremly dilute alloys containing between 10 and 40 ppm Mn /9/ the adopted analysis suggests that the impurity

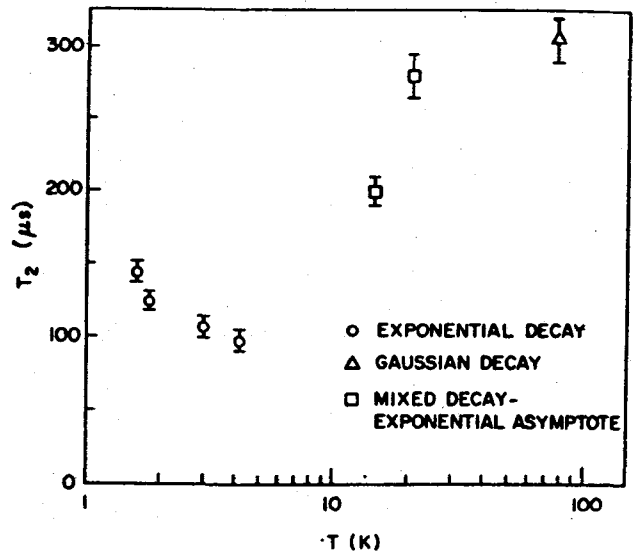spin correlation time decreases with increasing applied field.

Fig. 7 : The temperature dependence of the transverse relaxation time T_2 for a Cu- 1 at % Mn alloy measured at the resonance frequency of 25.5 Mhz. Results from ref. /7/.

Analysis of these results apparently show well defined temperatures for the onset of freezing of solute spins. These temperatures, further-more, are the same as those obtained from susceptibility measurements /33/. At the same time, however, the results suggest that the mean impurity spin correlation time increases continuously with decreasing temperature across the freezing temperature reaching a value of about $\sim 10^{-4}$s at a temperature roughly a factor 20 lower than the freezing temperature T_F.
The influence on the transverse relaxation time of a nucleus from a fluctuating source of hyperfine field h_i, such as the solute spin, may be divided into three regions depending on the relative time constants of the precession period $(\gamma h_i)^{-1}$ of the nucleus and the correlation time τ_c of the source (i.e. the spin). Thus,
a) when $(\gamma h_i) \ll 1/\tau_c$, the contribution to the relaxation time is given by $T_2^{-1} \sim (\gamma h_i)^2 \tau_c$.
b) However, when the correlation time τ_c of the impurity spin is comparable to $(\gamma h_i)^{-1}$ the transverse relaxation time is determined directly by τ_c, i.e. $T_2 \sim \tau_c$.
c) Finally, if the correlation time is very long compared with $(\gamma h_i)^{-1}$ i.e. $(\gamma h_i) \gg 1/\tau_c$, the hyperfine field only leads to static line broadening effects, the contribution to T_2 is then mainly from gradients in the hyperfine field.

Also in the interpretation of the nmr data it is recalled that the spectrometer dead time due to rf pulse overload is fairly long, $\sim$ 50 µs which puts a limit to the shortest relaxation time T_2 which can be measured in the experiments.

We can now see how the solute spin dynamics observed in neutron scattering measurements are consistent with the nmr data. At high temperatures, the Mn spin correlation time τ_c is short $\sim 10^{-13}$s, and the average RKKY field h_i at the Cu-nucleus ($\sim$ 600 Oe for 1 at % Mn) is such that $(\gamma h_i) \ll 1/\tau_i$ so that the contribution to T_2 is $T_2^{-1} \sim (\gamma h_i)^2 \tau_c$. Hence with decreasing temperature as the spin dynamics slow down i.e. as τ_c increases one observes a reduction of T_2. The neutron scattering results indicate a distribution of spin correlation times τ_c which together with a distribution of the hyperfine fields h_i suggests that for some of the nuclei T_2 would become much shorter than for others. When these times get shorter than the spectrometer dead time the corresponding nuclei are lost from the signal. As the temperature decreases and approaches T_F the spin dynamics pass rapidly from the fast fluctuation regime $(\gamma h_i) \ll 1/\tau_c$ to the slow fluctuation regime $(\gamma h_i) \sim 1/\tau_c$ leading not only to the reduction of the average measured T_2 but also of the intensity. Finally as the correlation times of the impurity spins begin to get very long $(\gamma h_i) \gg 1/\tau_c$, the hyperfine fields lead only to static line broadening effects, their contribution to T_2 of the observed nuclei being mainly through any resultant field gradients.

Thus at low temperatures as T_2's become longer again these nuclei return to the signal and we see not only an increase of the intensity but also of the average measured T_2. There is, however, a net loss of signal intensity at low temperatures (compared with higher temperatures) through nuclei which experience very large static hyperfine fields and are thus lost into the very large wings of the broadened signal, and through those that still experience rapid Mn spin fluctuations and have T_2 below the spectrometer dead time. Thus the nmr results are consistent with a wide spectrum of relaxation times as observed in neutron scattering measurements also.

3.4. **Muon Precession Technique.** - In this technique of measurement the muons are implanted into the sample to be investigated where they occupy interstitial sites, their spins precessing about an applied polarising field. The muon half-life is $\sim 2 \times 10^{-6}$s. It decays through positron (β^+) emission preferentially along its spin direction, so that oscillations in β^+-detection

rate are observed in a detector positioned at an angle to the applied field. This technique of probing the dynamics of solute spins is similar to the host nmr measurements in that in both cases the spin dynamics influence the transverse relaxation times T_2 of the probes. However, the dominant hyperfine field at the muon site is through the dipolar coupling with the solute spins which is an order of magnitude smaller than the RKKY field at the ^{63}Cu nuclei. The gyromagnetic ratio γ_μ of the muon is, however, an order of mangitude smaller than that for the Cu-nuclei, (I thank D. McLauglin for bringing this to my attention) so that the average $(\gamma_\mu h_i)$ at the muon sites is of the same order of magnitude as that for the Cu-nuclei. However, in the fast fluctuation regime, $\tau_c \ll (\gamma_\mu h_i)^{-1}$ the influence of the solute spin dynamics on the muon free precession decay time T_2^* is much weaker than would be expected from a straight comparison with the nmr results where the spin echo decay time T_2 begins to decrease markedly from a relatively high temperature. It appears that as the temperature approaches the freezing temperature T_F the spin dynamics are slowing down to the value $\tau_c \sim (\gamma_\mu h_i)^{-1}$ when the influence on muon's depolarisation rate becomes noticeable $(T_2^* \sim \tau_c)$. As the solute spin dynamics slow down even further i.e. as $\tau_c \ll (\gamma_\mu h_i)^{-1}$ the static fields which are produced with random directions and magnitudes, also cause the same effective depolarisation of the muons. In this respect the muon depolarisation technique is different from the host nmr measurements where the static fields mainly produce inhomogeneous line broadening (with some of the nuclei lost into the wings), i.e. the transverse relaxation time T_2 of the nuclei which are observed in nmr is not directly related to the line-broadening, whereas in the muon precession method T_2^* and line broadening are related.

In figure 8 we reproduce the results of Fiory /11/ on the variation of the muon depolarisation rate Δ in a Au-0.7% Fe alloy ($\Delta = (\gamma_\mu T_2^*)^-$), as a function of the reduced temperature T/T_o, where $T_o \equiv T_F$. In the diagram the vertical line through the freezing temperature $(T/T_o = 1)$ is added for emphasis. The results indicate a relatively narrow temperature range over which the depolarising fields develop and their rapid continuous increase across the temperature T_F (the freezing temperature in the static susceptibility) and the approach to saturation as $T \to 0$ K.

The relatively weak influence of the solute spins

on the T_2 of muons in the fast fluctuation regime
$(\tau_c \ll (\gamma h_i)^{-1})$ means that except over a very small
temperature range the muon depolarisation principal-
ly occurs with the onset of static fields (i.e.
$\tau_c > (\gamma h_i)^{-1})$. Thus the method becomes comparable
to the Mössbauer effect technique where the nuclear
hyperfine field due to the solute spin is "felt"
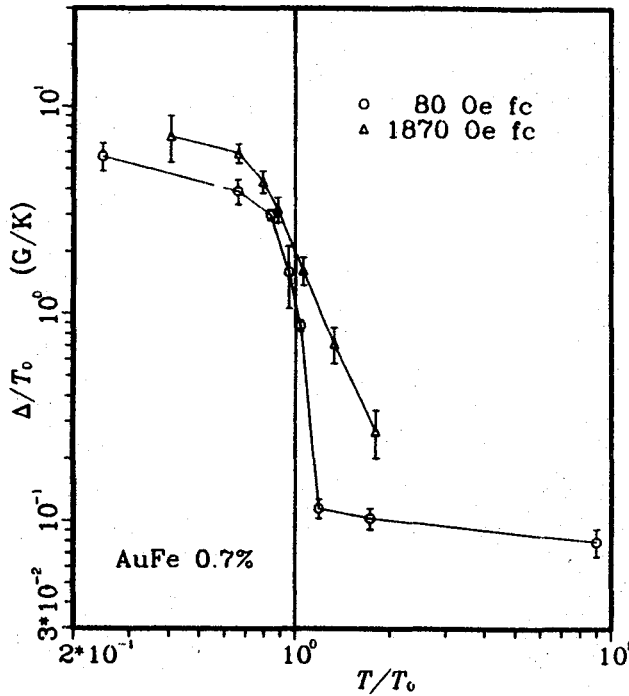only when $\tau_c \gtrsim (\gamma h_i)^{-1}$.

Fig. 8 : The muon depolarisation rate Δ for a Au-
0.7 at % Fe alloy as a function of the reduced tem-
perature T/T_0 of the maximum in the static suscepti-
bility, which is emphasised by the added vertical
line through $T/T_0 = 1$. Results from ref. /11/.

3.5. Mössbauer Effect.- As a probe of spin dynamics
the Mössbauer effect is somewhat different from the
host nmr technique in that it does not measure the
influence of solute spin fluctuations on the longi-
tudinal or the transverse relaxation times of the
probe nuclei in a resonance mode. In the Mössbauer
effect, the probe nucleus either emits or absorbs
a γ-ray in a recoiless process and thus undergoes
transition between a ground and an excited state.
The effect of a magnetic field at the nucleus is to
produce splitting of the nuclear energy levels so
that a hyperfine splitting of the energy spectrum
results. In order that a field h_i can cause obser-
vable splitting, however, it must remain stable at
least over the period τ_L of the nuclear Larmor pre-
cession $(\tau_L = (\gamma h_i)^{-1})$. The Mössbauer transition,
however, has an intrinsic line-width $\hbar/\tau_0$ so that

only splittings comparable to or larger than this
width can be observed. Thus τ_0 defines the time cons-
tant of the probe ($\sim 10^{-7}$s for ^{57}Fe). Hence, in a
spin glass at high temperatures when the spins are
fluctuating rapidly i.e. $\tau \sim 10^{-13}$s the time average
local magnetisation $\langle S_i \rangle_{\tau_0}$ over the Mössbauer tran-
sition period is zero. As the temperature is reduced
and the spins begin to slow down the $\langle S_i \rangle_{\tau_0}$ become
finite the Mössbauer spectrum begins to broaden and
finally the characteristic six line hyperfine spec-
trum is observed at low temperatures.

Figure 9 shows typical Mössbauer spectra for a Au -
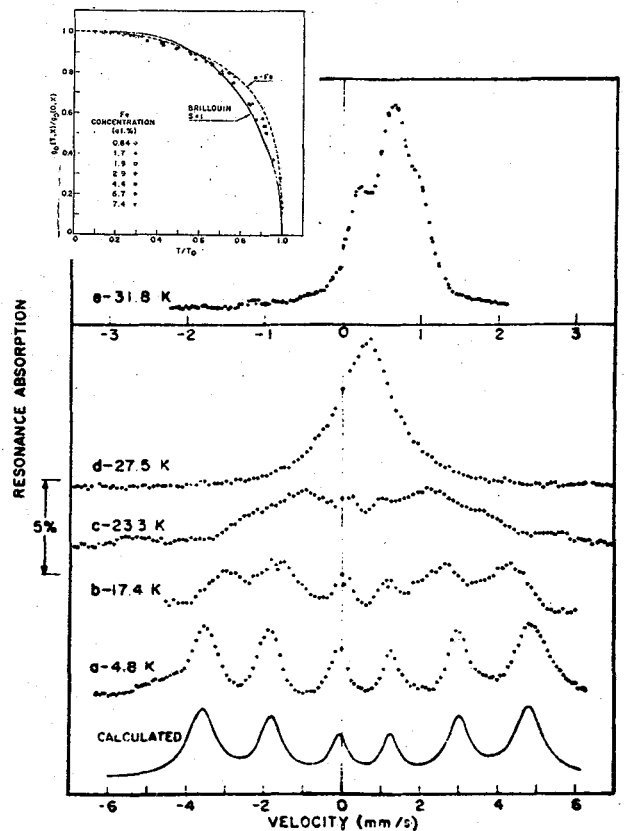6.7 at % Fe alloy observed by Violet and Borg /13/.

Fig. 9 : Mössbauer spectra for a Au- 6.7 % Fe alloy
observed at several temperatures, including the
calculated spectrum at low temperatures. The "free-
zing temperature" T_0 for this alloy obtained from
analysis of the spectra is 27.6 K which is close to
the temperature for the spectrum d. The inset shows
the temperature variation of the average hyperfine
field for a series of Au - Fe alloy plotted on the
reduced temperature scale relative to T_0 the "orde-
ring" or the "freezing temperature" obtained from
analysis of the spectra. Results from ref. /13/.

It is interesting to note that the average hyperfine
field h measured in a Mössbauer experiment is pro-
portional to the square root of the E-A order para-
meter q_{τ_0} defined by equation (5).

For,

$$h \propto \frac{1}{N} \sum_{\tau_\alpha > \tau_o} |\langle S_i \rangle_{\tau_\alpha}| = \overline{|\langle S_i \rangle_{\tau_o}|} = q_{\tau_o}^{\frac{1}{2}} \quad (7)$$

where all spins fluctuating with time constants τ_α longer than τ_o contribute to h.

The temperature variation of the average hyperfine fields for a series of Au-Fe alloys studied by Violet and Borg /13/ are shown in the inset in figure 9. The results apparently indicate well defined "ordering" temperatures T_o. It was seen in the neutron elastic scattering measurements with finite energy resolutions that the "freezing temperatures" obtained are systematically higher the shorter the time constant of measurement. Although detailed comparisions of the freezing temperatures from the Mössbauer and the static susceptibility measurements are not available for the Au-Fe system, results for the Rh-Fe /15/ and the Cu-Mn alloy systems /14/ (the latter with small amounts of Fe and Sn as probes) show systematically higher "freezing temperatures" compared with the temperatures of the maxima in static susceptibility. Also, the results presented in figure 9 give a somewhat false impression of the abruptness of the increase of the average hyperfine field at T_o. For example, Violet and Borg's analysis of the Mössbauer spectra for the Au-6.7 % Fe alloy shown in figure 9 suggest that the ordering temperature T_o is 27.6 K, spectrum d, whereas the spectrum e measured at 31.8 K still shows an appreciable broadening which can be translated into a measurable hyperfine field. Thus the measurements in reality indicate a gradual initial increase of the mean hyperfine field occuring well above the "freezing temperature", in agreement with the observations of muon precession and neutron scattering techniques.

4. SPIN DYNAMICS BELOW T_F.- In the results reviewed above we have seen that the broad quasielastic spectral intensity lying outside the elastic energy resolution of the neutron spectrometer and the quantity "resonance intensity x temperature" in the esr measurements show very similar temperature dependence and indicate the persistance of a finite, although continuously diminishing, component of rapidly fluctuating spins or spins which can respond to the microwave signal, down to the lowest temperatures of the measurements. Can these be regarded as free paramagnetic spins ? We have seen that the spectral width in the neutron spectra and the line-width in the esr measurements go through shallow minima at temperatures above the "freezing

temperatures T_F". Also , the resonance field in the esr begins to shift to lower values below about the same high temperature. Another important observation is furnished by the Mössbauer spectra on spin glasses. For example, the spectra for the Au-6.7 at % Fe alloy, figure 9, show no detectable unsplit paramagnetic component at finite temperatures some way below the "freezing temperature", c.f. spectrum c at 23.3 K, (figure 9). All these observations support the conclusion that the rapid spin dynamics at low temperatures are not due to free spins in their paramagnetic state, but may be thought of as due to spins which become correlated to certain time-average spatial directions over time scales longer than the typical measurement time constants of these probes and "oscillate" individually and collectively about these equilibrium mean positions. Thus the broad quasi-elastic spectrum observed in neutron scattering measurements below the freezing temperature must be regarded as arising from such diffusive "spin-wave" modes. The fact that the width of the spectrum increases with decreasing temperature below T_F indicates that these diffusive "spin-waves" get stiffer with decreasing temperature. It should be remembered, however, that the integrated intensity of these excitations steadily decreases with decreasing temperature, figure 3, as does the quantity "signal intensity x temperature" in the esr results, figure 6.

Recent neutron scattering measurements on Cu-Mn alloys at low temperatures by Scheuer et al. /34/ show an additional feature - a broad inelastic peak due to spin wave-like excitations around an energy transfer ω of 4.2 meV which they claim appears dispersionless in their q-range of measurement $0.5 < q < 1.5 \text{ Å}^{-1}$. Recently Ching and Huber /35/ have extended their earlier computer simulation calculations of zero temperature spin dynamics /36/ and show that this peak corresponds to the small maximum around a finite energy in the density of excitation states in their calculations.

5. CONCLUSIONS.- In the foregoing we have reviewed the spin dynamics of binary alloys observed with a range of measurement techniques. The results show the strong influence of the Korringa relaxation mechanism on the dynamics of the spins. Additionally the spin-spin exchange leads to important modifications of the dynamics which eventually result in spin glass freezing phenomena observed at low temperatures. We have seen that the results suggest a continuous evolution of correlations in the spin

system with decreasing temperature which may be
understood also in terms of an evolving spectrum of
quasi-free relaxation times $N(\tau)$ of the spin system.
In a spin glass alloy with a moderate concentration
of magnetic atoms the Korringa mechanism is domi-
nant at high temperatures so that the spectrum of
relaxation times is narrow - of the form of a delta
function peak which is broadened by the additional
channels of relaxation provided by the solute-solute
interactions. With decreasing temperature not only
does the Korringa relaxation rate get smaller
($\tau \propto T^{-1}$) but the spectrum N (τ) becomes wider also.
At temperatures below the "freezing temperature"
T_F it appears that the spectrum becomes very wide
extending from very short times ($\tau \sim 10^{-13}$s) to
very long times ($\tau \to \infty$). It is plausible that many
of the complex spin glass properties result from
this wide spectrum of relaxation times.

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