On the dynamics of the magnetic hyperfine interaction in the cluster glass $Y_{1.1}(Fe_{0.75}Al_{0.25})_2$ above the freezing temperature

1991 J. Phys.: Condens. Matter 3 2713

(http://iopscience.iop.org/0953-8984/3/16/010)

J. Phys.: Condens. Matter 3 (1991) 2713–2719. Printed in the UK

On the dynamics of the magnetic hyperfine interaction in the cluster glass $Y_{1.1}(Fe_{0.75}Al_{0.25})_2$ above the freezing temperature

A Pösinger†, H Winkler‡, W Steiner†, A X Trautwein‡ and M Reissner†

† Institut für Angewandte und Technische Physik, TU Wien, Austria

‡ Institut für Physik, Medizinische Universität Lübeck, Federal Republic of Germany

Received 23 July 1990, in final form 7 December 1990

Abstract. ^{57}Fe Mössbauer measurements were performed in applied fields up to 13.5 T in the temperature range $45 < T < 300$ K. Whereas at high temperatures the recorded spectra could be analysed using a set of subspectra with intensity ratios according to a binomial distribution taking into account nearest-neighbour environments, at lower temperatures the line shape indicates the presence of relaxation effects. For this temperature region a model is proposed, in which magnetic clusters are formed stochastically. These clusters are assumed to decay after a mean lifetime, which increases with increasing applied field and decreasing temperature.

1. Introduction

The spin dynamics of spin and cluster glasses has been investigated by experimental techniques such as ESR, NMR, muon precession, neutron scattering and Mössbauer spectroscopy (see, e.g., Murani 1978). The results suggest a gradual formation of correlations in the spin system with decreasing temperature. Neutron scattering (Murani 1981) and neutron spin-echo experiments (Mezei 1983) on **CuMn** and **AuFe** indicate that dynamical processes take place in concentrated spin glasses above the freezing temperature (T_f). Investigations of the static susceptibility of **CuMn** with concentrations between 1 and 6% Mn exhibit short-range ferromagnetic correlations and fluctuations of the Mn spins up to temperatures of $5T_f$ (Morgownik and Mydosh 1981).

By applying an external field ($\boldsymbol{B}_a$) to cluster glasses the spin dynamics is slowed down to characteristic times observable by Mössbauer spectroscopy (even in temperature regions where without $\boldsymbol{B}_a$ only a quadrupole interaction is present) allowing a detailed study of these magnetic short-range correlations. In $Y(Fe_xAl_{1-x})_2$ long-range magnetic order is only present for Fe concentrations higher than $x = 0.78$. In $\boldsymbol{B}_a = 0$ YFe_2 exhibits two hyperfine fields of 21.2 T and 20.8 T at 4.2 K and 18.5 T and 18.2 T at 300 K in good agreement with Dariel *et al* (1973). For lower Fe contents typical cluster glass behaviour is observed (Reissner *et al* 1984) with freezing temperatures (determined by magnetization measurements in low fields) decreasing from 39 K for $x = 0.75$ to $\simeq$4.5 K for $x = 0.25$. Thus these samples provide us with the possibility of studying the spin dynamics in a wide temperature range at high Fe contents.

0953-8984/91/162713 + 07 $03.50

The chosen compound $Y_{1.1}(Fe_{0.75}Al_{0.25})_2$ crystallizes in the cubic $MgCu_2$ structure type (C15). The Fe and Al atoms are distributed statistically over the 16d positions. Every Fe (Al) atom has six nearest and 12 next-nearest Fe (Al) neighbours and is surrounded by 6 Y nearest neighbours. The temperature (T_A) above which the magnetic splitting of the Mössbauer spectra disappears in the absence of an external field is 45 K. The linear field dependence of the magnetization diminishes for $T < 240$ K.

2. Experimental details

The sample $Y_{1.1}(Fe_{0.75}Al_{0.25})_2$ was prepared by induction melting of the constituent metals under purified argon atmosphere, followed by homogenization at 700 °C in an evacuated quartz tube for two weeks. The purity of the starting materials was 3N for Y and higher than 4N for Fe and Al. The composition was determined by weighing the constituent materials. The weight loss caused by the melting procedure was negligible. X-ray powder diffraction measurements were performed for structural analysis.

^{57}Fe Mössbauer spectra were recorded in transmission geometry on a powdered sample in applied fields of 4.5 to 13.5 T in the temperature range from 50 to 295 K which is in any case above T_f. Measuring times between two and three days were necessary for acceptable statistics of the spectra because of the large distance between source and counter in the spectrometer. The field, created by superconducting coils and applied parallel to the γ-ray direction, was stable during that measuring time. The precision of B_a was ± 0.01 T and the temperature stability for the sample was better than ± 0.5 K. The temperature was measured at zero field using a carbon glass resistor and controlled by a field-independent SrTi sensor. The source was kept at 4.2 K and was situated in a field-compensated area. Velocity calibration was made with an α-Fe foil simultaneously with the measurements.

3. Results and discussion

At high temperatures ($T \geq 280$ K) the shape of the spectra clearly indicates that the induced hyperfine field $\boldsymbol{B}_{ind}$ (defined by $\boldsymbol{B}_{eff} = \boldsymbol{B}_{ind} + \boldsymbol{B}_a$, where B_{eff} denotes the measured effective magnetic field) is oriented antiparallel to $\boldsymbol{B}_a$ (Reissner *et al* 1987). These spectra could be fitted using a set of subspectra with intensity ratios according to a binomial distribution taking into account nearest-neighbour environments and increasing B_{ind} for increasing number of nearest Fe neighbours. Decreasing the temperature still leads to spectra that are completely polarized, i.e. without any indication of the appearance of the second and the fifth line. Furthermore, the inner slopes of the first and sixth line are smooth (see especially the spectra for $T = 240$ and 200 K in figure 1). It was therefore assumed that $\boldsymbol{B}_{eff}$ is oriented antiparallel to $\boldsymbol{B}_a$ down to T_A. There remains uncertainty about the direction of $\boldsymbol{B}_{eff}$ for small B_a because of the small splitting of the spectra. Thus we restricted the present investigation to values of $B_a \geq 4.5$ T.

For the spectra recorded below 280 K a fit with a set of B_{ind} according to a binomial distribution is no longer possible. More subspectra were necessary to obtain a reasonable fit and the resulting B_{ind} turned out very high. This points to the formation of magnetically ordered clusters. An attempt to fit the spectra recorded at $T \leq 160$ K and $B_a = 13.5$ T by means of a distribution of hyperfine fields leads to cluster fields (B_{cl}) larger than 27 T. This value is higher than the hyperfine field measured for the magnetically ordered

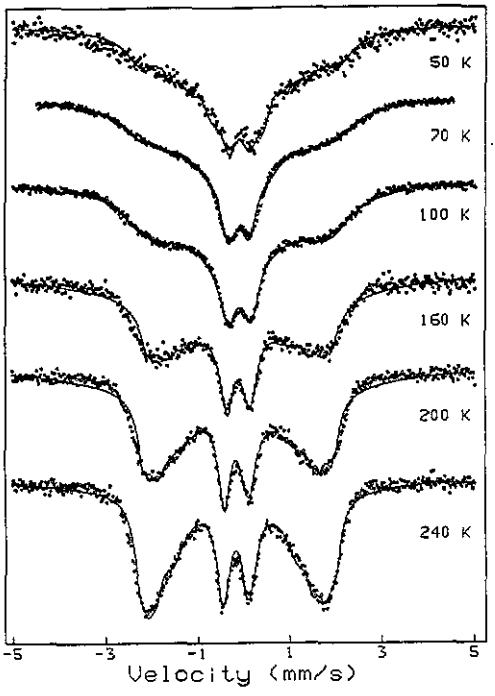

Figure 1. Recorded and fitted (full curve) spectra at $B_a = 13.5$ T for different temperatures.

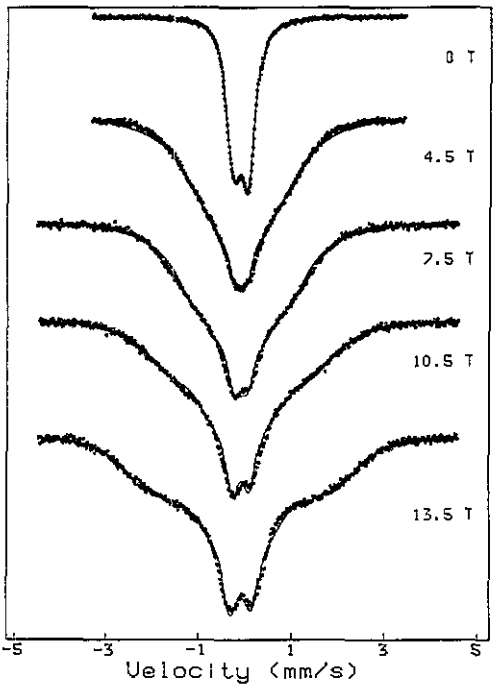

Figure 2. Recorded and fitted spectra at $T = 70$ K for different applied fields B_a.

undiluted compound YFe_2 (Pösinger *et al* 1989). Such a behaviour for a compound where the magnetic exchange interaction of Fe is diluted by Al is hard to explain. The shape of the spectra (see, e.g., those at $T = 70$, 100 and 160 K in figure 1 and figures 2, 3) indicates the presence of relaxation effects (small degree of structure at high and maximum intensity at low absolute velocity, due to both broadening of the outer lines and collapsing of the inner ones (Wickman 1966)). With increasing temperature the relaxation time decreases and therefore the broadening of the outer lines diminishes resulting in spectra with more structure (see, e.g., $T = 200$ and 240 K in figure 1).

A relaxation model that allows the cluster field B_{cl} to flip between the directions parallel and antiparallel to the applied field B_a cannot provide an explanation for the recorded spectra, because the value of $B_a - B_{cl}$ needed to fit the inner part of the spectra (with B_{cl} approximately 8 T) leads to contributions resulting from $B_a + B_{cl}$ in the calculated spectra that are not present in the measured ones.

To explain the observed spectra a simple alternative model is proposed, based on the observation that the magnetic hyperfine interaction is induced only by the application of an external magnetic field to the sample. Thus in strongly spin-correlated regions, which are supposed to form under the influence of this field, a net magnetization is generated giving rise to a magnetic hyperfine interaction at the Fe nucleus. By its action these regions are characterized by a drastic reduction of spin flips in their interior, so they appear as clusters of nearly static magnetic order on the Mössbauer time scale. It is then tempting to explain the dynamics via the assumption that these clusters are fluctuating in the sense that they move around in the sample or form and decay randomly in course of time. This leads to a model in which the individual Mössbauer nucleus is

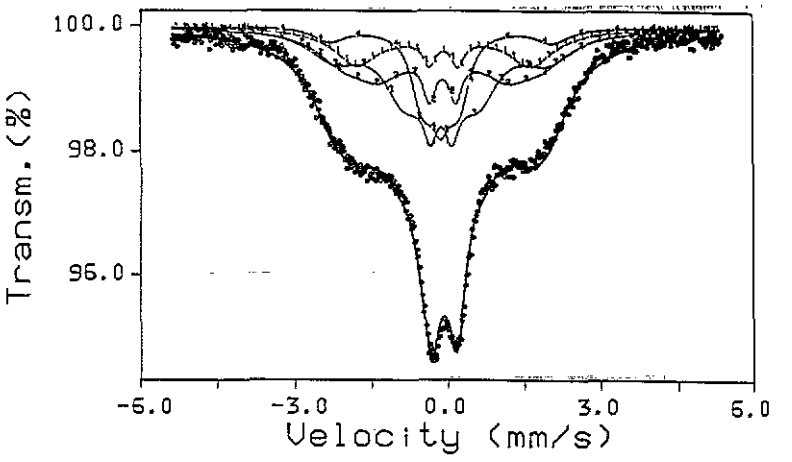

Figure 3. $Y_{1.1}(Fe_{0.75}Al_{0.25})_2$ at 100 K for $B_a = 13.5$ T. The four calculated subspectra are also shown.

assumed to sense a magnetic hyperfine field that is switched on and off stochastically. Such a behaviour can easily be cast into the form of a Blume model (Blume 1968). If there were only a magnetic hyperfine field that fluctuates between zero and some finite value without ever changing its direction, the Wickman formulas (Wickman *et al* 1966) for simple random frequency-modulated resonance absorption would be applicable, because the axis of quantization stays fixed during such a process. Inclusion of an electric field gradient with arbitrary orientation with respect to the effective magnetic field, however, makes this simple approach unsuitable. Therefore, we decided to work with the more general superoperator formulation of the stochastic theory of line shape as developed by Blume (1968) and Clauser and Blume (1971).

The Hamiltonian in terms of which the nuclear Zeeman and the electrostatic hyperfine interactions have been described was chosen to be of the form

$$\mathcal{H}_{hf} = -g\mu_n(B_a - B_{el}f(t))I_z + [eQV_{\dot{z}\dot{z}}/4I(2I-1)]\{\tfrac{1}{2}(3\cos^2\Theta - 1)(3I_z^2 - I^2) + \tfrac{3}{2}\sin 2\Theta[(I_zI_x + I_xI_z)\cos\Phi + (I_zI_y + I_yI_z)\sin\Phi] + \tfrac{3}{2}\sin^2\Theta[(I_x^2 - I_y^2)\cos 2\Phi + (I_xI_y + I_yI_x)\sin 2\Phi]\}$$

where Θ and Φ are the polar angles of $V_{\dot{z}\dot{z}}$ with respect to the laboratory frame and $f(t)$ denotes a random variable taking the values 0 or 1. The relaxation matrix was constructed from the transition probabilities

$$w_{1\to 0} = 1/\tau_{on} \qquad w_{0\to 1} = 1/\tau_{off}$$

which are determined by two mean times, i.e. the lifetime τ_{on} characteristic for the length of periods during which the individual Mössbauer ion is member of an ordered spin cluster and τ_{off} denoting the time intervals in between.

Based on this approach, the measured spectra could be reproduced with an algebraic routine of the kind described by Winkler *et al* (1988). The angle between the γ-ray and $\boldsymbol{B}_{eff}$ was set to zero in accordance with both the experimental set-up and the assumption that $\boldsymbol{B}_{eff}$ is collinear with the applied field $\boldsymbol{B}_a$. In the course of the work it turned out that the integration over Θ and Φ, which is necessary due to the uniform directional

distribution of the crystallites in the sample, could be replaced by taking $\Theta = 54.7°$ and any value for Φ as representative angles without visibly affecting the spectral line shape.

To fit the spectra a superposition of four subspectra was used (figure 3). The line width (FWHM) 0.28 mm s^{-1}, as measured for an α-Fe foil, was taken for all subspectra. Slightly different values were used for the centre shift (CS) of the subspectra ($\Delta CS \simeq$ 0.06 mm s^{-1}) in accordance with the analyses by Reissner and Steiner (1986) of magnetically unsplit spectra of the same sample at room temperature.

The number of subspectra is (in contrast with a fitting procedure that uses a quasi-continuous hyperfine field distribution) very low and might allow the study of the influence of the different Fe environments in an extended temperature range, since the intensity ratios of these subspectra are close to those obtained from a binomial distribution taking into account the nearest-neighbour Fe environments.

The measurements at $B_a = 0$ and room temperature lead to a mean value of 0.3 mm s^{-1} for the electric quadrupole splitting $|eQV_{zz}/2|$ (Reissner and Steiner 1986). Since in the magnetically split spectra the quadrupole interaction is small compared with the magnetic one the calculations are not very sensitive to eQV_{zz}, and thus a common value of -0.3 mm s^{-1} was used for all subspectra. The sign of eQV_{zz} was determined in external fields of 1.5 T at room temperature. The asymmetry parameter was set to zero.

The two fields between which the relaxation takes place are B_a and $B_a - B_{cl}$. For the different subspectra values of approximately 5 T to 12 T were obtained for B_{cl} (figure 3). The dynamical parameters are the relaxation time τ, defined as

$$1/\tau = 1/\tau_{on} + 1/\tau_{off}$$

and a_τ, the ratio of τ_{on}/τ_{off}. For fitting the spectra, a_τ was assumed to increase with increasing B_{cl} for the different subspectra. The differences in τ are small for three of the subspectra, but somewhat larger for the fourth subspectrum, whereas the values for a_τ and B_{cl} exhibit nearly the same differences for all four different subspectra.

Since up to now an assignment of the subspectra to the different Fe environments is uncertain, in the following discussion the mean values $\bar{B}_{cl}$, $\bar{\tau}$, $\bar{a}_\tau$, weighted by means of the area of the subspectra, are used.

$\bar{B}_{cl}$ is nearly constant for all temperatures T and for all applied fields B_a in the range investigated (figure 4).

Within the proposed model $\bar{\tau}_{on}$ is expected to decrease with increasing T and decreasing B_a, since the possibility of coupling to other Fe atoms diminishes, and for T high enough only paramagnetic behaviour is possible. The results obtained for $\bar{a}_\tau$ fulfil these requirements. The product $\bar{B}_{cl}\bar{\tau}_{on}/(\bar{\tau}_{on} + \bar{\tau}_{off})$ represents a mean moment for the stochastically varying time where the atoms are coupled to clusters and corresponds therefore to a magnetization, which on the other hand should also be observed in bulk magnetic measurements. The different characteristic measuring times of the two experiments are not essential, since B_{cl} which is proportional to the moment is always antiparallel to B_a and only $\bar{\tau}_{on}$ decreases with increasing T and decreasing B_a. The results of both sets of measurements are compared for two typical temperatures (160 and 240 K) in figure 5. The bulk magnetic measurements (left-hand axis) were performed in fields up to 6.7 T and in the temperature range $50 < T < 270$ K. The magnetization (σ) as a function of field is curved for $T < 240$ K. For all T the product $\bar{B}_{cl}\bar{\tau}_{on}/(\bar{\tau}_{on} + \bar{\tau}_{off})$ (right-hand axis in figure 5) is proportional to σ with 11 T/μ_B, which is a further test for the validity of the model.

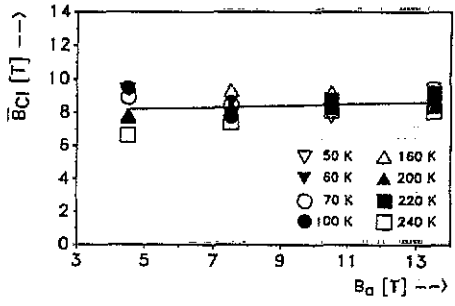

Figure 4. Dependence of the mean cluster field $\bar{B}_{cl}$ on the applied field B_a for different temperatures. The line is only a guide for the eyes.

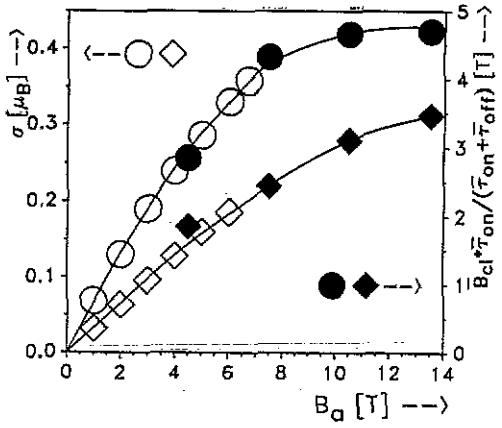

Figure 5. Field dependence of the magnetization σ (left-hand scale, open symbols) and $\bar{B}_{cl}\bar{\tau}_{on}/(\bar{\tau}_{on}+\bar{\tau}_{off})$ (right-hand scale, full symbols) for $T = 160$ K (●, ○) and 240 K (◆, ◇). The curves are only guides for the eyes.

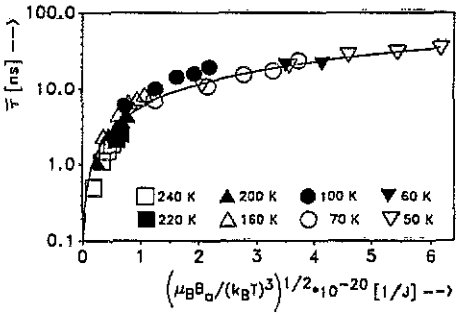

Figure 6. Mean relaxation time $\bar{\tau}$ versus $[\mu_B B_a/(k_B T)^3]^{1/2}$ on a semi-log scale.

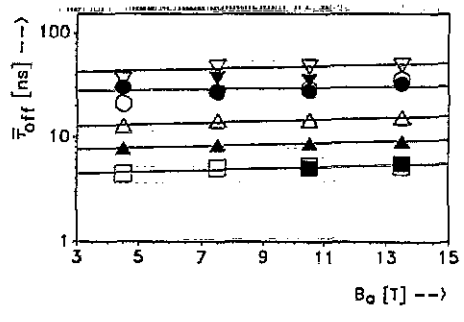

Figure 7. Mean $\bar{\tau}_{off}$ versus B_a for different temperatures (symbols as in figures 4 and 6). The lines are only guides for the eyes.

For $\bar{\tau}$ a dependence according to

$$\bar{\tau} = b[\mu_B B_a/(k_B T)^3]^{1/2}$$

with $b = 5.8 \times 10^{-29}$ J s (figure 6) was found for all values of T and B_a. The mean times $\bar{\tau}_{off}$, during which the Fe atoms are not incorporated into a cluster, are only weakly dependent on B_a (figure 7), indicating that B_a has little influence on the formation of magnetic clusters, whereas the increase of $\bar{\tau}$ with increasing B_a indicates that B_a hinders the decay of the clusters.

It is remarkable that relaxation effects are present over a very wide range in temperature up to $T \approx 7T_f$. Even below, but not far from T_f the clusters are not completely frozen, but the simple model with only one direction for B_{cl} fails for $T < T_f$.

At the present state of the investigations it is not certain whether the different subspectra of which the spectra are composed, are due to the different environments of the Fe atoms. We now try to prove that this is possible, and want to test the model of

forming and decaying clusters on other concentrated spin glasses. First measurements on AuFe point to the applicability of the proposed concept for these samples also.

To sum up: relaxation effects dominate the magnetic behaviour up to $7T_f$ in $Y(Fe_{0.75}Al_{0.25})_2$. For this temperature region the Mössbauer spectra recorded in applied fields up to 13.5 T support a model, formulated to explain the dynamic behaviour, in which magnetic clusters are formed stochastically far above the freezing temperature. These clusters are not stable, but are assumed to decay after a mean lifetime that increases with increasing B_a and decreasing temperature. The applied field hinders the decay of the clusters but has little influence on their formation.

Acknowledgments

This work was supported in part by the Austrian Fonds zur Förderung der wissenschaftlichen Forschung, grant number P6938.

References

Blume M 1968 *Phys. Rev.* **174** 351–8
Clauser M J and Blume M 1971 *Phys. Rev.* B **3** 583–91
Dariel M P, Atzmony U and Lebenbaum D 1973 *Phys. Status Solidi* b **59** 615–20
Mezei F 1983 *J. Magn. Magn. Mater.* **31–4** 1327–30
Morgownik A F J and Mydosh J A 1981 *Phys. Rev.* B **24** 5277–83
Murani A P 1978 *J. Physique Coll.* **39** C6 1517–26
—— 1981 *J. Magn. Magn. Mater.* **22** 271–81
Pösinger A, Reissner M and Steiner W 1989 *Physica* B **155** 211–4
Reissner M, Pösinger A and Steiner W 1987 *J. Magn. Magn. Mater.* **70** 165–7
Reissner M and Steiner W 1986 *Hyperfine Interact.* **28** 1017–20
Reissner M, Steiner W, Kappler J P, Bauer P and Besnus M J 1984 *J. Phys. F: Met. Phys.* **14** 1249–59
Wickman H H 1966 *Mössbauer Effect Methodology* vol 2, ed I J Gruverman (New York: Plenum) pp 39–66
Wickman H H, Klein M P and Shirley D A 1966 *Phys. Rev.* **152** 345–57
Winkler H, Bill E, Trautwein A X, Kostikas A, Simopoulos A and Terzis A 1988 *J. Chem. Phys.* **89** 732–40