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1998 J. Phys.: Condens. Matter 10 9849

(http://iopscience.iop.org/0953-8984/10/43/029)

J. Phys.: Condens. Matter **10** (1998) 9849–9858. Printed in the UK PII: S0953-8984(98)92892-2

Spin correlation times in $Cr_{0.825}Fe_{0.175}$ above the freezing temperature

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Received 31 March 1998

Abstract. Magnetic correlations up to ten times the freezing temperature are determined in the spin glass $Cr_{0.825}Fe_{0.175}$ by high-field Mössbauer spectroscopy. The model based on stochastic formation and decay of these correlated regions, used to analyse the spectra of the spin glass systems $Y(Fe, Al)_2$ and **Au**Fe, is again successfully applied, irrespective of the completely different matrix materials. In agreement with the results obtained for these systems a power law for the temperature dependence of the correlation time with an exponent close to -2 is observed. This exponent is also found for the temperature dependence of the relaxation rate if the μSR measurements on the same sample reported by Telling and Cywinski are analysed according to a Kohlrausch law.

1. Introduction

Dynamics of magnetic correlations have been observed in spin glasses well above the freezing temperature (T_f). Detailed investigations were performed especially for **Au**Fe by μSR (Uemura *et al* 1985, Keren *et al* 1996), neutron spin echo (Mezei 1983) and high-field Mössbauer (HFM) measurements (Pösinger *et al* 1993, Bogner *et al* 1998b). For the analysis of the Mössbauer spectra in an applied magnetic field a model was proposed based on a two level jump process, between a state in which the probe atom is embedded in a correlated region—the lifetime of this state is τ_{on}—and a state in which the atom experiences no correlation. This type of analysis was also applied successfully to explain the recorded spectra of $Y(Fe,Al)_2$ in a broad range of Fe concentrations. In contrast to **Au**Fe, where Fe is embedded in a diamagnetic host, in the latter series a Pauli paramagnetic matrix is present. The temperature (T) dependence of τ_{on} follows between $2\ T_f \leqslant T \leqslant 10\ T_f$ a power law. Two different exponents were obtained: -2 for spin glasses with high Fe content (e.g. $Au_{0.85}Fe_{0.15}$, $Y(Fe_{0.75}Al_{0.25})_2$) and -1 for samples with low Fe content ((e.g. $Au_{0.95}Fe_{0.05}$, $Y(Fe_{0.40}Al_{0.60})_2$) (Pösinger *et al* 1993, Bogner *et al* 1998b).

To check if this power law is more generally valid, we extended the investigations to $Cr_{1-x}Fe_x$, for which a change is reported from ferromagnetism to antiferromagnetism with increasing Cr content. The spin glass behaviour appears in the concentration range $x = 0.16$–0.19 (Burke *et al* 1983, Burke and Rainford 1983b, Dubiel *et al* 1985). From this region a sample with $x = 0.175$ was chosen for the present investigation.

Moreover the same sample has already been measured by the μSR technique by Telling and Cywinski (1995), which to our knowledge for the first time allows a comparison of the dynamics detected by both methods. Whereas the application of a large external magnetic

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field is necessary to slow down the dynamics of the magnetic correlations to times observable by Mössbauer spectroscopy (1 ns to 100 ns, given by the Larmor period of the absorbing nucleus), no external field is used in these μSR measurements, which can prove magnetic correlations with lifetimes in between 50 ns and 50 μs (given by the lifetime of the muon and slightly varied by properties of the muon beam used (Schenck 1985)).

A short description of the HFM investigations has already been presented at *ICAME'97* (Bogner *et al* 1998a).

2. Experiment

The investigated $Cr_{0.825}Fe_{0.175}$ sample (kindly donated by S K Burke) crystallizes in the bcc structure. The description of preparation and sample characterization can be found in Burke and Rainford (1983a).

A vibrating sample magnetometer was used to estimate the field dependence of T_f from the d.c. magnetic measurements between 2 K and 100 K in various external magnetic fields.

All ^{57}Fe Mössbauer experiments were performed in transmission geometry using conventional constant acceleration setups. The zero-field spectra, some of which can be seen in figure 1, were analysed solving the complete Hamiltonian for magnetic and quadrupole interaction in the fast relaxation limit.

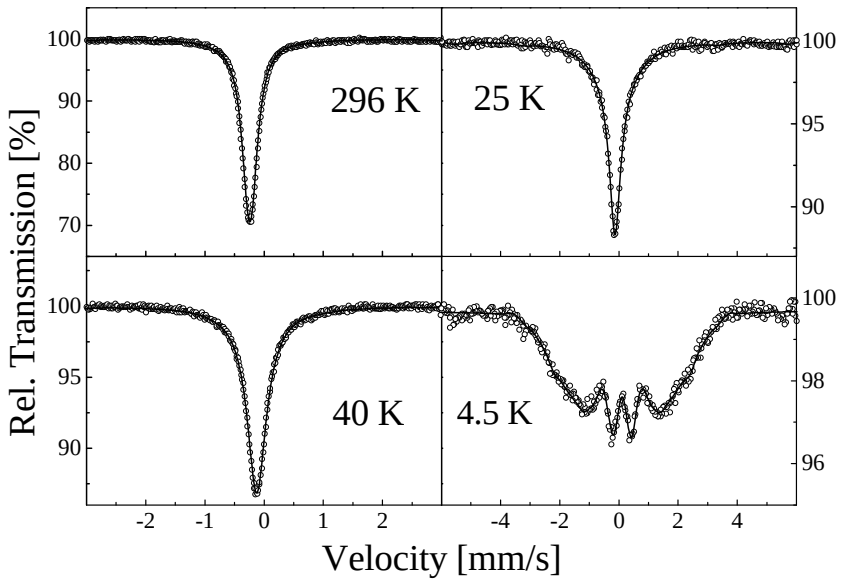

Figure 1. Examples of zero-field Mössbauer spectra recorded at various temperatures.

At room temperature without external field unsplit spectra with slightly asymmetric shape were recorded. Additionally a small magnetic hyperfine split contribution with 3% relative intensity was detected and ascribed to α-Fe precipitates in the sample. As a first approximation we assume a random distribution of the remaining amount of 17.0 at.% Fe in the Cr matrix, and use a binomial distribution function to calculate probabilities for the different environments for the first nearest neighbour shell of the ^{57}Fe probe atom. Five distinct surroundings (zero, one, two, three and four nearest (nn) Fe neighbours) with relative intensities higher than the resolution of the experiment are obtained. To conform with the

HFM analyses we reproduced the asymmetric shape by a superposition of five single line subspectra with relative intensities according to the various probabilities of the surroundings, and with a change in isomer shift (IS) of 0.015 mm s^{-1} per Fe atom in the neighbourhood. With -0.155 mm s^{-1} (IS given relative to the ^{57}Co**Rh** source at room temperature) the IS of the 4 nn Fe surroundings is the largest one. The difference in IS was kept fixed for all temperatures. The obtained line widths (0.26 mm s^{-1}) are the same as found for the α-Fe calibration spectra.

HFM measurements in the temperature range 50 to 300 K with applied fields ($\boldsymbol{B}_a$) of 7.5 T, 10.5 T and 13.5 T were performed. The direction of $\boldsymbol{B}_a$ was parallel to the γ-ray direction and the source (again ^{57}Co**Rh**) was placed in a field compensated area ($\boldsymbol{B}_a < 0.1$ T). The temperature of the source was kept between 4.2 K and 35 K, depending on the absorber temperature, which was measured by a carbon glass resistor and controlled by a field-independent SrTi sensor. During the rather long measuring times (3–5 days), which were due to the large distance of 50 cm between source and proportional counter, a temperature stability of ± 0.1 K was achieved (for details of the experimental HFM setup see Pösinger *et al* (1989)).

3. Results

The temperature dependence of the magnetization shows the typical maximum in the zero-field cooled curve and the difference to the field cooled curve as characteristic for spin glasses (figure 2). As expected, the maximum shifts to lower temperatures and the peak broadens with increasing measuring field. Defining the freezing temperature, T_f, as the maximum in the zero-field cooled curve a value of $T_f = 20$ K is determined for the smallest applied field ($B_a = 1.6$ mT). This value is in good agreement with findings of Palumbo *et al* (1982). The field dependence of T_f fits to a power law with exponent ~ 0.32, which is close to 0.43 reported for a sample with $x = 0.16$ (Palumbo *et al* 1983). With regard to the experimental uncertainties in the determination of T_f a value of two thirds, as theoretically proposed by de Almeida and Thouless (1978), is not unreasonable; an exponent of two according to Gabay and Toulouse (1981) must be, however, excluded (figure 3). It is also worth noting that the presently determined exponent differs from the exponent 1 as found for the $Cr_{75}Fe_{25}$ alloy (Dubiel *et al* 1987). In the latter case, however, the system undergoes a re-entrant transition into the spin glass state with non-vanishing spontaneous magnetization (ferromagnetic spin glass).

A freezing temperature, T_f^*, can also be determined in two ways from the Mössbauer spectra recorded in zero field as a function of temperature: (i) from the mean line width at half maximum, HWHM_{av}, and (ii) from the average hyperfine field, B_{av}, which was obtained by the integration of the hyperfine field distribution histograms derived from the spectra (figure 4) (Dubiel *et al* 1985). The results obtained in this way can be seen in the inset of figure 3. The former yielded $T_f^* \approx 39$ K, the latter $T_f^* \approx 32$ K.

The difference of 12–19 K between the freezing temperature determined from Mössbauer spectroscopy and the one determined from d.c. magnetic measurements is by far larger than the one found in **Au**Fe, in which the different time windows of the methods used was assumed to be responsible for (Sarkissian 1981). In the case of superparamagnetic particles, the ratio between the blocking temperature determined from the Mössbauer spectra and that found from the magnetization after zero-field cooling can be taken as a measure of the strength of interactions between the particles (Hanson *et al* 1995). If this value lies in the range 4–7, the particles do not interact with each other, while a value of 1 indicates a strong interaction. The magnetization curves for superparamagnetic particles and those for spin

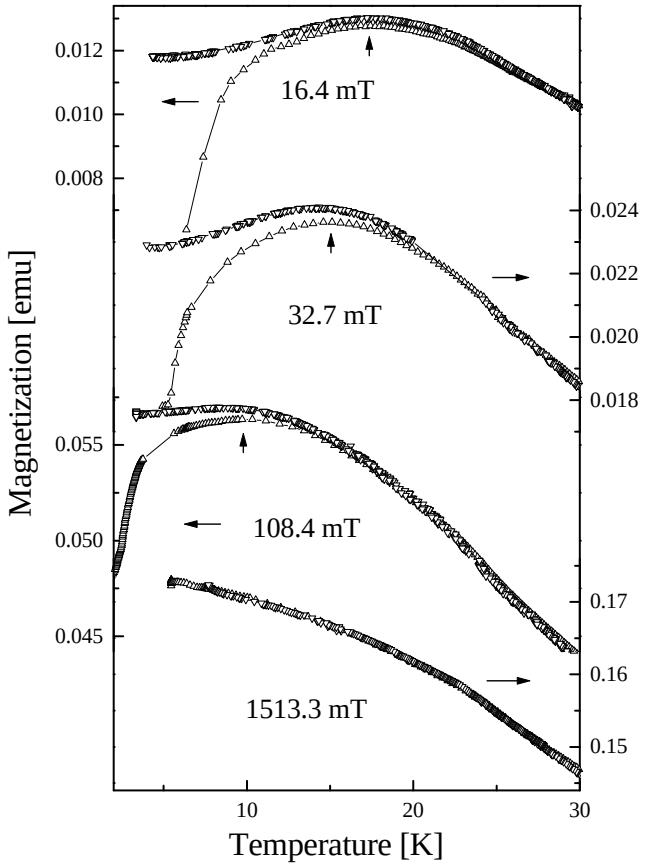

Figure 2. Some examples for the temperature dependence of the zero-field (△) and field (▽) cooled bulk magnetization in different applied fields. The upward arrows indicate the maximum of the zero-field cooled bulk magnetization, which determines the freezing temperature T_f.

glasses are similar to each other. This suggests that also for spin glasses the ratio between the spin freezing temperature found from the Mössbauer spectra and the one determined from the zero-field magnetization curves can be taken to measure the strength of interaction between spins. As in the present case the ratio lies in the range 1.6–2.0, this might be an indication that the spins localized at iron atoms interact with each other rather strongly.

The shape of the HFM spectra of $Cr_{0.825}Fe_{0.175}$ above T_f is characteristic for the influence of dynamics—pronounced minimum at low and broad shoulders at high absolute velocities (figure 5). The similarity of the spectra with those found for **Au**Fe and $Y(Fe,Al)_2$ in the spin glass region suggests the applicability of the same analysis (Pösinger *et al* 1993, Bogner *et al* 1998b), which takes into account dynamics based on a Blume formalism (Blume 1968). The model process can be described by a jump in time between two discrete hyperfine field levels, $\boldsymbol{B}_{hf1} = \boldsymbol{B}_{cl} - \boldsymbol{B}_a$ and $\boldsymbol{B}_{hf2} = \boldsymbol{B}_{ind} - \boldsymbol{B}_a$. For a mean lifetime τ_{on} the ^{57}Fe nucleus is in addition to the applied field $\boldsymbol{B}_a$ exposed to a field $\boldsymbol{B}_{cl}$, which is caused by strong interaction of the on-site spin with surrounding spins. This state of strong correlation alternates stochastically with a non-correlated state. In this second state the nucleus feels now the field $\boldsymbol{B}_{ind}$ (caused by the action of $\boldsymbol{B}_a$ on the atomic moment), which is present for a stochastic time τ_{off}. During this time the moment behaves in a paramagnetic way.

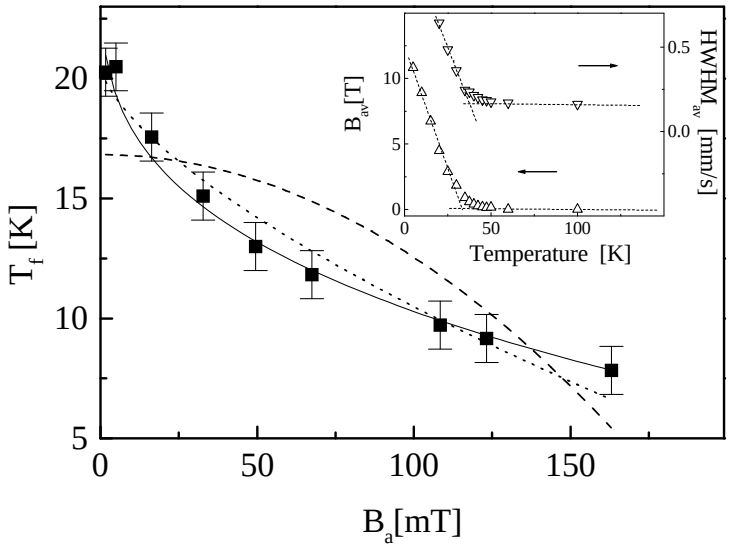

Figure 3. Field dependence of T_f determined by d.c. magnetic measurements. The lines represent power laws with an exponent of 0.32 (——) resulting from a least squares fit of the data and $2/3$ (· · · · · ·) as proposed by de Almeida and Thouless (1978), as well as 2 (– – –) according to Gabay and Toulouse (1981). Inset: estimation of the freezing temperature T_f^* from the analysis of the zero-field Mössbauer measurements using the mean hyperfine fields B_{av} (△) and mean half widths HWHM_{av} (▽).

Taking for the different nn Fe surroundings the relative intensity ratios, the differences in IS from the zero-field Mössbauer spectra and allowing distinct $\boldsymbol{B}_{cl}$ and $\boldsymbol{B}_{ind}$, both increasing with number of nearest Fe neighbours, all HFM spectra could be fitted with one set of five subspectra (figure 6). With no quadrupole interaction present, the number of free parameters in the analysis reduces significantly and thus supports a unique description.

Table 1. Selected values for Fe with 1 nn Fe in $Cr_{0.825}Fe_{0.175}$ at 13.5 T.

	50 K	150 K	250 K
B_{cl} (T)	21.0	15.7	12.0
B_{ind} (T)	8.6	1.0	0.0
τ_{on} (ns)	28	5.2	2.0
τ_{off} (ns)	56	33	17

The high temperature ($\geqslant$250 K) spectra recorded with $\boldsymbol{B}_a = 7.5$ T, 10.0 T and 13.5 T can be described without taking into account dynamic effects. The intensities for the $\Delta m = 0$ transitions are vanished indicating complete polarization of the moments by the applied field. At 250 K, however, the spectrum becomes more broadened. A consistent fit using the fast relaxation limit as for the spectra at higher T would now either require an increase of the number of subspectra or an unreasonable larger half width of the subspectra with smaller hyperfine field. The fit by means of our stochastic model gives the more satisfying answer of slightly larger hyperfine fields $\boldsymbol{B}_{ind}$ and $\boldsymbol{B}_{cl}$ as well as longer stochastic times τ_{on} and τ_{off}. Thus it is obvious that on the one hand at about 12.5 T_f and 13.5 T the upper border of the time window of the present experiment is reached. This means on the

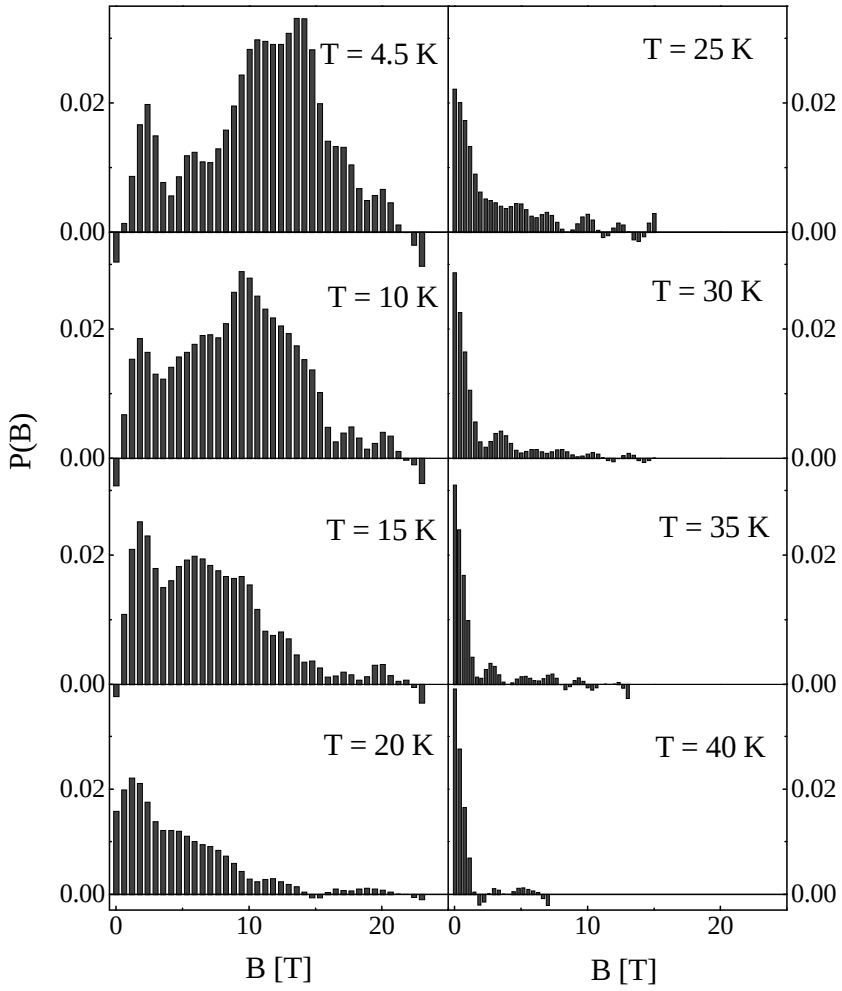

Figure 4. Hyperfine field distribution histograms derived from the zero-field Mössbauer spectra.

other hand that the influence of the dynamics can be resolved in the used field range up to temperatures of approximately 10 T_f.

For comparable Fe nn environments $\boldsymbol{B}_{cl}$ is 10–20% smaller than the one found for the **Au**Fe system (Bogner *et al* 1998b) but exhibits a similar temperature dependence. In the whole temperature range investigated $\boldsymbol{B}_{cl}$ is oriented antiparallel to the applied field, since for each subspectrum the intensity of the $\Delta m = 0$ transitions vanishes. The values for $\boldsymbol{B}_{ind}$ as a function of $\boldsymbol{B}_a/T$ follow Langevin functions with saturation fields of 20 T, 18 T, 16 T, 14 T and 6 T for the 4, 3, 2, 1 and 0 nn Fe surroundings, respectively. This supports strongly the model assumption of a paramagnetic state for a certain stochastic time.

In the temperature range 12.5 T_f down to 5 T_f only one single set of stochastic times τ_{on} and τ_{off} for a given temperature was used, disregarding the different local environments. If the temperature is decreased further, different times for these environments become necessary. Below 7 T_f the influence of $\boldsymbol{B}_a$ on the correlation time τ_{on} becomes more pronounced indicating that correlated states are more long lived at higher applied fields.

Both τ_{on} and τ_{off} increase with decreasing temperature similar to $Au_{0.85}Fe_{0.15}$ (Bogner

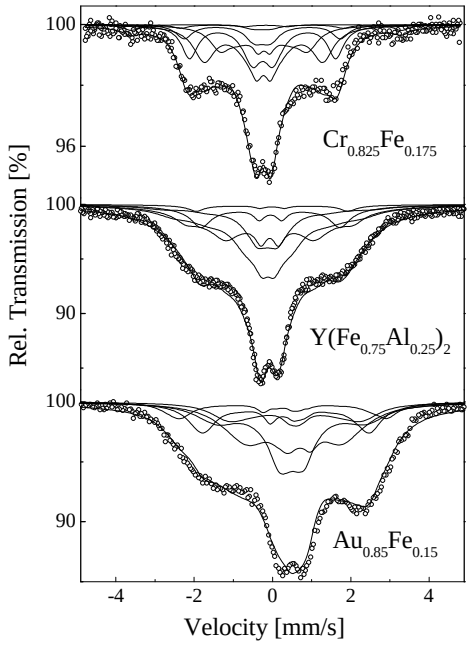

Figure 5. Typical high-field Mössbauer spectra of $Au_{0.85}Fe_{0.15}$, $Y(Fe_{0.75}Al_{0.25})_2$ and $Cr_{0.825}Fe_{0.175}$ at 100 K measured in an applied field of 13.5 T. The subspectra according to the nn environments used in the analysis are also shown.

et al 1998b). In a double logarithmic representation of τ_{on} as a function of the normalized temperature T/T_f a linear dependence can be determined (figure 7). The slope is close to -2 as for the Fe rich compounds $Au_{0.85}Fe_{0.15}$ and $Y(Fe_{0.75}Al_{0.25})_2$. To stress the point, this exponent is -1 for low Fe content samples of the **Au**Fe and $Y(Fe,Al)_2$ systems. The slope remains the same if $\tau_{on}(\boldsymbol{B}_a = 0)$ is considered, which can be obtained from the linear extrapolation of τ_{on} measured at different $\boldsymbol{B}_a$ to zero field (figure 7). Thus the high fields necessary to obtain a proper resolution in the Mössbauer experiments seem to be of minor influence on the formation of the correlated regions, which can be seen as precursors for the appearance of a spin glass state at low temperature.

Zero-field μSR measurements above the freezing temperature have already been performed on the same $Cr_{0.825}Fe_{0.175}$ sample (Telling and Cywinski 1995). The time differential μSR spectra are described by a stretched exponential relaxation function of the (Kohlrausch) form

$$G(t, T) \propto \exp[-(\lambda(T)t)^{\beta(T)}] \tag{1}$$

where λ is the muon depolarization rate and β an empirical parameter, which describes the distribution of depolarization rates as function of temperature. This exponent β is close to 1 at high temperatures (300 K), which points out that all muons detect a single depolarization rate, and decreases to $\beta = 0.33$ at approximately 50 K, indicating that a distribution of depolarization rates must be present. If we take λ to be directly proportional to the spin correlation time of the system (and multiply it with a unit factor 1 s^2), it can be compared with the time τ_{on}. In the temperature range of the μSR experiment a power law with exponent -2 is also found (figure 7). It must be stressed that the muon is stopped at an interstitial site and thus is influenced by the fluctuating fields at these sites. The ^{57}Fe probe

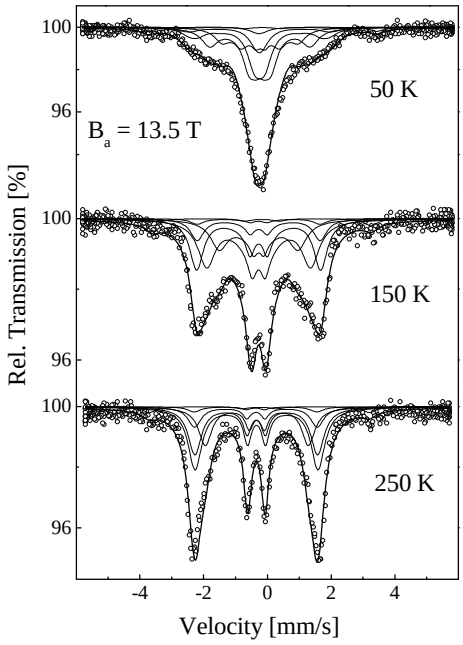

Figure 6. ^{57}Fe Mössbauer spectra of $Cr_{0.825}Fe_{0.175}$ at different temperatures measured in an applied field of 13.5 T. The subspectra according to the nn environments are also shown. Selected values for Fe with 1 nn are given in table 1.

atom is in contrast embedded on a well defined lattice site. The difference in the absolute values of λ and τ_{on} is of the order of one magnitude and is obviously due to the different experimental methods used. In addition, in the temperature range in which β remains close to 1, just one value for τ_{on} for all different environments was obtained. Thus not only a similarity in the temperature dependence of the correlation time but also of the highly probable distribution of correlation times follows from both experiments.

Whereas the HFM analysis starts from a given microscopic picture and can thus provide detailed information on hyperfine fields and correlation times for distinct nn Fe surroundings, the analysis of μSR spectra with a stretched exponential lacks this microscopic information. Therefore an analysis for both measuring techniques, starting from a common microscopic model of the spin glasses above the freezing temperature, seems highly desirable.

4. Conclusion

Dynamics of magnetic correlations above the freezing temperature in the spin glass phase of the alloy **Cr**Fe can be observed by high-field Mössbauer spectroscopy up to at least 10 T_f. A recently developed model, which makes use of a stochastic switching in time between different states of correlation, and which was used to describe the spin glass systems **Au**Fe and $Y(Fe,Al)_2$, is again successfully applied irrespective of the magnetically highly different matrix materials. Concerning the temperature dependence of the stochastic times, $Cr_{0.825}Fe_{0.175}$ can be classified into the group of Fe rich spin glasses of **Au**Fe and $Y(Fe,Al)_2$, with both stochastic times decreasing with temperature and the power law exponent of -2 for τ_{on}. The small concentration range of the spin glass phase in the **Cr**Fe system makes further investigation in this phase (i.e. at the ferromagnetic border of the spin glass phase)

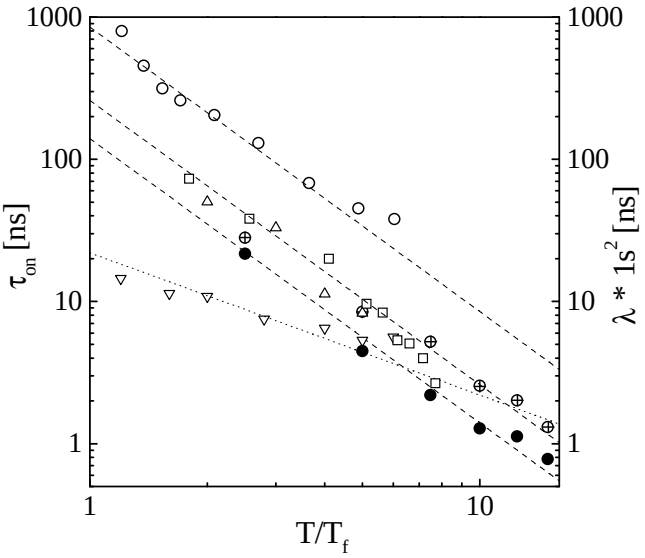

Figure 7. Mean lifetime of correlated states at $\boldsymbol{B}_a = 13.5$ T as a function of reduced temperature of $Cr_{0.825}Fe_{0.175}$ (⊕), $Au_{0.85}Fe_{0.15}$ (△) $Y(Fe_{0.75}Al_{0.25})_2$ (□) and $Au_{0.95}Fe_{0.05}$ (▽). Mean lifetime of correlated states of $Cr_{0.825}Fe_{0.175}$ extrapolated to $\boldsymbol{B}_a = \mathbf{0}$ (●) in comparison with the muon depolarization rate λ (Telling and Cywinski 1995), multiplied by a unit factor 1 s^2 (○). The lines indicate the slopes according to a power law with exponent -2 (– – –) and -1 (· · · · · ·), respectively.

difficult if one keeps in mind the uncertainties in sample preparation, especially in achieving the necessary homogeneity of the distribution of the Fe atoms in the Cr matrix. From the good agreement of both the temperature dependence of the depolarization rate with the HFM correlation time and the temperature interval for a narrow distribution of these quantities, as well as from the fact that the μSR measurements were performed in zero applied field, it can be concluded that the high applied fields in the Mössbauer experiments do not alter the distribution of magnetic correlations drastically, but only slow down their dynamics.

Acknowledgments

We are grateful to R Cywinski and I A Campbell for fruitful discussions. This work was partly supported by the Austrian FWF (project No P11584). One of us (SMD) thanks the State Research Committee (KBN), Warsaw, for financial support within the Polish–Austrian project No 12.

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