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Spin dynamics in the reentrant spin glass $(Fe_{0.65}Ni_{0.35})_{1-x}Mn_x$ *

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The spin dynamics in the reentrant spin glass $(Fe_{0.65}Ni_{0.35})_{1-x}Mn_x$ has been studied by zero, longitudinal and transverse field μSR. In the ferromagnetic reentrant and pure spin glass regimes ($x \leqslant 0.175$), zero field experiments reveal a stretched exponential muon relaxation with a universal behaviour of the dynamic exponent β above the spin glass transition. There are no qualitative differences between the ferromagnetic and paramagnetic phases. In transversal field μSR experiments the divergence of the relaxation rate close to the spin glass transition is suppressed for manganese doping up to $x = 0.113$ but enhanced for slightly higher doping ($x \geqslant 0.12$). We understand this behaviour as a crossover from an itinerant to a more localized state of the 3d electron system. This is also supported by the fact that in the highly doped regime with dominant antiferromagnetic interactions the muon relaxation rate diverges above the antiferromagnetic transition temperature.

The Mn-doped invar system $(Fe_{0.65}Ni_{0.35})_{1-x}Mn_x$ is a metallic model system to study the competition of ferromagnetic and antiferromagnetic exchange between densely packed moments. Depending on the Mn concentration ferromagnetic, reentrant spin glass, spin glass and antiferromagnetic order transitions are found in different macroscopic and microscopic experiments [1–3]. For systems where the mean magnetic coupling strength J_0 is of the order of the width of the magnetic coupling strength distribution δJ two reentrant spin glass phases are predicted [4,5]: first a freezing and irreversible behaviour of transverse spin components (phase M1), and then at lower temperatures an additional irreversible behaviour of the longitudinal spin component (phase M2). At lowest temperatures the long range longitudinal order should coexist with the frozen transverse spin components.

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To study the local spin ordering and spin dynamics in the different magnetic regimes we have performed zero field (ZF), longitudinal field (LF) and transverse field (TF) μSR. The samples with Mn doping $x = 0.09$, 0.113, 0.12 are in the ferromagnetic reentrant spin glass regime, $x = 0.14$ is the triple point concentration of paramagnetic, ferromagnetic and spin glass phases, $x = 0.175$ in the pure spin glass regime, and $x = 0.24$ in the antiferromagnetic reentrant regime. The ZF and LF experiments were performed partly with the MUSR spectrometer at the ISIS pulsed muon facility and with the GPD spectrometer at the PSI decay beamlines, where also the TF experiments have been done. To investigate the non-ergodic behaviour in the spin glass phases we used two different paths: zero field warming after zero field cooling and zero field warming after longitudinal field cooling.

For all samples except the $x = 0.24$ specimen the muon spin relaxation in the temperature range 15–300 K is mainly due to spin fluctuations controlled by the spin glass transition which occurs between 50 K and 70 K [2]. A difference between the zero field and 0.01 T field cooled experiments has not been observed. The ZF experiments show an increase of the relaxation rate with decreasing temperature until the relaxation rate diverges between 80 and 100 K, depending on the Mn concentration. Above this temperature the μSR spectra have been fitted with a stretched exponential relaxation $\propto \exp[-(\lambda t)^{\beta}]$ representing the electronic damping. By using a product of this relaxation function with a static Gaussian Kubo–Toyabe function we have included the additional damping contribution from the ^{55}Mn nuclear moments determined from higher temperature ($\approx$ 200 K) longitudinal field experiments. Figures 1 and 2 show the obtained temperature dependences of the relaxation rate λ and the exponent β in the zero field experiments. Surprisingly, there is no qualitative difference in the muon relaxation between the ferromagnetic and the paramagnetic phases above the spin glass transitions. Therefore in the ferromagnetic phase the mean local magnetic field at the muon site must be zero which can be well understood for a muon site of cubic symmetry in this fcc lattice. In addition, the field distribution has to be very narrow. We interpret this with the very itinerant character of the 3d magnetic moments which leads to an effective averaging over the solid solution of iron, nickel and manganese atoms.

The dynamical exponent β shows a universal temperature dependence. It varies from ≈ 1 at higher temperatures ($T > 150$ K) to 0.3–0.5 in the vicinity of the spin glass transition. The variation in the absolute values between the different samples is caused by an uncertainty in the signal amplitude. The same behaviour has been found in μSR studies of disordered systems like amorphous DyAg [6], Al-doped YMn_2 [7] and dilute spin glasses [8]. The *temperature dependent* deviations from a pure exponential muon relaxation function may have several reasons. The commonly used interpretation assumes a distribution of exponential relaxation times of the magnetic moments due to their inhomogeneous coupling. This cannot be differentiated from a nonexponential relaxation of the individual magnetic moments due to a feedback in the relaxation process as proposed by different hierarchical models for spin glasses (e.g. [9]). A third scenario is a distribution of local muon relaxation times due to an inhomogeneous hyperfine coupling to the magnetic environment. In principle,

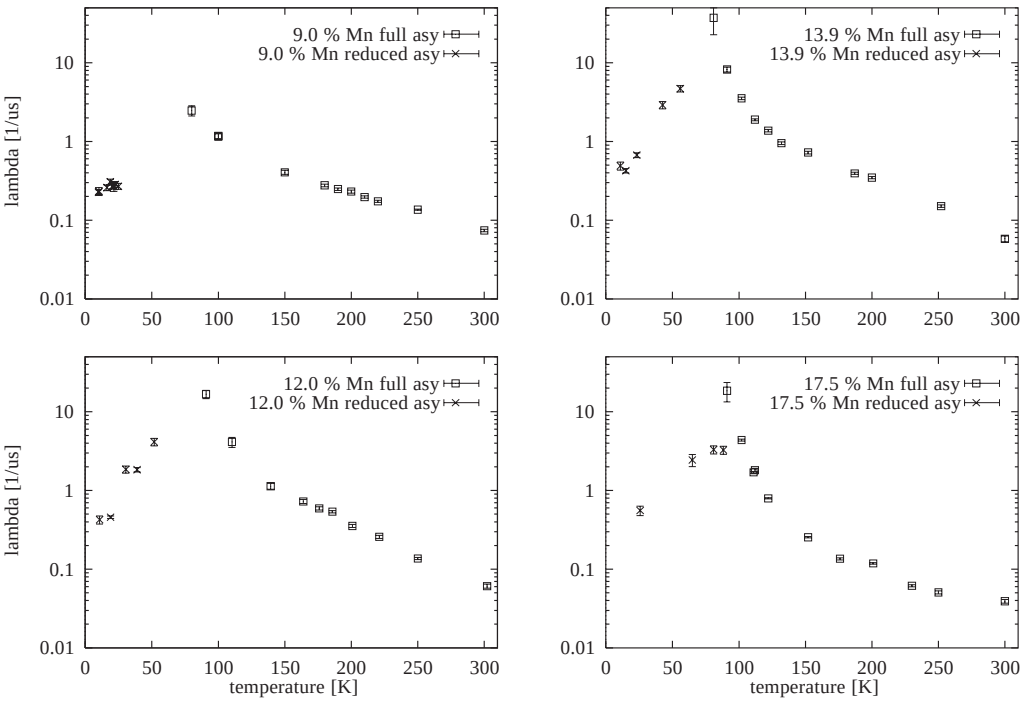

Fig. 1. Temperature dependence of the relaxation rate λ in $(Fe_{0.65}Ni_{0.35})_{1-x}Mn_x$ at four different manganese concentrations. The open squares represent the high temperature fits with the full signal amplitude, the crosses the low temperature fits with a reduced signal amplitude (see text).

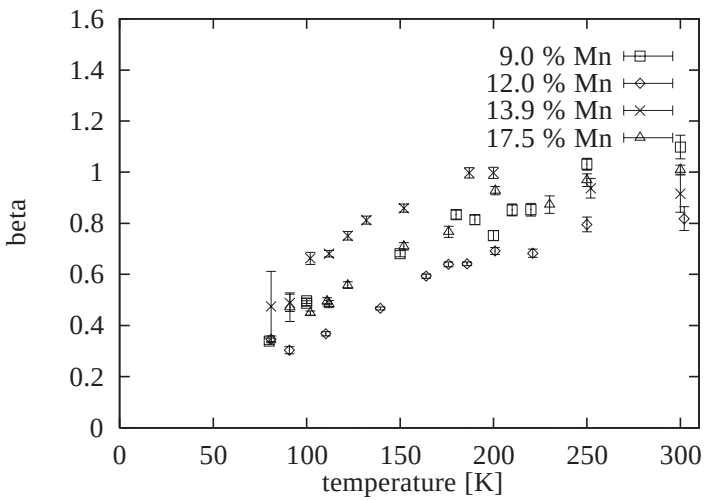

Fig. 2. Temperature dependence of the dynamical exponent β in the high temperature fits for different manganese concentrations.

this is a temperature independent process but for a Lorentzian field distribution a comparison of the muon time window with a time scale proportional to the temperature dependent mean magnetic relaxation rate ν_{eff} [10] can cause a temperature variation of the exponent β. In this picture the observed change of β would reflect the reduction of ν_{eff} with the decrease in temperature.

Below 80 K a recovery of $\approx 1/3$ of the high temperature signal amplitude indicates a typical ZF relaxation in the slow dynamic limit. Due to the broad static linewidth ($\Delta B \geqslant 0.1$ T), the fast decay of polarization of the full signal is not observed. The 1/3-signal has been fitted using a pure exponential decay where the relaxation rate λ reflects the internal field fluctuation rate at the muon site. The monotonic decrease of the relaxation rate upon further cooling for all samples is due to the disappearance of spin excitations.

Characteristic changes of the spin dynamics between the M1 and M2 phases have not been observed. This indicates that the magnetic domains and spin structures of the M1 and M2 phases differ only on a length scale of several hundred nm as derived from neutron depolarization [3] but not on the μSR scale of less than one nanometer.

The sample with $x = 0.24$ Mn doping showed a different behaviour. The muon relaxation rate increases only slightly from 0.07 μs^{-1}at 300 K to 0.29 μs^{-1}at 200 K. The signal amplitude decreases rapidly between 200 K and 185 K. Below this temperature only $\approx 1/6$ of the initial amplitude can be resolved which has been fitted with a pure exponential relaxation. The relaxation rate decreases from $1.9(5)$ μs^{-1}at 150 K to $1.4(5)$ at 10 K. The antiferromagnetic transition (in Mössbauer experiments located at $\approx$ 130 K [3]) causes a very broad field distribution at the muon site in contrast to the ferromagnetic transition. This indicates a far more localized state of the 3d electrons compared to lower manganese doping.

TF μSR experiments have been performed on samples with $x = 0.0$, 0.113, 0.12 and 0.24. The undoped $Fe_{0.65}Ni_{0.35}$ shows a nearly constant relaxation rate of $0.30(3)$ μs^{-1} and frequency shift of $-1.1(2)$% in 0.1 T external field between 300 K and 20 K. The relaxation rates of the doped samples are shown in fig. 3. The TF experiments reveal a striking difference between two samples with only slightly differing manganese doping. In the sample with $x = 0.113$ no divergence of the relaxation rate as in the ZF experiments on the $x = 0.09$ and 0.12 samples is observed. Instead the relaxation rate levels off at ≈ 0.4 μs^{-1} below 150 K and the frequency shift in 0.5 T changes from zero at 300 K to $-0.6(1)$% below the ferromagnetic transition at $\approx$ 200 K. In the 0.5 T experiments on the sample with $x = 0.113$ the formation of a spin glass phase with transverse spin freezing is inhibited and the long range ordered ferromagnetic state is stabilized. The $x = 0.12$ sample shows an increase of the relaxation rate with the external field and a similar damping behaviour like the sample in the antiferromagnetic doping regime ($x = 0.24$). In these samples the disorder in the spin structures is enhanced by the external field. We understand this behaviour as a crossover from a mainly itinerant long range coupling of the 3d electrons at lower manganese concentrations to a localized coupling with short range order at higher doping.

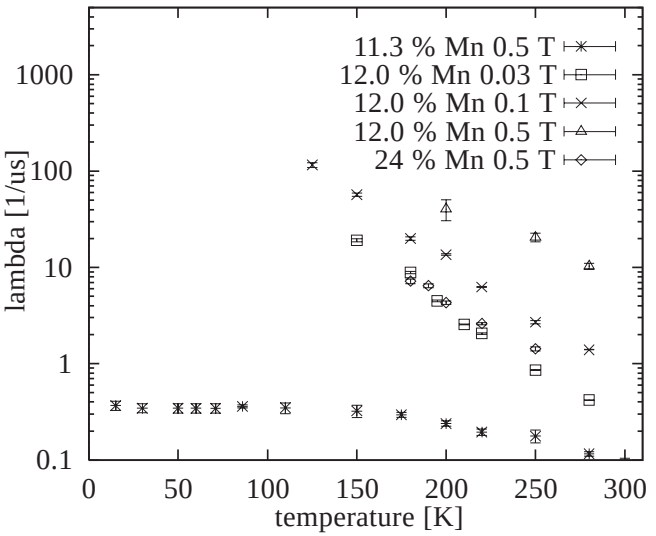

Fig. 3. Temperature dependence of the TF relaxation rates in $(Fe_{0.65}Ni_{0.35})_{1-x}Mn_x$ with $x = 0.113$, 0.12 and 0.24 in different external fields.

The present μSR studies on the FeNiMn reentrant spin glasses reveal significant differences between the spin dynamics in the low manganese doping (ferromagnetic) and the high doping (antiferromagnetic) regimes which could be understood in terms of a localization of the 3d electrons at higher doping. A universal behaviour of the dynamic relaxation function exponent has been found similar to studies on quite different systems with disordered spin freezing. A qualitative difference between the local spin dynamics in reentrant and ordinary spin glass phases has not been observed. In contrast to neutron depolarization, which reveals different degrees of spin freezing within the reentrant spin glass, μSR could not resolve these phases. Mössbauer spectroscopy is similarly insensitive. This indicates that these phases are of mesoscopic character.

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