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A neutron polarisation analysis study of the spin-glass phase of $Y(Al_{1-x}Fe_x)_2$

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Abstract. Diffuse neutron scattering with full polarisation analysis has been used to characterise the spin correlations in the extended spin-glass phase of the C15 Laves-phase compound, $Y(Al_{1-x}Fe_x)_2$, at concentrations of $x = 0.25$ and $x = 0.65$. Whilst the nuclear diffuse scattering indicates weak anticlustering of Fe atoms on the Al sublattice, the magnetic scattering can be interpreted as arising from small ferromagnetic clusters of less than 10 Å in diameter, with these continuing to exist at much higher temperatures than the spin-glass temperature. The ferromagnetic correlations which give rise to the clusters are short-range, and some evidence is found for weak antiferromagnetic correlations.

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The substitution of iron onto the tetrahedrally coordinated aluminium sites in the C15 Laves phase compound YAl_2 leads to an unusually extended spin glass-like regime and a greatly retarded percolation threshold for long range magnetic order at a concentration of approximately 75 at. % Fe [1]. There have been numerous attempts to model this rather anomalous magnetic phase diagram. One approach has invoked an itinerant electron rigid band model [2], within which it is assumed that each Al atom donates three electrons to the $3d$ sub-bands of YFe_2, whilst other approaches have used local environment models, suggesting that Fe atoms might possess either zero moment or a fully saturated moment (later shown to be incorrect [3]), depending upon the nearest neighbour configuration of Fe and Al atoms [4, 5]. However, neither of these models, nor later versions [6, 7], provide an adequate explanation of the experimental results and, despite several recent experimental studies, the magnetic properties of the pseudo-binary series $Y(Al_{1-x}Fe_x)_2$ are still not fully understood.

The experimentally determined magnetic phase diagram of $Y(Al_{1-x}Fe_x)_2$ is illustrated in Fig. 1. Recent Mössbauer spectroscopy and muon spin relaxation (μSR) measurements [8–10] have confirmed the existence of the spin glass-like or cluster-glass-like state which persists from $x = 0.25$ ($T_g = 4.6$ K) to $x = 0.75$ ($T_g = 50$ K). Above this concentration the compounds are generally regarded as ferromagnetic with a Curie temperature which rises rapidly with concentration to 540 K at $x = 1.0$. It should be noted, however, that band structure calculations [11, 12] and polarised neutron diffraction measurements [13] indicate a ferrimagnetic ground state for YFe_2 in which yttrium has been shown to possess a strongly delocalised moment of $-0.67\mu_B$ aligned antiparallel to the spatially localised iron moment of $1.77\mu_B$.

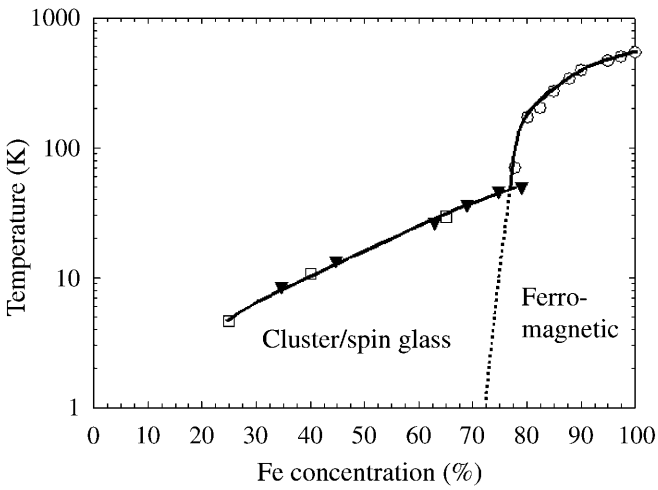

Fig. 1. The magnetic phase diagram of $Y(Al_{1-x}Fe_x)_2$ where T_g (*squares*) was determined by μSR [10] and T_g (*triangles*) and T_C (*circles*) were determined by dc magnetisation and Mössbauer spectroscopy effect measurements [8]

Our extensive susceptibility, Mössbauer spectroscopy effect and μSR measurements [8–10] have provided considerable evidence for extremely inhomogeneous magnetisation processes within the spin glass regime, with ferromagnetic correlations, or "clusters", persisting to temperatures well above the spin glass temperature, T_g. Whilst the spatial extent

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of the clusters appears to be relatively temperature independent to temperatures as high as $8\,T_g$ [14], there is evidence from our Mössbauer effect measurements and complementary muon spin relaxation measurements that the clusters are dynamic. Indeed, μSR measurements [10] suggest a broadly distributed cluster relaxation rate leading to Kohlrausch-like non-exponential relaxation with a mean relaxation time that increases critically as T_g is approached.

It is tempting to conclude that the inhomogeneous spin glass phase and the delay of the onset of ferromagnetic order is a consequence of topological frustration associated with weak antiferromagnetic correlations between the Fe spins superimposed upon the tetrahedral geometry of the Al sublattice. However, as yet there has been no direct evidence for such antiferromagnetic correlations. We have therefore carried out a series of diffuse neutron scattering measurements on $Y(Al_{1-x}Fe_x)_2$ with $x = 0.25$ and $x = 0.65$, using full neutron polarisation analysis to enable unambiguous separation of the magnetic and nuclear scattering cross sections in an attempt to probe the concentration and temperature dependence of the spin correlations in this intriguing system.

1 Neutron polarisation analysis measurements

The $Y(Al_{1-x}Fe_x)_2$ compounds, $x = 0.25$ and $x = 0.65$, were prepared by melting stoichiometric amounts of yttrium, aluminium and iron in an argon arc furnace (with extra Y for $x = 0.65$ to avoid forming YFe_2) and then annealing the Al-rich compound at 300 °C for 24 hours and the Fe-rich compound at 750 °C for 2 weeks. Ac susceptibility measurements were carried out on both samples from below T_g to room temperature, confirming the previously observed ferromagnetic correlations up to $\sim$ 200 K. The polarisation analysis measurements of the diffuse scattering were made using the D7 spectrometer at ILL, Grenoble.

Typical scattering cross-sections for the nuclear and magnetic diffuse scattering are shown in Fig. 2. These cross-sections were analysed using a modified reverse Monte Carlo procedure (RMC) [15] which initially distributes Fe atoms of an appropriate concentration over the Al sublattice of YAl_2, and then randomly exchanges Al and Fe atoms on the sublattice until an optimal fit to the nuclear cross section is achieved (see Fig. 2(a)). The resultant structural model enables the extraction of the Warren Cowley short-range order parameters, α_i, where $\alpha_i = (p_i^{BB} - c)/(1-c)$ with p_i^{BB} being the probability of finding a B atom at shell i from another B atom of concentration c. These parameters indicate a slight anti-clustering of Fe atoms for both $Y(Al_{1-x}Fe_x)_2$ compounds, with values of $\alpha_1 = -0.02 \pm 0.01$ and $\alpha_2 = -0.04 \pm 0.01$, with $\alpha_i = 0$ for third near neighbours and beyond.

The structural model produced by the RMC procedure is then used to analyse the magnetic cross section in a similar way. Magnetic (Heisenberg) spins are placed at the Fe sites and their directions and magnitudes varied until a fit to the magnetic cross-section is achieved (Fig. 2(b)). It should be noted that all the Fe spins are of identical magnitude, but that this magnitude is varied in the RMC procedure. The resulting magnetic model enables $S(S+1)$ and the normalised radially dependent magnetic spin correlations $\langle \underline{S}_0 . \underline{S}_i \rangle / S(S+1)$ to be extracted for both compounds. The results are shown in Table 1 and Fig. 3 for both compounds at several

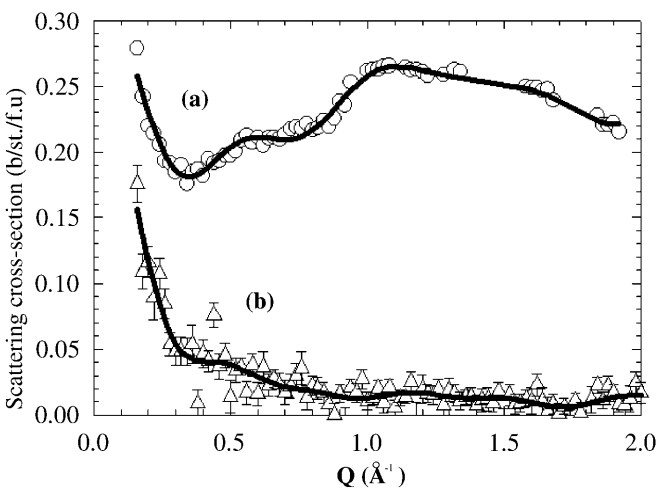

Fig. 2. The nuclear (a) and magnetic (b) scattering cross-sections of $Y(Al_{0.75}Fe_{0.25})_2$ at 300 K, together with fits resulting from the Monte Carlo procedure

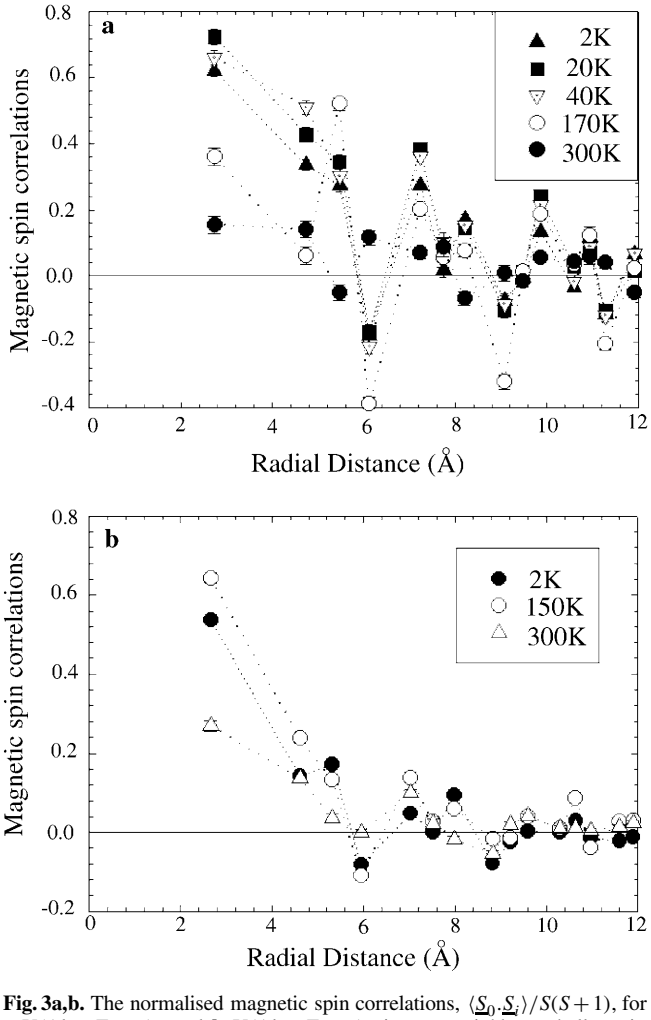

Fig. 3a,b. The normalised magnetic spin correlations, $\langle \underline{S}_0 . \underline{S}_i \rangle / S(S+1)$, for **a** $Y(Al_{0.75}Fe_{0.25})_2$ and **b** $Y(Al_{0.35}Fe_{0.65})_2$ in near neighbour shells at increasing radial distance. (The lattice parameter $a = 7.7426$ Å and 7.526 Å respectively.)

Table 1. Values of $S(S+1)$ estimated from the magnetic diffuse scattering cross section measured at several temperatures

Temperature (K)	$Y(Al_{0.75}Fe_{0.25})_2$	$Y(Al_{0.35}Fe_{0.65})_2$
2	0.06 ± 0.03	0.33 ± 0.05
20	0.05 ± 0.02	–
40	0.04 ± 0.01	–
150	–	0.19 ± 0.01
170	0.07 ± 0.02	–
300	0.09 ± 0.01	0.23 ± 0.04

temperatures. (The uncertainties in the respective parameters are estimated by averaging over the RMC generated configuration.) It can be seen that the self correlation parameter $S(S+1)$ is largely independent of temperature, taking a mean value of 0.06 for the $x = 0.25$ compound and 0.25 for the $x = 0.65$ compound. From Fig. 3 it is apparent that at short radial distances ($R < 5$ Å) the spin correlations are predominantly ferromagnetic. Whilst there is some evidence of a modest antiferromagnetic component to the spin correlations at and beyond 6 Å, the spin correlations are rapidly damped to zero at large distances. It should also be noted that the spin correlations decrease at the highest temperatures, particularly for the $x = 0.25$ compound.

2 Discussion

The magnetic cross section of the $Y(Al_{1-x}Fe_x)_2$ compounds show clear evidence of ferromagnetic correlations which lead to small clusters of approximately 5 Å radius. These clusters vary little in extent with concentration between $x = 0.25$ and $x = 0.65$, and show relatively weak temperature dependence to temperatures well above T_g. The spin correlations also provide indications of weak antiferromagnetic correlations at longer distances. In analysing the magnetic diffuse scattering, our model has assumed that all the Fe spins for a particular alloy are of the same magnitude. Within this constraint, we obtain values for the effective Fe moments of $1.20\,\mu_B$ and $1.74\,\mu_B$ for $x = 0.25$ and $x = 0.65$ respectively at 2 K. Integrating over the extended Fe correlations shown in Fig. 3, we estimate effective cluster moments of $\sim 4.5\,\mu_B$ and $\sim 9.6\,\mu_B$ for the $x = 0.25$ and $x = 0.65$ compounds respectively, again at 2 K. The values for both the effective Fe moments and effective cluster moments compare favourably with those determined by Mössbauer spectroscopy measurements and reported in the literature [8, 14].

3 Conclusions

Our neutron diffuse scattering measurements with polarization analysis indicate that the extended spin glass phase of $Y(Al_{1-x}Fe_x)_2$ is characterised by relatively small magnetic clusters formed by ferromagnetic Fe spin correlations which extend to approximately 5 Å. The mean effective Fe moment, and correspondingly the effective cluster moment, increase between $x = 0.25$ and $x = 0.65$, although the normalised pair correlations are similar for both concentrations. The spin correlations persist to temperatures well above T_g, although with decreasing magnitude. The weak antiferromagnetic correlations at distances greater that 6 Å, cannot alone account for the anomalous extended spin glass phase of $Y(Al_{1-x}Fe_x)_2$.To understand fully the nature of the magnetic correlations in $Y(Al_{1-x}Fe_x)_2$ it may be necessary to develop a more complex, and possibly dynamic, model of extended magnetic defects on the Fe sites associated with the substitution of Al atoms, a task that will be facilitated by our forthcoming polarized neutron studies of magnetic defects associated with relatively dilute (< 5 at. %) Al substitution for Fe in ferrimagnetic YFe_2.

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