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Magnetic field dependence of the spin dynamics in $Fe_{28}Cr_{72}$

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Abstract. The field dependence of the magnetic excitations in the re-entrant spin glass (RSG) $Fe_{28}Cr_{72}$ is investigated over a wide range of temperatures. The zero-field spin dynamics are anomalous not only in the spin-glass phase but also in the paramagnetic phase. In an applied field ($H = 8.8$ kOe) spin waves are induced at low temperatures with an energy gap $g\mu_B H \simeq 0.1$ meV. This observation is in marked contrast with results from $Fe_{1-x}Al_x$, where no gap is induced. The results suggest that the ground state of RSG is not unique.

1. Introduction

In the wide field of diluted magnetic systems, the problem of re-entrant spin glasses (RSGs) has proven difficult to solve, both experimentally and theoretically [1]. Despite nearly universal behaviour of the measured AC susceptibility, the spin dynamics appear to be system specific and it remains difficult to classify the various materials. Table 1 gives a partial listing of various RSG and spin-glass (SG) systems and a summary of the observed features. All these systems show spin waves appearing just below the onset of ferromagnetic-like ordering at $T = T_C$. The magnetic stiffness D associated with these spin waves increases with decreasing temperature T and then decreases again at lower T. It is in this low-temperature regime, the RSG phase, that the behaviour varies from system to system. For example in Ni_xMn_{1-x} [2] and amorphous Fe_xMn_{1-x} [4], D decreases and then increases again at lower T. $Fe_{1-x}Al_x$ [5–7] and amorphous Fe_xNi_{1-x} show no well-defined spin waves at lower T. In Fe_xCr_{1-x} [8] it was originally implied that D vanished at low T, but a recent reexamination [9] suggests that D may level off to a finite value.

Experimentally it is very difficult to determine the nature of the excitation at low T. One reason is that the intensity of the inelastic scattering is weak because of the thermal population factor and that the spectral response is dominated by the resolution-limited central peak. In addition the T-dependence of D frequently depends upon the particular choice of lineshapes used to analyse and deconvolve the spectra.

Hennion and co-workers [4] suggested that one possible way to separate out the inelastic scattering from the central peak is the application of a magnetic field which induces a gap $\Delta = g\mu_B H$ in the magnetic scattering. Under the assumption that the spin-glass state is not affected by H it is indeed possible to measure the stiffness D of the spin

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Table 1. Summary of typical properties of RSGs in an applied magnetic field.

Sample	System	Elastic peak observed at $Q \neq 0, H \neq 0$	Presence of spin wave	References
$Ni_{0.784}Mn_{0.216}$	RSG	Yes	Yes	[2, 3]
$(Fe_{0.765}Mn_{0.235})_{75}P_{16}B_6Al_3$	RSG	Yes	Yes	[3, 4]
$Fe_{70.4}Al_{29.6}$	RSG	Yes	No	[5]
$Fe_{69.9}Al_{30.1}$	RSG	Yes	No	[6]
$Fe_{68.5}Al_{31.5}$	SG	Shoulder	No	[6]
$Fe_{28}Cr_{72}$	RSG	No	Yes	Present work
$Fe_{0.65}(Ni_{0.866}Mn_{0.134})_{0.35}$	RSG	Yes	Yes	[6]

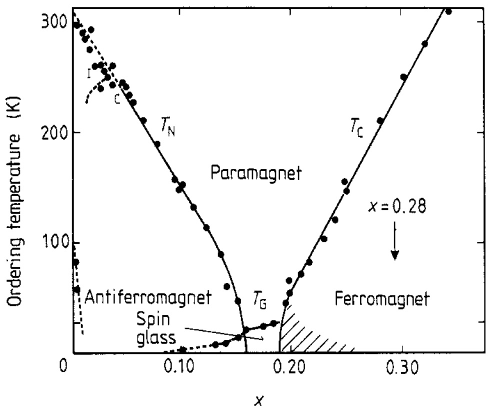

Figure 1. Temperature–concentration phase diagram or Fe_xCr_{1-x}. The shaded area indicates the re-entrant phase. After Burke and co-workers [12].

waves unambiguously after subtraction of the induced gap. If a dispersion is induced i.e. $D > 0$ spin waves are considered to persist in the spin-glass phase even in zero field.

There is, however, strong evidence from neutron depolarisation measurements [10, 11] that fields of a few Oerstedt affect the spin glass state in $Fe_{1-x}Al_x$ dramatically. Moreover it was shown very recently [5], that no spin waves are induced in $Fe_{70.4}Al_{29.6}$ in applied fields up to 10 kOe, although the sample was close to saturation. This latter result is in contrast with the early experiments on Fe_xCr_{1-x} ($x = 0.26$) where spin waves are induced in the spin-glass phase [8]. In this work we reexamine another alloy of Fe_xCr_{1-x} ($x = 72$) with particular emphasis on the magnetic field dependence of the excitations.

2. Experimental

The measurements were performed on the same spherically shaped polycrystalline sample of $Fe_{28}Cr_{72}$ used in the previous studies by Shapiro and co-workers [8]. The diffuse scattering measurements showed that the samples exhibited some chemical short-range order. A temperature–concentration phase diagram for Fe_xCr_{1-x} [12] in the Fe-poor region is shown in figure 1. The present sample with $x = 28$ is very close to being a ferromagnet even at very low temperatures and was not studied in any detail in the earlier work.

For the neutron scattering experiments we used the triple axis spectrometer H9, which is situated at the cold source at the High Flux Beam Reactor and is equipped with a double monochromator system. The final neutron energy E_f was kept fixed at 3 meV and the collimations were 30′ 40′ 30′ before and 20′ 40′ after the sample. The sample was mounted between the pole pieces of an electromagnet with the field direction parallel to $\boldsymbol{Q}$ (horizontal) for $E = 0$. Higher order neutrons were removed by means of a cooled Be filter. The energy resolution is 0.042 meV (FWHM). The measurements were conducted near the forward direction mostly with $0.025\ \text{Å}^{-1} \leq q \leq 0.06\ \text{Å}^{-1}$.

3. Results

In the earlier work it was not clear whether Fe_xCr_{1-x} with $x = 0.28$ entered a spin-glass state at low temperature, because the measurements were only extended down to 15 K and the energy resolution was relatively coarse. As a first step we confirmed that our sample indeed entered a spin-glass state at very low T. Figure 2 gives a summary of representative scans taken for a fixed momentum transfer of $q = 0.04\ \text{Å}^{-1}$. It can be clearly seen that the spin-wave energy decreases in energy with decreasing T, as opposed to a levelling off in energy expected in a ferromagnet. Also a central peak at $E = 0$ develops on cooling. These characteristics are typical of RSG behaviour. In order to put out data on a more quantitative basis we fit the data with the following cross section (convolved with the instrumental resolution function)

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E} = \{CE/[1-\exp(-E/k_B T)]\}F(q, E)/q^2 + (B/q^2)\delta(E). \tag{1}$$

The first term represents inelastic or quasi-elastic magnetic scattering with C containing trivial constants and $F(q, E)$ being a normalised spectral weight function. The second term with the normalisation constant B represents a resolution-limited central component which includes incoherent scattering from the sample and sample container as well as the magnetic scattering which develops at low T. For convenience we choose a Lorentzian form of $F(q, E)$

$$F(q, E) = (1/2\pi)[\![\{\Gamma/[(E-E_q)^2+\Gamma^2]\}+\{\Gamma/[(E+E_q)^2+\Gamma^2]\}]\!] \tag{2}$$

where Γ is the width and E_q is the peak position of the spin wave. At small q the dispersion is given by

$$E_q = \Delta_0 + g\mu_B H + Dq^2 \tag{3}$$

where Δ_0 is a crystal anisotropy gap, D is the magnetic stiffness and g the Landé g-factor ($g = 2$ for Fe). During the course of fitting the data to the above cross section we found that the linewidth Γ was smaller than the energy resolution of the spectrometer. Therefore we fixed Γ at 0.01 meV in the final fitting procedure. Figure 3 shows the temperature dependence of E_q, C and B for $q = 0.04\ \text{Å}^{-1}$. The spin-wave energy exhibits the characteristic behaviour of an RSG for $H = 0$: E_q decreases near T_C and again at low temperatures. C increases by 25% with decreasing temperature mostly because of experimental uncertainties, since the spin wave intensities become very small compared with the nonmagnetic contributions and the central peak. At T_C C increases by exactly 50% because of the contributions of the evolving longitudinal fluctuations as expected for a normal ferromagnet. On the other hand B increases by about a factor of 2 when

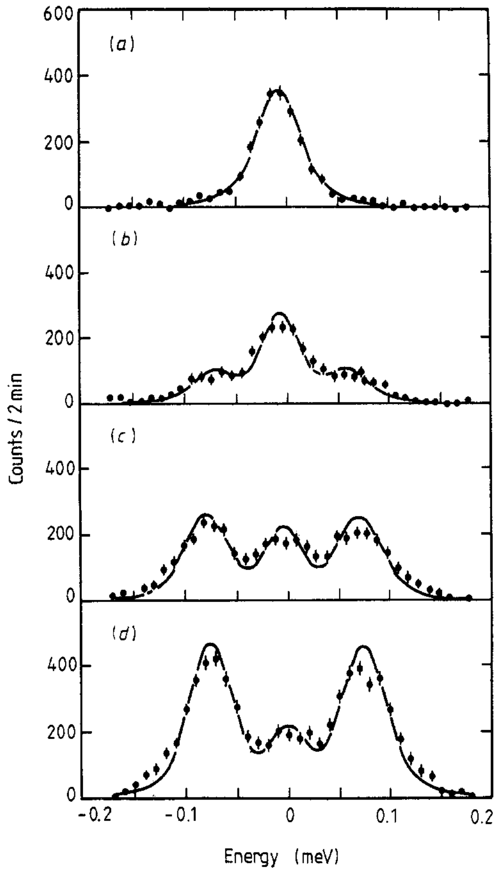

Figure 2. Temperature dependence of zero-field spectra of $Fe_{28}Cr_{72}$ measured at $q = 0.04$ Å^{-1}. The scattering at $E = 0$ increases by almost a factor of 2 with decreasing T. (*a*) $T = 5$ K; (b) $T = 15$ K; (*c*) $T = 40$ K; (*d*) $T = 80$ K.

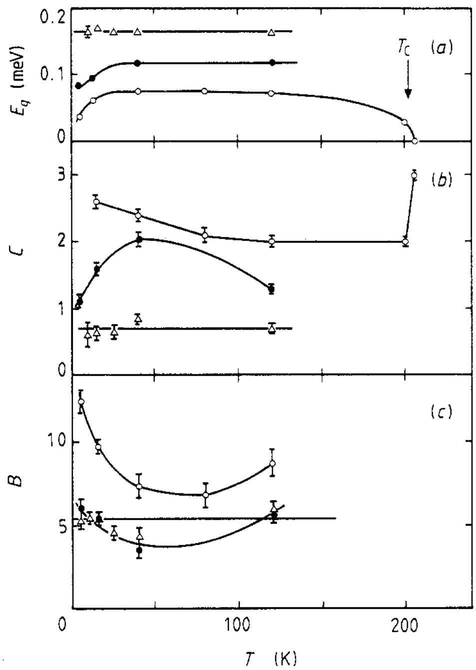

Figure 3. Summary of the temperature and field dependence of (*a*) the spin wave energy E_q, (*b*) the inelastic intensity C and (*c*) the central component B of $Fe_{28}Cr_{72}$ for $q = 0.04$ Å^{-1}. ○, $H = 0$ kOe; ●, $H = 5$ kOe; △, $H = 8.8$ kOe. The full curves are guides to the eye. The error bars indicate the statistical errors and are omitted when they were smaller than the size of the symbols.

the temperature reaches 5 K. Such an increase is common in RSGs. The above results suggest that $Fe_{28}Cr_{72}$ can be classified as a typical RSG.

As a next step we applied a horizontal field H of 5 and 8.8 kOe in order to measure the dispersion of the induced spin waves at low T. Figures 4 and 5 show some representative spectra measured at $q = 0.04$ Å^{-1}. In $H = 5$ kOe, the spin waves are shifted with respect to the zero field data by 0.045 meV, which is smaller than $g\mu_B H = 0.058$ meV because of the energy gap $\Delta_0 \simeq 0.01$ meV ([8] and present work) in zero field. In 8.8 kOe, however, the spin-wave energy is nearly independent of T below 120 K. We conclude that $Fe_{28}Cr_{72}$ behaves like a normal ferromagnet in large fields, because the spin waves do not soften at low T and the amplitude for the magnetic scattering C is independent of T within the experimental uncertainty.

In number of RSGs it was found that some Q dependent structure develops in the elastic intensity in a magnetic field [3, 13–15]. Therefore we also performed some Q-scans at $E = 0$ in order to find out if $I(Q, 0)$ exhibits a peak at finite Q under an applied

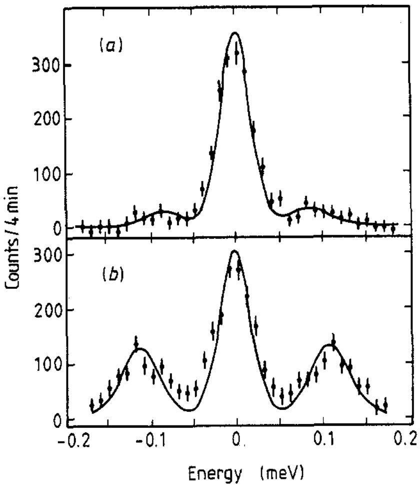

Figure 4. Some representative scans of $Fe_{28}Cr_{72}$ measured at $H = 5$ kOe: (*a*) $T = 5$ K; (*b*) $T = 15$ K. Final neutron energy $E_f = 3$ meV; beam collimations 30′ 40′ 30′ before and 20′ 40′ after the sample.

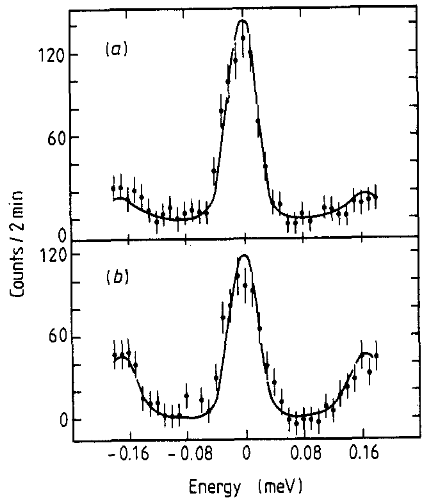

Figure 5. Some representative scans of $Fe_{28}Cr_{72}$ measured at $H = 8.8$ kOe: (*a*) $T = 10$ K; (*b*) $T = 25$ K. E_f and beam collimations as for figure 4.

field H. With increasing H the small angle scattering decreased monotonically implying that the moments become aligned and contribute to the (0 0 0) Bragg peak. Hence low temperature effects caused by chemical order can be ruled out. In contrast $Fe_{1-x}Al_x$ and some Ni_xMn_{1-x} and amorphous Fe–Mn alloys, a peak was not visible in our raw data for $q = 0.03$ Å^{-1}. However, it was demonstrated recently for $Fe_{28}Cr_{72}$ that a more sophisticated subtraction of the tails of the inelastic scattering at $E = 0$ can yield a broad bump at finite Q [14].

In addition to the measurements at low T we also studied the field dependence of the magnetic scattering at T_C. In zero field the scattering is diffuse and the q-dependence of the linewidth can be parametrised by $\Gamma^C = Aq^z$ with $z = 1.76 \pm 0.3$ and $A \simeq 7.7$ meV Å$^{1.76}$. The value of z is significantly smaller than for a normal localised ferromagnet like EuO [16] where $z = 2.5$. It is, however, within error equal to the value found recently in the invar alloy $Fe_{65}Ni_{35}$ ($z = 2$) [17]. The latter value was interpreted in terms of scattering of spin fluctuations by impurities [18].

Another interesting feature of the magnetic scattering from Fe_xCr_{1-x} are the energy scales involved. They are similar in the ordered and disordered phase for isotropic ferromagnets like BCC Fe. Although the exchange is significantly reduced in $Fe_{28}Cr_{72}$ with respect to Fe ($D_{Fe}/D_{Fe_{28}Cr_{72}} \simeq 12$) the linewidths of the paramagnetic scattering are comparable (at $q = 0.05$ Å^{-1}: $\Gamma^C_{Fe}/\Gamma^C_{Fe_{28}Cr_{72}} \simeq 2$). In other words $\Gamma^C_{Fe_{28}Cr_{72}}$ is very large at T_C and cannot be explained solely on the basis of the spin dynamics below T_C (i.e $D_{Fe_{28}Cr_{72}}$), which are a measure of the magnitude of the exchange interactions. By the same reasons dipolar interactions can be ruled out too. Therefore the large Γ of $Fe_{28}Cr_{72}$ at small q is a signature for the scattering of the spin fluctuations by the inhomogeneities. Under application of a field ($H = 8.8$ kOe) a ferromagnetic-like state is induced at $T = T_C$ with a gap of 0.12 ± 0.02 meV and $D = 15 \pm 5$ meV Å^2, the latter value being a factor of 2 smaller than the maximum value of D in the ferromagnetic phase [8].

4. Discussion

The present results show that the RSG state of Fe_xCr_{1-x} differs significantly from the RSG state in $Fe_{1-x}Al_x$. In the former system spin waves are induced in the spin-glass phase and in the latter a peak in $I(Q, 0)$ appears in a magnetic field. In table 1 (§ 1), we give a summary of similar studies on other RSG samples. Apparently we can divide the systems in at least three categories on the basis of the appearance of spin wave peaks and (or) peaks at finite Q, suggesting that no unique ground state exists in RSGs. The Mn-containing compounds have a peak at finite q and spin waves are induced in a field. In $Fe_{1-x}Al_x$ the peak in $I(Q, 0)$ is most pronounced for higher Fe concentration, i.e. when the frustration is reduced, however, no measurable spin waves can be induced. On the other hand in $Fe_{28}Cr_{72}$ we observe clearly ferromagnetic spin waves under an applied field in the RSG phase for an Fe concentration close to the ferromagnetic border (see figure 1). Therefore stronger ferromagnetic correlations do not guarantee the occurrence of a peak at finite q in the RSG phase of $Fe_{28}Cr_{72}$. It would be of great value to extend the present measurements in Fe_xCr_{1-x} to lower Fe concentrations in order to measure the influence of the ferromagnetic correlations in detail.

Another interesting aspect of our measurements is the field dependence of the spin-wave energy. Although our present data are not entirely conclusive, it appears that no full long-range order develops in a field of 5 kOe (figure 3), in contrast to $H = 8.8$ kOe. The observed decrease of E_q for $q = 0.04$ Å^{-1} and $T = 10$ K may indicate that the correlation length does not exceed $2\pi/0.04$ Å^{-1} = 150 Å^{-1} below 10 K.

Obviously the application of a field changes the ground state properties of the RSG phase significantly. At intermediate T, E_q is just proportional to $g\mu_B H$ as expected for a Zeeman splitting of a simple Heisenberg ferromagnet. At low temperatures, however, a ferromagnetic-like state is induced for sufficiently large fields and E_q becomes independent of T. Therefore the dynamics in the ground state are affected significantly by the application of a magnetic field.

Acknowledgments

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