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Neutron depolarization measurements on the reentrant spin-glass bcc Cr–Fe–Mn alloy

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Abstract. Neutron depolarization and diffraction measurements were performed on bcc $Cr_{60}Fe_{20}Mn_{20}$ with the aim of studying the low temperature reentrant spin-glass phase. This alloy is reported to undergo successive magnetic transitions as the temperature is lowered, *viz.*, antiferromagnetic, ferromagnetic and spin glass. Transmitted neutron polarization was measured as a function of temperature for zero-field-cooled (ZFC) and field-cooled (FC) cases. The transition to a ferromagnetic phase is indicated by a decrease in the polarization at $T_C \sim 100$ K. It exhibits a minimum at 55 K below which it increases and at 14 K, in the spin-glass phase, for the ZFC case it recovers to the paramagnetic value. Large irreversibility and time dependent effects are also observed in the spin-glass phase. These results indicate that the ferromagnetic order which grows below T_C is destroyed in the spin-glass phase at 14 K. Neutron diffraction measurements indicate the presence of an antiferromagnetic (100) peak below $T_N \sim 290$ K. The intensity of the (100) peak increases as the temperature is lowered and persists down to 15 K. The low temperature phase is, therefore, a mixed phase where spin-glass and antiferromagnetic order coexist.

1. Introduction

The magnetic properties of bcc Cr–Fe alloys have been of interest for both experimental and theoretical studies. The $Cr_{1-x}Fe_x$ alloys with $x > 19$ at.% are found to exhibit a ferromagnet to reentrant spin-glass transition [1]. Inelastic neutron scattering measurements on several of these alloys confirm the existence of ferromagnetic (FM) to spin-glass (SG) crossover [2]. Spin-wave excitations are found within the ferromagnetic phase. The spin wave frequency decreases as the spin-glass phase is approached. Within the spin-glass phase no well defined excitations are found. However on application of field the excitations reappear. Addition of Mn to Cr stabilizes the antiferromagnetic phase over a wide concentration range of Mn. Recently a number of studies have been reported on ternary Cr–Fe–Mn alloys where Cr has been partially substituted by Mn [3–5]. Based upon bulk magnetization, transport and Mossbauer experiments, a magnetic phase diagram has been obtained for $Cr_{80-x}Fe_{20}Mn_x$ ($3.9 \leqslant x \leqslant 39$) [3] which shows that the compositions with $3.9 \leqslant x \leqslant 39$ undergo successive magnetic transitions, namely, antiferromagnetic (AF), ferromagnetic (FM) and spin-glass (SG) order as the temperature is reduced. Neutron scattering studies on one of the compositions ($x = 15$)

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show that the magnetic peak (100) develops below $T_N \sim 250$ K [4]. It increases in intensity as the temperature is lowered and becomes constant below 50 K. Magnetization measurements made on the same sample show ferromagnetism below 100 K and spin-glass behaviour below 40 K. Since the antiferromagnetic (100) line remained in the range of temperature where ferromagnetism and spin glass exist it was inferred that at low temperatures FM and SG phases coexist with AFM phase [4]. However, it is not clear from these studies whether the ferromagnetic order which grows below T_C coexists with the spin-glass phase. The study of ferromagnetic domains using the neutron depolarization technique was initiated by Halperin and Holstein [6]. A general introduction to the application of neutron depolarization technique in the study of reentrant spin glasses has been given by Mitsuda *et al* [7]. We have used this sensitive technique to study the nature of the reentrant spin glass phase on a representative alloy, $Cr_{60}Fe_{20}Mn_{20}$, of the series. It is reported to have three transitions, $T_N \sim 270$ K, $T_C \sim 140$ K and $T_G \sim 50$ K [3].

2. Experimental details

The polycrystalline sample was prepared as reported earlier [3]. For depolarization measurements a pellet of dimensions approximately $2 \times 8 \times 10$ mm^3 was used. The one dimensional depolarization measurements were performed with $Cu_2MnAl(111)$ as the polarizer and $Co_{0.92}Fe_{0.08}$ (200) as the analyser using the polarization analysis spectrometer at Dhruva reactor, Trombay. A detailed description of the spectrometer is given in [8]. An rf flipper was used to flip the incident beam polarization. The sample was attached to the cold head of a close cycle refrigerator (CCR). Beam size was restricted to 5×5 mm^2 to be within the sample.

The following procedure was adopted to measure the zero-field-cooled (ZFC) and field-cooled (FC) depolarization measurements. For ZFC measurements the sample was cooled from 300 to 14 K with magnetic field on the sample set to zero. At 14 K the field was increased to 50 G. The flipping ratio of the intensities for the two spin states of incident neutron beam polarization was recorded in the warming cycle after stabilizing the temperature at each point. In the case of FC measurement the sample was cooled from 300 down to 14 K in the presence of magnetic field, $H = 50$ G and the flipping ratio was recorded in the warming cycle as for ZFC measurements.

For relaxation measurements the sample was cooled in $H = 1.2$ kG from 300 K to the respective measuring temperature, i.e., 14 and 60 K. The field was then reduced to 50 G and the transmitted intensity recorded as a function of time. The polarizer and analyser were used in the crossed polarized mode. The rf flipper was off. In this configuration a large transmitted intensity corresponds to large depolarization.

The diffraction patterns were recorded on the PSD based profile analysis spectrometer ($\lambda = 1.094$ Å) and high-Q diffractometer ($\lambda = 1.278$ Å) on a button shaped sample between 300 and 15 K in a CCR. The low field magnetization was measured using a SQUID magnetometer.

3. Results and discussion

Neutron depolarization measurements provide useful information on the magnetic inhomogeneity in a length scale >1 μm. In the paramagnetic state the temporal spin fluctuations are too fast for neutron polarization to follow the variation of the magnetic field acting on the moving neutron. Therefore, no depolarization is observed. In a classical

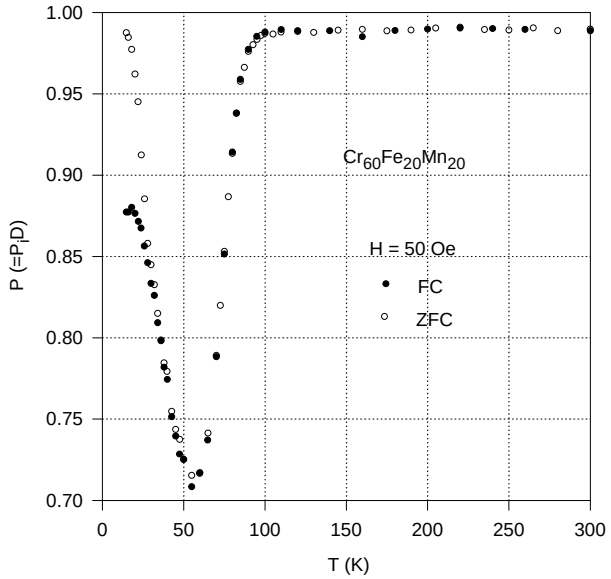

Figure 1. Temperature dependence of the transmitted neutron polarization, P for zero-field-cooled (○) and field-cooled (●) cases in $H = 50$ G.

spin-glass state each spin is frozen with its direction pointing in a random direction. The spatial fluctuation of the effective field due to the random orientations of the spins does not affect the neutron polarization and hence no depolarization is observed. Experimentally, no depolarization is observed in systems exhibiting pure spin-glass state, namely, amorphous Fe–Mn [9], Au–Fe [10], amorphous Fe–Ru–Zr [11] etc. In antiferromagnets the net magnetization is zero and no depolarization is expected. In traversing uniformly magnetized domains, as in a ferromagnet, the neutron spin experiences a torque as a result of which it precesses about the magnetization direction. In the presence of a number of such domains oriented randomly (if the precession angle is $<2\pi$ in each domain) the incident neutron undergoes depolarization.

In this study we have measured the flipping ratio R (ratio of the transmitted intensities for two spin states of the incident neutron spin) which is a measure of the transmitted beam polarization. R for the particular configuration where we have used Cu_2MnAl as polarizer and $Co_{0.98}Fe_{0.02}$ as analyser is expressed in the form [8]

$$R = \frac{1 - P_i D P_A}{1 + (2f - 1) P_i D P_A}$$

where P_i is the incident beam polarization, P_A is the efficiency of the analyser crystal, f is the rf flipper efficiency and D is the depolarization coefficient. In the absence of any depolarization in the sample $D = 1$. $P_i D$ is thus the transmitted beam polarization.

Figure 1 shows the temperature dependence of the transmitted neutron beam polarization ($P = P_i D$) in a field of 50 G, for ZFC and FC cases. Between 300 and 100 K polarization remains constant within experimental error. It decreases rapidly below 100 K and reaches a minimum at 55 K. Below 55 K polarization again increases and at 14 K in the ZFC case it recovers to the paramagnetic value. For the FC case it recovers partially. A pronounced difference between ZFC and FC polarization is observed below 30 K.

The variation of the low-field dc magnetization ($H_{ext} = 50$ G) with temperature for ZFC and FC states is shown in figure 2. The magnetization increases below 140 K and peaks at

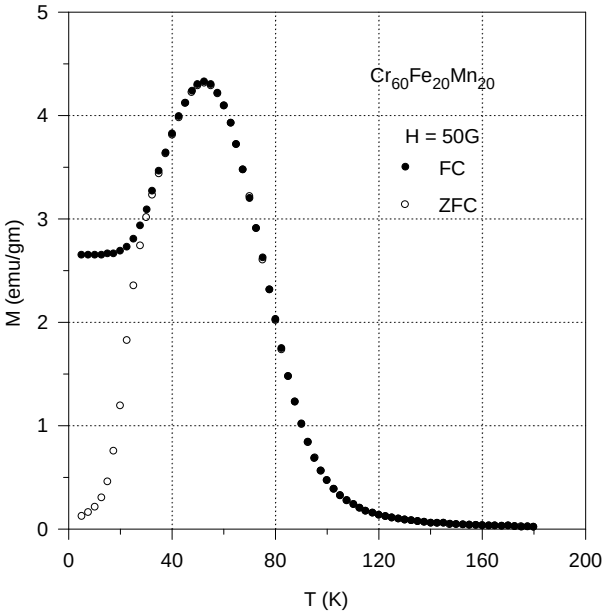

Figure 2. Temperature dependence of magnetization for ZFC and FC cases in $H = 50$ G.

$T_G = 55$ K. In the FC state magnetization departs significantly from the ZFC state below 30 K. It may be noted that though the magnetization in the ZFC state approaches zero as $T \to 0$, the spontaneous magnetization obtained from the Arrott plot (figure 3 in [3]) does not show any anomalous behaviour below 55 K. It increases below 140 K and remains nearly constant below 60 K.

The depolarization measurements are fairly consistent with the magnetization behaviour. The only difference we find with the magnetization data is that the depolarization starts decreasing only at 100 K. In this experiment T_C is identified with the onset of depolarization, in which case we find that T_C ($\sim$100 K) is lower than the value reported from magnetization experiments ($\sim$140 K). The T_C in the magnetization experiments is associated with the appearance of non-zero spontaneous magnetization (M_S) in the Arrott plot. Because the low-field region in the Arrott plot is non-linear, M_S is determined by extrapolation from high field. The difference in the value of T_C obtained, therefore, corresponds to different field regimes of measurement. In the region $55 < T < 100$ K polarization continuously decreases where it exhibits ferromagnetic behaviour. The temperature at which a minimum in the polarization occurs coincides with the maximum in the ZFC magnetization data. Below 55 K the increase in the polarization indicates the onset of breakdown of ferromagnetic ordering. At 14 K the total recovery of polarization to the paramagnetic value is a surprising result. Magnetization measurements do not provide any further clue except for the observed drop in magnetization which is identical in all RSG systems. Unfortunately due to the limitation of the experimental setup the temperature could not be lowered any further. The observation that the polarization in the ZFC condition at 14 K is the same as that in the paramagnetic state is a definite indication that there exists no ferromagnetic order over the mesoscopic length scale. A similar observation of complete recovery of polarization in the reentrant spin-glass phase has been made in a-$(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ [12], $Fe_{17.65}Ru_{2.35}Zr_{10}$ [13] and a-$(Fe_{0.3}Mn_{0.7})_{75}P_{16}B_6Al_3$ [9]. A common feature in these results is that the reported compositions are very close to the

critical concentration beyond which ferromagnetic order disappears. In the present study the composition is sufficiently away from the critical concentration $Cr_{41.6}Fe_{20}Mn_{38.4}$ [3]. The recovery of polarization has been attributed to either the shrinkage of ferromagnetic domain size or decrease in spontaneous magnetization or both [9, 13]. In the present sample the spontaneous magnetization does not decrease for $T < 55$ K. Therefore the observed increase of polarization below 55 K could only be due to a gradual decrease of domain size. An estimate of the domain size in the ferromagnetic region is obtained using the expression [6, 12]

$$P_f = P_i \exp(-\alpha(d/\delta))\langle\phi_\delta\rangle^2$$

where P_f and P_i are the transmitted beam and incident beam polarization, respectively, α is a dimensionless parameter $= 1/3$, d is the sample thickness, δ is a typical domain length and the precession angle $\phi_\delta = (4.63 \times 10^{-10}\ \mathrm{Oe}^{-1}\ \mathrm{A}^{-2})\ \lambda\delta B$. The domain magnetization, B is obtained from the bulk magnetization. This expression is valid in the limit where the domains are randomly oriented and the Larmor phase of the neutron spin due to the internal magnetic field of the sample $<2\pi$ over a typical domain length scale. Our measurements were carried out in low field far away from the saturation field and therefore satisfies the assumption of the model. The estimated domain size in the present sample at $T = 55$ K, where the depolarization is maximum, is $\sim 2\ \mu$m. The size of the domains obtained is in fair agreement with the estimated domain sizes in a-$(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ [12], a-$Fe_{90+x}Zr_{10-x}$ [14] and a-Fe–Mn [8] alloys. However, a better quantitative estimate of domain size can be obtained by measuring the wavelength dependence of polarization, $P(\lambda)$.

In the present alloy, the occurrence of a maximum in the ZFC magnetization at 55 K and the irreversible behaviour is associated with the spin-glass behaviour [3]. Depolarization measurements exhibit similar behaviour. No irreversibility appears above 55 K. The difference between ZFC and FC measurements appears below 55 K and becomes significant below 30 K, though at present we do not associate strong and weak irreversibility regions. The depolarization is larger in FC than in ZFC as is observed in other systems [16]. When the sample is cooled in a field domains/clusters of finite dimension grow in size. Since the measuring field is low ($\sim$50 G) the domains are randomly oriented due to pinning of domain walls. This results in a larger depolarization in the FC case.

Magnetic relaxation on a macroscopic time scale is one signature of a spin glass [17, 18]. Recently neutron depolarization measurements have been found to exhibit relaxation behaviour in the spin-glass phase of amorphous Fe–Mn alloys [9] and a-$Fe_{85}Ru_5Zr_{10}$ [11]. We have performed similar time dependent depolarization measurements on our sample as follows. In two different measurements the sample was cooled to 14 and 60 K under a field of 1.2 kG. The field was then reduced to 50 G and the transmitted beam intensity was measured as a function of time with polarizer and analyser in crossed mode. In figure 3 we show the measured intensity as a function of time at 14 and 60 K. There exists a strong relaxation at 14 K and practically no relaxation at 60 K. Since polarization measurements follow magnetization behaviour relaxation in the field cooled state is associated with the relaxation of the domains/clusters induced by the field. The time dependence of magnetization in various magnetic phases with differing relaxation functions has been reported [19]. We find that the field-cooled transmitted intensity at 14 K follows a logarithmic time dependence. The inset of figure 3 shows the semilog plot of intensity versus time and the continuous line is the linear fit to the data. Thermoremanent magnetization in spin glasses has been reported to obey a $\ln T$ dependence over a long time [20, 21]. The $\ln T$ dependence is shown to arise in an ensemble of noninteracting particles with moment and having a distribution of energy barriers to moment rotation. Magnetic relaxation in various phases including antiferromagnets has been found. Since the present sample appears to be magnetically complex and the relaxation behaviour is governed by too many parameters,

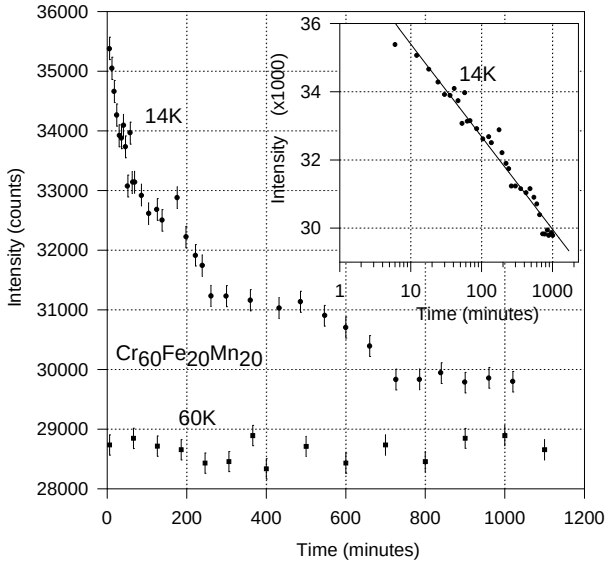

Figure 3. Time dependence of the transmitted intensity at 14 and 60 K. The inset shows the intensity as a function of $\log t$. The continuous line is a linear fit to the data.

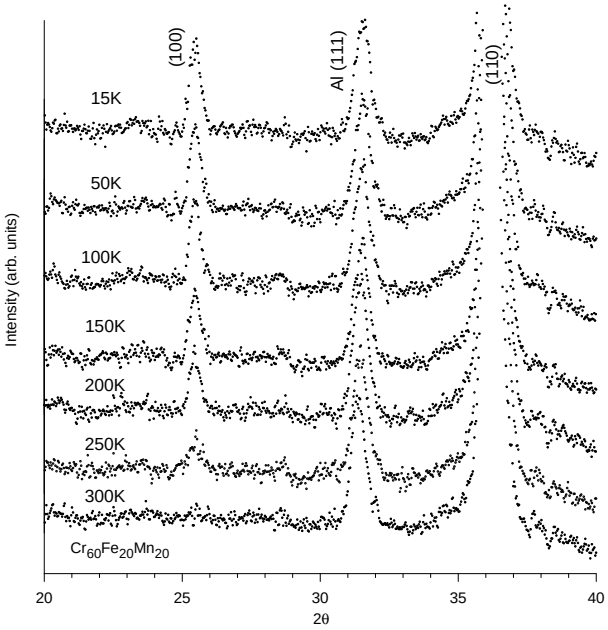

Figure 4. Diffraction patterns at various temperatures indicating the evolution of the antiferromagnetic (100) peak.

namely, the field at which it is cooled, the measuring temperature T/T_G, wait time etc, it needs to be separately studied. Our present results show that relaxation behaviour exists and is observed only in the irreversible regime.

Neutron diffraction patterns at several temperatures between 15 and 300 K were recorded on the high-Q diffractometer. Figure 4 shows a portion of the diffraction pattern. The magnetic

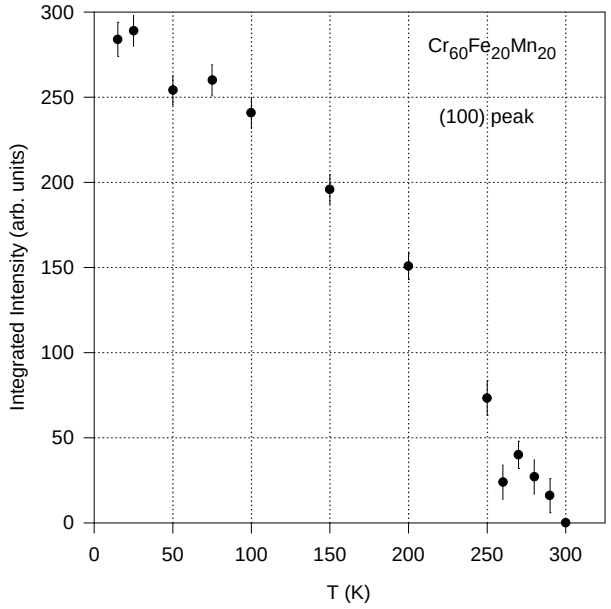

Figure 5. Temperature dependence of the integrated intensity of the antiferromagnetic (100) peak.

(100) peak which is absent at 300 K clearly builds up as the temperature is lowered. The temperature dependence of the integrated intensity of the (100) peak is shown in figure 5. It shows a monotonic increase as the temperature is lowered. The behaviour is similar to that observed in the case of $Cr_{65}Fe_{20}Mn_{15}$ [4]. We associate the antiferromagnetic transition $T_N \sim 290$ K to the observation of the (100) peak. This is in agreement with the value of T_N for this composition obtained from resistivity measurements [22], which shows an increase in resistivity at T_N as the temperature is lowered. This measurement, therefore, confirms the presence of antiferromagnetic order below T_N and its existence down to the lowest temperature. The antiferromagnetic moment has not been estimated as the patterns were recorded on a button shaped sample with preferred orientations. Also due to the limitations with the resolution of the diffractometer we have not been able to carry out peak width analysis to comment upon the nature and extent of antiferromagnetic ordering. The temperature dependence of the integrated intensity of the (110) fundamental reflection follows a trend similar to magnetization and depolarization behaviour. The temperature dependence of the integrated intensity of the (110) peak is shown in figure 6. The intensity increases below 150 K and reaches a maximum at 50 K. The gain in intensity between 150 and 50 K as a result of the ferromagnetic ordering is about 4%. The small change is due to the low spontaneous magnetization, $\sim 0.2\ \mu_B$/atom reported in this alloy. Below 50 K, the integrated intensity apparently decreases. It is of interest to note that in RSG transitions the peak intensity has been reported to decrease, which has been associated with line broadening effects [23]. The decrease in intensity coincides with increase of polarization below 50 K, indicating its origin to the loss of ferromagnetic order.

The picture which emerges is that as temperature is lowered below 55 K small clusters of spin-glass regions appear in the ferromagnetic region which grow in size at the cost of FM regions. At the lowest temperature the FM regions are smaller than the length scale required for neutron depolarization. Therefore, we suggest that at low temperatures the alloy is a mixed phase where spin-glass and antiferromagnetic order coexist. Experimentally, coexistence of antiferromagnetic and spin-glass phases has been reported in $Fe_{0.55}Mg_{0.45}Cl_2$ [24] and

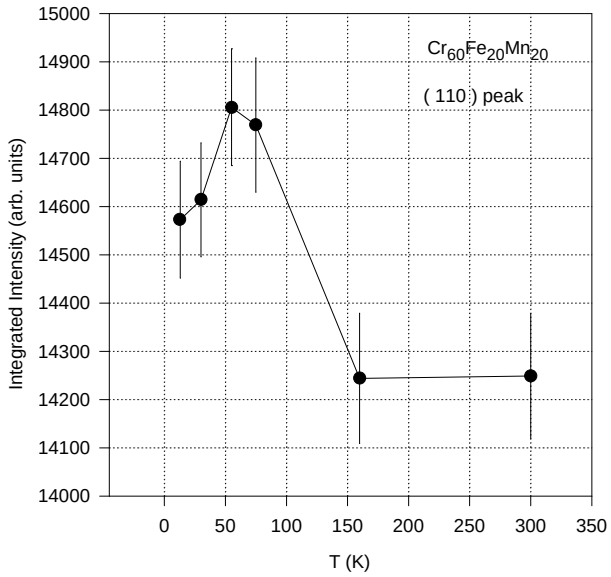

Figure 6. Temperature dependence of the integrated intensity of the (110) fundamental peak.

$Fe_xMn_{1-x}TiO_3$ [25]. However, it is different in the present case as the spin-glass phase which forms below T_G destroys the ferromagnetic order but coexists with the antiferromagnetic order. Theoretically, the reentrant spin-glass phase has been understood on the basis of two different models, the mean field treatment of the Heisenberg spins by Gabay and Toulouse (GT) [26] and the random field effects model proposed by Aeppli *et al* [27]. In the GT model, ferromagnetism and spin glass coexist due to the freezing of spin component transverse to the mean magnetization. Experimentally a number of systems has been found in which ferromagnetic order and spin-glass order coexists [17]. According to the model by Aeppli *et al* [27], in the presence of competing interactions there exists frustrated cluster of spins coupled to the ferromagnetic network. In the SG phase, the frozen spins act to impose a random field on the FM network. The presence of random fields destroys the FM order in a 3d short range order Heisenberg magnet. The breakdown of FM order in the studied RSG alloy may be associated with the random field effect model. However, more experiments are required, particularly SANS and inelastic measurements which are planned.

4. Conclusions

Neutron depolarization has been measured as a function of temperature on $Cr_{60}Fe_{20}Mn_{20}$ alloy. It undergoes a ferromagnetic transition at $T_C \sim 100$ K indicated by a rapid fall in the polarization. The recovery of polarization in the spin-glass phase is attributed to a gradual reduction in the ferromagnetic domain size. At 14 K the polarization in the ZFC condition recovers to the paramagnetic value at 300 K indicating the absence of ferromagnetic order. Neutron diffraction show the appearance of magnetic (100) peak as the temperature is lowered below $T_N \sim 290$ K. It is suggested that at low temperatures a spin-glass phase coexists with an antiferromagnetic phase. A decrease in the integrated intensity of the (110) Bragg peak below 55 K, irreversibility effects and relaxation behaviour in the SG phase are found to be consistent with the above picture.

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