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Magnetic behaviour of $(Fe_{0.90}Mn_xCo_{0.1-x})_2P$

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$(Fe_{0.90}Mn_xCo_{0.1-x})_2P$ with x=0.03, 0.05 and 0.07 (hereafter referred to as FMC37, FMC55 and FMC73, respectively) crystallize in Fe_2P like hexagonal symmetry. DC magnetization measurements have been made on these alloy systems in external fields up to 8.5 kOe and in the temperature range 85-400K. The alloys are ferromagnetic with T_c~325K for FMC37, ~250K for FMC55 and ~220K for FMC73. While FMC37 shows a clear paramagnetic to ferromagnetic transition, in FMC55 and FMC73, the transition anomaly is field-dependent and gets suppressed by rather low fields. In these two systems, FM order keeps evolving with cooling below these Curie points and at still lower temperatures negative exchange gains in strength, giving rise to disordered magnetic state, which shows long time magnetic relaxation. Below 85K, there could be another modification in magnetic states of the two systems with positive exchange interaction regaining in strength.

[Keywords: Disordered magnetic systems, Ferromagnetic alloys, Magnetic properties]

1 Introduction

Di-metal iron phosphide Fe_2P, a ferromagnet with a Curie temperature of 216 K, has been widely studied[1-4]. Its magnetic structure is known to be sensitive to metallic substitutions such that, just 3 atomic% of Cr substituted for Fe, drives the magnetic behaviour to a complex one with changing sequence of temperature[3] and simultaneous substitutions of 5% Cr and 5% Ni introduce a re-entrant spin glass phase[4]. The studies on three systems with simultaneous substitutions of 3 atomic% Mn and 7% Co, 5% Mn and 5% Co, and 7% Mn and 3% Co have been reported. When substituted individually in Fe_2P, it is known that, Co effects to strengthen the FM behaviour and Mn tends to weaken it.

2 Experimental Details

The alloys were prepared using solid-state diffusion technique. Amounts of Fe, Mn, Co and P (of 4N purity) were weighed in stoichiometric proportions and sealed in evacuated quartz ampoules for heating at ~1000 °C. Temperature was slowly raised in about 10 days and then maintained for about a week. The ingot obtained was quenched, powdered, vacuum-sealed and re-annealed at ~1000°C for another week. This process of annealing was repeated once more to ensure homogenization. For structural characterization, the samples were pulverized and subjected to X-ray diffraction measurements on Philips make diffractometer model PW 1840 using FeK_α radiation. DC magnetization measurements were made using PARC vibrating sample magnetometer (model 155).

3 Results and Discussion

X-ray diffraction patterns confirmed the formation of single-phase material with Fe_2P-like hexagonal symmetry in all the three cases. The lattice constants are a = 5.89Å, c = 3.48Å for FMC37, a = 5.89Å, c= 3.49Å for FMC55, and a = 5.90Å, c=3.47Å for FMC73. Figs 1(a) and 1(b) show magnetization-temperature (M–T) measure-ments for FMC37, in the temperature range 90–300K and 300–425K, respectively. In the range 90–300K, the measurements are made in zero field cooled (ZFC) and also field cooled (FC) modes. For measurements in ZFC mode, the sample is first cooled from paramagnetic (PM) state (in this case from ~350K) down to 90K in the absence of any magnetic field. Then, a measuring field is applied and the sample is again heated up to say 300K and moment is recorded during this heating cycle. For measurements in FC mode, the sample is cooled from PM state down to 90K in the presence of the magnetic field and moment is recorded either during this cooling cycle or as the sample is again heated up to 300K. Fig. 1(b) shows that, FMC37 undergoes a well-defined

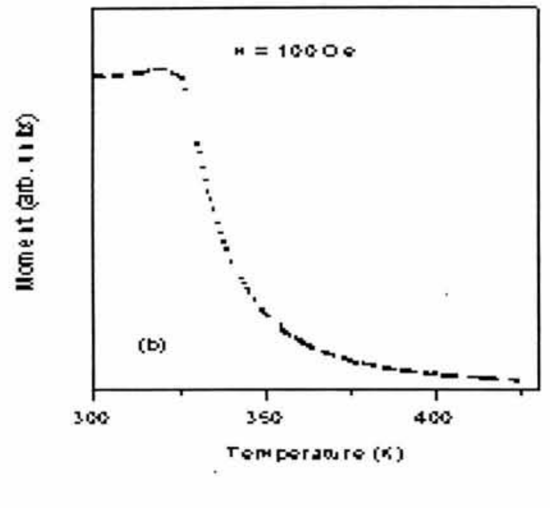

Fig. 1 — *M* versus *T* plots for FMC37 sample in the temperature range, a) 90 – 300K and b) 300 – 425K

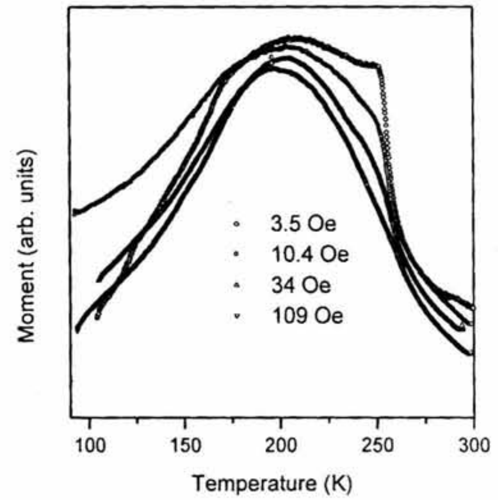

Fig. 2 — *M* versus *T* plots for FMC55 under different fields

paramagnetic-ferromagnetic (PM–FM) transition at ~325K.

The other two alloys with higher Mn content, viz., FMC55 and FMC73, do as well show PM–FM transitions, but not so clearly. Figs 2 & 3 show M–*T* measurements for FMC55 and FMC73, respectively, recorded in ZFC mode in the presence of a few different fields. The first anomaly, occurring at ~250K for FMC55 and at ~220K for FMC73, as the samples are cooled from the high temperature side signify PM-FM transitions, but the transition is not complete; instead the FM state keeps evolving with cooling. Also the *M-T* anomaly at the transition gets suppressed as the measuring field is increased. This signifies first-order transitions

Another point worth noting is that, while in FMC37, once the FM state is attained, magnetic structure does not show any change down to 85K, in the other two alloys, viz., FMC55 and FMC73, the FM state keeps evolving with cooling even below the respective Curie points. Further down below a certain temperature (~220K in FMC55 and ~180 K in FMC73), *M* begins to drop (Fig 2 and 3). It is referred to these anomaly temperatures as, T_a. In both the alloys, this anomaly temperature T_a is field- dependent, decreasing with increasing *H*.

M–T curves, for the two alloys, recorded in FC mode broadly follow the shapes of the ZFC curves. Fig. 4 shows, as an illustrative example, *M–T* measurement in FC mode for the system FMC73 in a field of 203 Oe. The fact of FC curve broadly following the ZFC, one would suggest that, at the anomaly points of ~220 and ~180K negative exchange interaction starts dominating

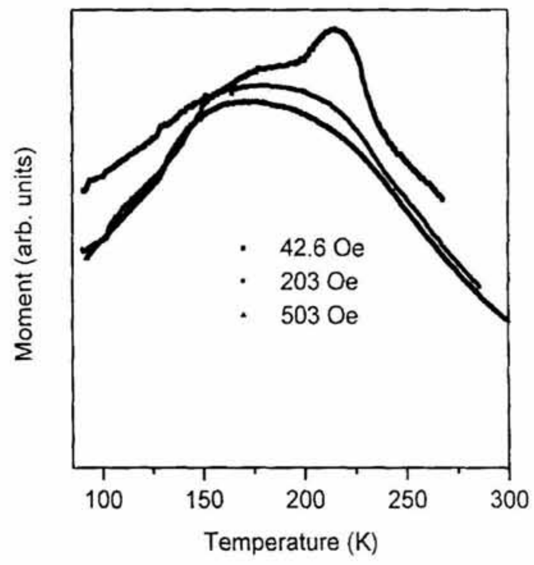

Fig. 3 — *M* versus *T* plots for FMC73 under different fields

the positive exchange one. The alloys do not go to anti-ferromagnetic states; in that case, the moments would drastically drop and FC and ZFC curves would overlap. The magnetic state below these anomaly points would be best described as a disordered one. The fact that T_a shifts downwards with increasing fields, supports this picture. Another fact in support is that of an observed temporal relaxation of magnetization.

Fig. 5 shows, as a typical example, variation of *M* as a function of log of observation time (log *t*) for FMC55 in a small field of 5.5 Oe after zero field cooling. Another

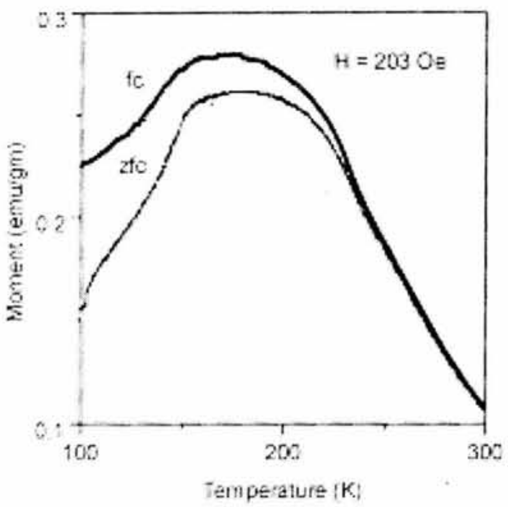

Fig. 4 — *M-T* plot for FMC73 recorded in ZFC and FC modes

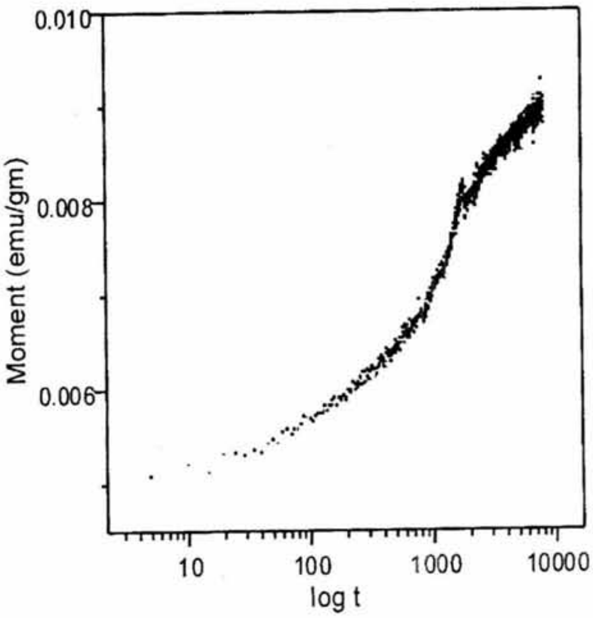

Fig. 5 — *M* versus time plot for FMC55 at 85K in 5.5 Oe after ZFC

worth noting point is, the decreased downward trend of FC curve, i.e., an increasing gap between FC and ZFC curves, at still lower temperatures in FMC73 (cf. Fig. 4) and FMC55 and a continually increasing M in FC curve of FMC37. These observations might indicate towards some other interesting change, involving increased ferromagnetic exchange, in the magnetic structures at temperatures below 90K.

M–H curves at 85K show magnetization at 8.5 kOe to be 58.6, 17.0 and 6.2 emu/g, for FMC37, FMC55 and FMC73, respectively. Compared to the saturation magnetization of 120 emu/g for single crystal of Fe_2P (and 48 emu/g in a field of 8.5 kOe at 10 K for powder sample) these values are very small, further indicating weakened FM order. Remanence and coercivity values at 85K are, 5.15 emu/g and 0.277 kOe for FMC37, 0.73 emu/g and 0.350 kOe for FMC55, and 0.262 emu/g and 0.269 kOe for FMC73. These values also generally indicate to low magnetic anisotropy.

In conclusion, the three alloys with simultaneous substitutions of Mn and Co for Fe, in the ferromagnetic Fe_2P, undergo PM-FM transitions –7 atomic% Co (even with 3% Mn), drives T_c from 216K in Fe_2P to 325K; increased Mn effects to reduce T_c to ~250K in FMC55 and ~220K in FMC73. In FMC55 and FMC73 PM-FM transition anomaly is quite sensitive to applied magnetic field and the transition is of first-order type. Also, in these two higher Mn containing systems, below some anomaly temperature (~220K for FMC55 and ~180K for FMC73) negative exchange gains in strength, giving rise to disordered magnetic structure. There are signatures suggesting that, below 90K new magnetic structures may show up.

In the unit cell of the parent system Fe_2P, there are four different metal-metal bond lengths giving rise to simultaneous presence of positive and negative exchange interactions. Observed complex magnetic behaviour of the studied alloys would owe to this fact[3].

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