Composition dependence of magnetoresistance in Fe$_{1-x}$Ni$_x$ alloys

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Abstract

Resistance of Fe$_{1-x}$Ni$_x$ ($x=0.1$, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7 and 0.9) has been measured using four probe method from 5K to 300K with and without a longitudinal magnetic field of 8T. The zero field resistivity of $x=0.1$ and 0.9 alloys, predominant contribution to resistivity above near room temperature is due to electron-phonon scattering, whereas for $x=0.5$ and 0.7 alloys electron-magnon scattering is dominant. Alloys with $x=0.1$ and 0.9 exhibit positive magnetoresistance (MR) from 5K to 300K. For $x=0.5$ and 0.7 alloys, magnetoresistance changes sign from positive to negative with increase in temperature. The temperature at which sign changes increase with Ni concentration in the alloy. The field dependent magnetoresistance is positive for $x=0.1$, 0.7 and 0.9 alloys whereas it is negative for $x=0.5$ alloy. MR follows linear behaviour with field for $x=0.1$ alloy. MR of all other alloys follow a second order polynomial in field.

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1 Introduction

FeNi alloys exhibit anomalous magnetic, elastic and structural properties. The phase diagram of Fe$_{1-x}$Ni$_x$ alloy system is very complicated and is still being investigated [1] [2]. Pure Fe metal possesses bcc structure ($\alpha$-phase) and exhibits a structural transition to fcc structure ($\gamma$-phase) upon alloying with Ni at around a Ni concentration of 30% and maintains the same structure upto 100% of Ni concentration. Both $\alpha$ and $\gamma$ phases form disordered alloys by metastable quenching. Ordered alloys form for 50% (FeNi) and 75% (FeNi$_3$) of Ni concentrations. But FeNi ordered phase can not be formed by simple annealing. The fcc phase exhibits complex magnetic behavior and is known to exist in different magnetic states [3] similar to pure fcc Fe [4]. Charles Edouard Guillaume was awarded the 1920 Physics Nobel Prize for the discovery of Invar effect (invariance of thermal expansion over a wide range of temperature) in iron-nickel ferromagnetic alloys with a nickel concentration of 35% [5].

In Fe$_{1-x}$Ni$_x$ alloys, Fe magnetic moment increases with Ni concentration both in bcc and fcc phases. Abrikosov et.al [6] predicted that Fe$_{65}$Ni$_{35}$ FM alloy first transforms into a ferrimagnetic system with local moments on Fe atom, surrounded mostly by Fe atoms, pointing antiparallel to the net magnetization. Such spin flip sites promote non-collinear magnetic structures inside FM matrix. Local Fe and Ni magnetic moments in FeNi alloys are marginally different from pure metal values and exhibit a week dependence on concentration of the alloy [7]. In these alloys Ni moment ranges from 0.6-0.7 $\mu$B and that of Fe ranges from 2.5-2.7 $\mu$B [8]. Several theoretical studies

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indicated $J_{FeFe}$ near neighbour exchange integral is negative in FeNi alloys suggesting antiferromagnetic coupling between Fe atoms [9]. For high Fe concentrations magnetic moment decreases due to the increase of antiferromagnetic behaviour as Fe-Fe nearest neighbour interactions start to dominate [10]. Re-entrant spin glass (RSG) behaviour has been reported at low temperatures in Fe$_{1-x}$Ni$_x$ alloys for $x$ values ranging from 0.3 - 0.5 [11].

Fe-Ni alloys are extensively used in recording industry. These alloys are also used in making transformers, electric motors, turbine blades of jet engine, space craft and nuclear reactors. Fe-Ni Invar alloys are used in making watches and liquid helium containers. Fe-Ni alloys exhibit exotic physical properties and possess complex magnetic structures, the origin of which demands further attention. These alloys are important from both fundamental and applied points of view. This paper reports the study of magnetoresistance (MR) of Fe$_{1-x}$Ni$_x$ alloys. Our results indicated a negative field dependent MR for $x = 0.5$ alloy at low temperatures suggesting re-entrant spin glass behaviour. All other alloys investigated in this study exhibit positive MR.

### 2 Sample Preparation and Characterization

Fe$_{1-x}$Ni$_x$ ($x=0.1$, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9) have been prepared using arc melting method by co-melting the high purity metals in an inert atmosphere of Ar gas. Homogeneity of the samples has been ensured by remelting the ingots several times keeping in different orientations. The samples were wrapped in a molybdenum foil and were annealed at high vacuum in an evacuated quartz tube. The alloys were structurally characterised using X-ray diffraction technique. XRD patterns have been indexed and the lattice parameters have been refined using PowderX software [12] [13]. The resistivity of the samples has been measured with (8T, longitudinal) and without applied magnetic field (0T) down to 5K using four probe method. Magnetic properties of Fe$_{1-x}$Ni$_x$ binary alloys were measured using SQUID (Quantum design-MPMS-XL) magnetometer at T.I.F.R. Mumbai. The magnetic hysteresis loops were measured up to 8 T magnetic field at 300 K, 100 K, 40 K, 20 K and 5 K temperatures. The temperature dependence of magnetization was measured from 5 K to 300 K.

### 3 Results and Discussion

Figure 1 (a), (b) and (c) show the XRD patterns of Fe$_{1-x}$Ni$_x$ alloys obtained using CuK$_{α}$ radiation. Fe$_{1-x}$Ni$_x$ alloys for $x=0.1$, 0.2 are in bcc phase and alloys having nickel content range 0.4 to 0.9 are in fcc phase. However both bcc and fcc phases coexist for Fe$_{0.7}$Ni$_{0.3}$ alloy. This alloy contains predominantly fcc phase with small amount of bcc phase as indicated by the intensities of 111 and 110 reflections in figure 1(b). A structural phase transition from bcc to fcc is observed for the Fe-Ni alloys between $x=0.3$ and 0.4. XRD patterns have been shifted along the vertical axis for clarity.

Figure 2 shows composition dependence of lattice parameter for both (a) bcc and fcc phases and (b) fcc phase. The lattice parameter of fcc alloys follows a linear behaviour with Ni concentration. According to Vegard’s law the linear behaviour of lattice parameter confirms the formation of a random alloy [14]. Temperature dependent resistivity for Fe$_{0.9}$Ni$_{0.1}$, Fe$_{0.8}$Ni$_{0.2}$, Fe$_{0.7}$Ni$_{0.3}$ and Fe$_{0.6}$Ni$_{0.4}$ alloys recorded in both cooling
Figure 1: XRD patterns of Fe$_{1-x}$Ni$_x$ alloys for (a) x=0.1, 0.2, (b) x=0.3 and (c) x=0.4, 0.5, 0.6, 0.7 and 0.9 recorded using CuK$_\alpha$ radiation. Alloys having low nickel content upto 20%, are in bcc phase whereas alloys having nickel content starting from 40% onwards possess fcc phase. Both bcc and fcc phases coexist for 30% nickel alloy.

Figure 2: Variation of lattice parameter of Fe$_{1-x}$Ni$_x$ alloys with Ni concentration (a) for both bcc and fcc phases and (b) for fcc phase. In the fcc region lattice parameter exhibits linear dependence on nickel concentration.

and heating cycles are shown in figure 3 (a), (b), (c) and (d) respectively. The Fe$_{0.8}$Ni$_{0.2}$ bcc alloy exhibits a large hysteresis with temperature due to Austenite (bcc phase) to martensite (fcc phase) transition occurring during cooling cycle. The Fe$_{0.9}$Ni$_{0.1}$ alloy also exhibits a weak hysteresis as shown in the inset of figure 3 (a). The alloys with x=0.3 and 0.4 do not show hysteresis as shown in figure 3 (c) and (d). No hysteresis was observed for other alloys with x$\leq$0.4 (not shown). The martensite start and finish temperatures ($M_s$ and $M_f$) reported for Fe$_{0.8}$Ni$_{0.2}$ alloy are around 550 K and 300 K respectively. The data presented in figure 3 indicates the $M_s$ value of 54 K and $M_f$ is above room temperature for Fe$_{0.8}$Ni$_{0.2}$ alloys. Martensitic transition is clearly observed for Fe$_{0.8}$Ni$_{0.2}$ alloy as shown in figure 3 (b). For Fe$_{0.9}$Ni$_{0.1}$ alloy martensitic transition is indicated in the inset of figure 3 (a) after magnifying in a temperature range from 263.5 K to 234 K. Though martensitic transition has been started much above room temperature during cooling for Fe$_{1-x}$Ni$_x$ alloys with low nickel content ranges from 10% to 30% reported in [15], but the former is observed to be finished at around 17 K and 54 K for Fe$_{0.9}$Ni$_{0.1}$ and Fe$_{0.8}$Ni$_{0.2}$ alloys respectively. For Fe$_{1-x}$Ni$_x$ alloys having nickel content more than 30%, signature of martensitic transition is absent in the temperature range studied as indicated in figure 3 (c) and (d).

Figure 4 shows field dependent magnetoresistance at 5 K temperature for Fe$_{1-x}$Ni$_x$ (x=0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7 and 0.9) alloys. The field dependent
Figure 3: Temperature dependent resistivity for Fe$_{1-x}$Ni$_x$ alloys recorded in both cooling and heating cycles for (a) 10%, (b) 20%, (c) 30% and (d) 40% nickel concentration. Martensitic transition is clearly observed for Fe$_{0.8}$Ni$_{0.2}$ alloy whereas for Fe$_{0.9}$Ni$_{0.1}$ alloy martensitic transition is shown in the inset of (a).

Magnetoresistance is calculated as 5 K as

$$\frac{\Delta \rho}{\rho} = \frac{\rho(H,T) - \rho(0,T)}{\rho(0,T)}$$

(1)

Magnetoresistance (MR) values calculated as above are shown in figure 4 for Fe$_{1-x}$Ni$_x$ alloys. For x=0.1, 0.3, 0.7 and 0.9 alloys, MR is positive whereas x=0.2, 0.4 and 0.5 alloys show negative MR at 5 K temperature. The positive MR indicates the normal metallic behaviour with insignificant magnetic scattering of electrons. The negative MR suggests magnetic ordering. When the alloy is ordered magnetically, the magnetic scattering decreases with magnetic field and the MR becomes negative. Fe$_{0.6}$Ni$_{0.4}$ invar alloy exhibits spin glass precipitation below 20 K, due to which it exhibits more negative MR at 5 K.

Figure 5 shows the numerical fitting to the data shown in figure 4. Fe$_{1-x}$Ni$_x$ alloys with x=0.1 and 0.4 exhibit linear dependence whereas the other alloys with x= 0.2, 0.3, 0.5, 0.6, 0.7 and 0.9 show quadratic dependence on the applied field. The positive MR with quadratic dependence indicates predominantly the free electron type conduction. The linear dependence suggests scattering due to spin...
It decreases to 83 emu/gm for x=0.5 and 119 emu/gm for x=0.1, 0.2 and 0.3 respectively and is 163 emu/gm for x=0.1, 0.2 and 0.3. Magnetization is 183 emu/gm, 130 emu/gm occurs at 1100 Oe and it decreases gradually for x=0.1 sample saturation of magnetization is 164 emu/gm and 143 emu/gm at 5 K and 300 K temperatures. However for Fe$_{0.6}$Ni$_{0.4}$ alloy saturation magnetization is 164 emu/gm and 143 emu/gm at 5 K and 300 K temperatures respectively.

The hysteresis loops of all the alloys except Fe$_{0.6}$Ni$_{0.4}$ coincide at 300 K and 5 K temperatures. For x=0.1, 0.2, 0.3, 0.4, 0.5 and 0.9 alloy systems for Fe$_{1-x}$Ni$_x$ alloys measured at 300 K and 5 K. All samples exhibit hysteresis loops similar to ferromagnetic materials. Interestingly, magnetization saturation occurs at lower fields with increase of Ni concentration. For x=0.1 sample saturation of magnetization occurs at 1100 Oe and it decreases gradually to 500 Oe for x=0.9 sample. Saturation magnetization is 183 emu/gm, 130 emu/gm and 163 emu/gm for x=0.1, 0.2 and 0.3 respectively and is 119 emu/gm for x=0.5 sample. It decreases to 83 emu/gm for x=0.9. The hysteresis loops of all the alloys except Fe$_{0.6}$Ni$_{0.4}$ coincide at 300 K and 5 K temperatures. However for Fe$_{0.6}$Ni$_{0.4}$ alloy saturation magnetization is 164 emu/gm and 143 emu/gm at 5 K and 300 K temperatures respectively.

### 4 conclusion

A structural phase transition from bcc to fcc phase is clearly observed in Fe$_{1-x}$Ni$_x$ alloy system. Alloys with Ni content upto 20% are in bcc phase and the alloys with Ni content more than 30% are in fcc phase. A large negative MR at low temperatures exhibited by the invar alloy, Fe$_{0.6}$Ni$_{0.4}$ suggests the formation of RSG ground state. The signature of RSG state is also evident from the increase in saturation magnetization at 5 K to that at 300K for the...
invar alloy. The linear dependence of MR with field at 5K indicates scattering due to spin disorder.

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