Effect of Adding of V and Co Atoms on the Spin Glass Transition Temperature of $Fe_{0.7-x}M_xAl_{0.30}$ Alloys

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Magnetization measurements on zero field cooled $(M_{\rm ZFC})$ and field cooled $(M_{\rm FC})$ with temperature T were made on Fe_{0.7-x}M_xAl_{0.30} alloys (where M = Co or M = V). Compared to Co and V free alloy Fe_{0.7}Al_{0.30}, our results revealed that an addition of small amount of M = Co with $0 \le x \le 0.10$ shows linear decrease in the transition temperature $T_{\rm f}$ as x increases. Addition of small amount of M = V with $0 \le x \le 0.1$ shows gradual decrease in $T_{\rm f}$ with increasing x. The relative changes in $T_{\rm f}$ show that an addition of V atoms is of more significance than addition of Co atoms. There is a twofold decrease which could be attributed to the ferromagnetic nature of Co atoms and to the random fields.

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1. Introduction

Soon after the nature of spin glasses began to be identified in dilute magnetic alloys, which exhibit paramagnetic to spin-glass transition [1, 2] more concentrated magnetic alloys were discovered to have an even more unusual sequence of phase transitions as the temperature is lowered. Some of these systems were found to undergo a paramagnetic (PM) to ferromagnetic (FM) transition then followed by transition into a spin glass state as the temperature is lowered where we refer to this temperature as freezing temperature $T_{\rm f}$. This re-entrant sequence of transitions was found in crystalline [3, 4], amorphous [5, 6] and ionic crystals [7].

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In $\text{Fe}_{1-x}\text{Al}_x$, studies of the magnetic behavior research were focused on composition of x = 0.30. It was reported that the $\text{Fe}_{0.7}\text{Al}_{0.3}$ exhibits re-entrant spin glass behavior. It goes from PM to FM at $T_c = 430$ K followed by PM state below new $T_c = 170$ K and then transition to spin glass state at temperatures below $T_f = 90$ K [8–11]. The disappearance of the spontaneous magnetization in alloys near this concentration was first observed in 1959 [12, 13]. Kouvel pointed out a co-existence of ferromagnetic and antiferromagnetic phases [14]. Later neutron diffraction measurements revealed no evidence for antiferromagnetic ordering above 4.2 K for Al concentration in the range 30–50 at.% [9].

Low field ac susceptibility and dc magnetization measurements as function of temperature (T) and concentration (x) further establish the phase diagrams and the nature of atomic ordering in Fe–Al alloys [15]. Careful studies of atomic order in Fe_{1-x}Al_x alloys with x near to 0.30 revealed both FeAl and Fe₃Al type order with differences in Fe and Al arrangements in the two cases [16, 17]. One of the most unusual aspects of this work was the establishment of the existence of a PM through FM to PM followed by spin glass (SG) phase as mentioned above. However, both diffuse and polarized neutron scattering performed on Fe_{0.7}Al_{0.3} alloy indicated FM clusters at temperatures in the range 10–500 K [18]. Furthermore, the data suggested that the interaction of spin clusters gives rise to magnetic transitions.

Several studies were done using X-ray and neutron diffraction on FeAl_{1-x}T_x alloys with solutes T of V, Cr, Ti, Mn, and Co [19, 20]. Also, Mössbauer studies were performed on FeAl alloys [21] and FeAl_{1-x}T_x with solutes of V and Mn [22]. Recently our studies of the Mössbauer spectra of Fe_{0.7-x}Ti_xAl_{0.3} with $0 \le x \le 0.3$ and Fe_{0.7-x}V_xAl_{0.3} showed that all samples keep CsCl (B2) structure and change from ferromagnetic phase to paramagnetic phase as x increases [23, 24]. In the case of FeCoAl alloys it was found that Co atoms exercised a preference for Fe sites and ferromagnetic behavior was only found for Co concentration above x = 0.02.

In this work we have investigated the magnetic behavior of $\text{Fe}_{0.7-x}M_x\text{Al}_{0.30}$ where M = Co, V with $0 \le x \le 0.10$. We performed zero field cooled (ZFC) and field cooled (FC) magnetization measurement as function of temperature.

Since the critical value of x = 0.3 in $\text{Fe}_{1-x}\text{Al}_x$, we focused on the effect of adding small amount of x < 0.30 of Co and V in the FeAl alloy.

2. Sample preparation

Our samples of $\text{Fe}_{0.7-x}M_x\text{Al}_{0.3}$ with M = Co, V were prepared for $0 \leq x \leq 0.10$ using elements of at least 99.99% purity. Stoichiometric elements were arc melted in a water cooled copper boat in a flowing argon gas. Alloys were melted several times to ensure homogeneity. The samples were wrapped separately in tantalum foil and heat treated in a vacuum furnace (less than 3×10^{-6} Torr) at 830°C for 24 hours and subsequently quenched in water. They were cut with a diamond slicing wheel to various sizes of order 0.5 mm \times 0.5 mm \times 3.5 mm.

3. Results and discussion

The magnetic measurements were carried out using a commercial superconducting quantum interface device magnetometer (Quantum Design) SQUID magnetic properties measurement system (MPMS) equipped with a low field option. The magnetization measurements were made by first demagnetization of the whole system at room temperature using flux gate. The sample was then cooled down to 10 K in zero field (ZFC). A magnetic field of 100 Oe was then applied at this temperature. The sample temperature was raised slowly to 300 K and the magnetization M versus the temperature T was recorded at every 2 K. After reaching 300 K the sample was then field cooled (FC) down to 10 K and M versus T was recorded.

We have made measurements of the magnetization M versus T on Fe_{0.7}Al_{0.3} alloy to determine the freezing temperature $T_{\rm f}$, where we found that the transition to spin glass phase occurs at $T_{\rm f} = 70$ K and the transition to ferromagnetic phase occurs at T = 170 K. This is consistent with earlier work done on this compound [9]. In what follows we will use the value of $T_{\rm f} = 70$ K as a reference to study the effect of addition of Co atoms and V atoms on the freezing temperature $T_{\rm f}$. The results of $M_{\rm ZFC}$ and $M_{\rm FC}$ versus T for the Fe_{0.7-x}Co_xAl_{0.30} are shown in Fig. 1. The figure shows the data for x = 0.04 and 0.08. The departure between $M_{\rm ZFC}$ and $M_{\rm FC}$ gives the value of $T_{\rm f}$. For this particular concentration $T_{\rm f} = 60$ K for x = 0.04 and $T_{\rm f} = 55$ K for x = 0.08 as can be found directly from the figure. The same sort of behavior was found for the other values of x. Getting $T_{\rm f}$ for each x we plot the results in Fig. 2 of $T_{\rm f}$ versus x. The figure shows a linear decrease in $T_{\rm f}$ as x increases. The relative change in $T_{\rm f}$ is almost of 30% at x = 0.30 compared to the free Co alloy. Even an addition of small amount of x = 0.02 showed a very significant decrease in $T_{\rm f}$ of about 9%. These relative changes are shown in Fig. 5 which can be easily fitted by a straight line.



Fig. 1. Magnetization M versus T for $\text{Fe}_{0.7-x}\text{Co}_x\text{Al}_{0.3}$ for x = 0.04 and x = 0.08. Arrows indicate the increase in T for the M_{ZFC} and the decrease in T for M_{FC} .



Fig. 2. The transition temperature $T_{\rm f}$ versus x for the ${\rm Fe}_{0.7-x}{\rm Co}_x{\rm Al}_{0.3}$ alloy.



Fig. 3. Magnetization M versus T for $\text{Fe}_{0.7-x} V_x \text{Al}_{0.3}$ for x = 0.02 and x = 0.10. Arrows indicate the increase in T for the M_{ZFC} and the decrease in T for M_{FC} .

The same measurements were repeated in the case of the Fe_{0.7-x}V_xAl_{0.30}. Our result is shown in Fig. 3 where, as a typical, behavior of M versus T is shown for x = 0.06. The same behavior was found for the values of x. The depicted values of $T_{\rm f}$ are shown in Fig. 4; there is plotted $T_{\rm f}$ versus x. The figure shows gradual decrease in $T_{\rm f}$ as x increases. The change in $T_{\rm f}$ is very fast at lower values of x. It is 20% for x = 0.02 and 56% at x = 0.1. These relative changes of $T_{\rm f}$ are shown in Fig. 5. In fact, in Fig. 5 we plot both data for Co and V for comparison. Comparing the relative changes in $T_{\rm f}$ for both samples indicates that an addition of small amounts of V is suppressing $T_{\rm f}$ more than the addition of Co. This may be attributed to the ferromagnetic nature of Co atoms.

The competition between the paramagnetic clusters and the ferromagnetic clusters which exists both in the Co free and V free alloy is responsible for the disappearance of the ferromagnetic clusters and appearance of a paramagnetic phase where the ferromagnetic structure breaks up into finite sized clusters. In this work the change in the freezing temperature $T_{\rm f}$, which is sensitive more to the



Fig. 4. The transition temperature $T_{\rm f}$ versus x for the ${\rm Fe}_{0.7-x}{\rm Co}_x{\rm Al}_{0.3}$ alloy.



Fig. 5. The relative change in $T_{\rm f}$ versus x. Closed circles for Co addition and open circles for V addition.

addition of the V atoms than Co atoms, may be attributed to the ferromagnetic nature of the Co atoms which has more effect in the ferromagnetic phase. The V atoms have more effect on the paramagnetic phase. It is worth mentioning that our Mössbauer study in the Fe_{0.7-x}V_xAl_{0.30} alloy revealed paramagnetic phases above x > 0.3 and definite magnetic order exists at $x \leq 0.1$ [24].

Finally, we suggest that neutron scattering studies will give more insight into the magnetic environment where one can look to spin clusters behavior.

4. Conclusions

In this study we found that there is a significant effect on the freezing temperature $T_{\rm f}$ when we add small amounts of Co or V to the Fe_{0.7-x}M_xAl_{0.30} alloy. This effect is associated with linear decrease in $T_{\rm f}$ in case of Co substitution and gradual decrease in $T_{\rm f}$ in case of V substitution. At the small values of x, the effect of V addition is of more significance on $T_{\rm f}$ than Co. This addition of V atoms is associated with more than twofold decrease in $T_{\rm f}$ compared to the addition of Co atoms.

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