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Effect of site disorder on the magnetic properties of weak itinerant ferromagnet $Ni_{75}Al_{25}$*

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Abstract. Detailed study of $Ni_{75}Al_{25}$ samples with varying degree of site disorder reveals that site disorder promotes magnetic excitations such as spin waves and local spin-density fluctuations and thereby reduces both spin-wave stiffness and Curie temperature. Irreversibility lines in the T–H phase diagram of the weak itinerant ferromagnet $Ni_{75}Al_{25}$ have been determined for the first time and the effect of site disorder on them has been ascertained.

Keywords. Itinerant ferromagnet; Ni_3Al; site disorder; spin wave stiffness; magnetic irreversibility; spin fluctuations.

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1. Introduction

Intense scientific activity in the field of metallic (band) magnetism has led to a substantial progress in understanding various physical phenomena associated with weak itinerant-electron (WI) ferromagnetism, in particular. Nevertheless, certain aspects of WI ferromagnetism are still not understood completely. For instance, the role of site disorder is not clear even in the most extensively studied WI ferromagnet, Ni_3Al. The spin fluctuation theories [1], proposed for the WI ferromagnets such as the ordered Ni_3Al intermetallic compound, do not deal with site disorder, which is invariably present in any real alloy system. Early magnetic investigations [2,3] on plastically deformed $Ni_{75}Al_{25}$ have shown that a complete breakdown of long-range ferromagnetic order occurs and superparamagnetic behaviour persists down to 4.2 K. Recently, a detailed comparative bulk magnetisation study [4] of (disordered) nanocrystalline and ordered polycrystalline samples of Ni_3Al revealed that

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the nanocrystalline counterpart exhibits exchange-enhanced Pauli spin paramagnetism for $T \geq 5$ K. Both these observations provide ample evidence for the extreme sensitivity of the ground-state as well as finite-temperature magnetic properties of $Ni_{75}Al_{25}$ to site disorder. This prompted us to undertake a detailed study of the correlation between site disorder and magnetic properties of the WI ferromagnet $Ni_{75}Al_{25}$.

2. Sample preparation

Starting with 5-N purity Ni and Al, a polycrystalline rod (diameter : 10 mm, length : 100 mm) of $Ni_{75}Al_{25}$ was prepared using RF induction melting technique. Spheres of 3 mm diameter and discs of 10 mm diameter and 5 mm thickness were spark-cut from the rod. One sphere and one disc were annealed at 520°C for 16 days and subsequently water-quenched. A portion of the as-prepared polycrystalline rod was melt-quenched to form long thin ribbons of width 2 mm and thickness $\sim$30 μm. The samples in the annealed, 'as-prepared' and quenched forms are henceforth referred to as S1, S2 and S3, respectively. From a single crystal grown by zone-refining technique, a small cylinder of 2 mm diameter and 3 mm length S4 was spark-cut such that [111] direction, which is the easy direction of magnetisation, coincides with the cylindrical axis. A textured polycrystalline $Ni_{75}Al_{25}$ sample S5 with a high degree of chemical inhomogeneity was prepared by Bridgeman technique. The rest of the pieces left after spark-cutting the samples S1–S5 were analysed for chemical composition so as to confirm that the samples indeed conform to the stoichiometric composition $Ni_{75}Al_{25}$.

3. Results, discussion and conclusions

X-ray diffraction patterns were analysed to obtain accurate values for the long-range atomic order parameter, $\mathscr{S}$, which is a measure of atomic order, and interatomic spacing a (figure 1). Magnetisation (M) was measured as a function of temperature from 15 K to 300 K at fixed magnetic fields (H) up to 15 kOe in the zero-field-cooled (ZFC) and field-cooled (FC) modes, and hysteresis loops were recorded on a vibrating sample magnetometer. In addition, M vs. H ($H \leq 70$ kOe) isotherms at $T = 5$ K and M vs. T data at $H = 1$ kOe and 10 kOe were taken on a SQUID magnetometer. From the spontaneous magnetization, $M(T,0)$, at $T = 5$ K (deduced from the M vs. H isotherms) atomic moment per Ni atom, μ_{Ni}, was calculated for all the samples (figure 1). The spin-wave [5] and critical-point [6] analyses of the $M(T,H)$ data yield accurate values for the spin-wave stiffness at 0 K, $D(0)$ and the Curie temperature, T_{C}. The $D(0)/T_{\mathrm{C}}$ ratio and T_{C} are plotted against $\mathscr{S}$ in figure 1. As site disorder increases (i.e., decreasing $\mathscr{S}$), a and $D(0)/T_{\mathrm{C}}$ increase while μ_{Ni} and T_{C} decrease rapidly for $\mathscr{S} \leq 0.85$. With decreasing $\mathscr{S}$, the number of the nearest neighbours for a given Ni atom decreases and hence μ_{Ni} decreases. Since $D(0)/T_{\mathrm{C}} \propto \mu_{\mathrm{Ni}}^{-1/2}$ [1], this ratio increases as μ_{Ni} decreases. Site disorder promotes the local spin-density fluctuations with the result that $M(T,0)$ diminishes at a faster rate as the temperature is increased. Consequently, T_{C} falls steeply below a certain threshold value of $\mathscr{S}$.

The difference between M_{FC} and M_{ZFC} at a given temperature and field is a direct measure of irreversibility at that temperature and field. The features presented by the

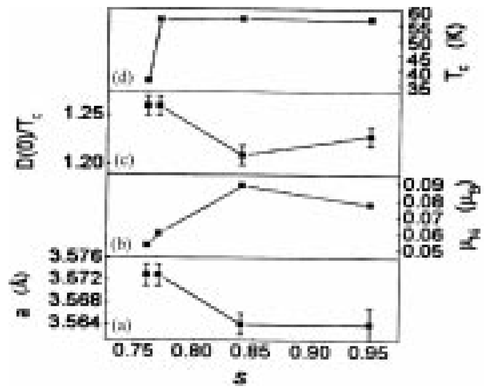

Figure 1. Lattice parameter *a* (**a**), magnetic moment per Ni atom, μ_{Ni} (**b**), the ratio of spin-wave stiffness $D(0)$ and T_{C} (**c**), and as functions of Curie temperature T_{C} long-range order parameter $\mathscr{S}$ (**d**).

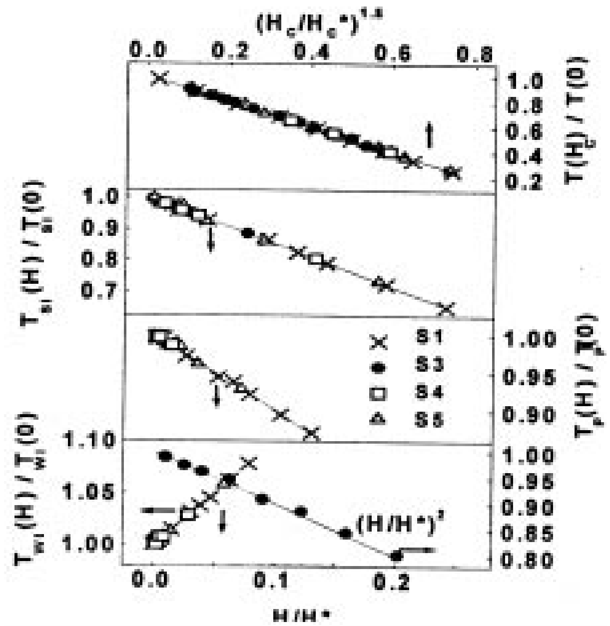

Figure 2. Reduced temperatures for weak irreversibility (WI), strong irreversibility (SI), peak irreversibility (P) and coercivity as functions of reduced magnetic field (H/H^*)/reduced coercive field $(H_{\mathrm{C}}/H_{\mathrm{C}}^*)$ for the samples S1, S3, S4, S5.

$[M_{\mathrm{FC}}(T) - M_{\mathrm{ZFC}}(T)]$ curves taken at fixed H are: (i) the temperature $T_{\mathrm{WI}}(H)\{T_{\mathrm{SI}}(H)\}$ below which the difference $[M_{\mathrm{FC}}(T) - M_{\mathrm{ZFC}}(T)]$ deviates from zero (increases steeply) and which marks the onset of weak irreversibility (WI) (strong irreversibility (SI)) and (ii) the temperature $T_{\mathrm{P}}(H)$ at which the difference goes through a peak (observed in all the samples except for the quenched sample S3). For the samples S1 and S4, the irreversibility lines (loci of $T_{\rm WI}$, $T_{\rm SI}$ and $T_{\rm P}$ points) in the T–H phase diagram follow the relations $\tau_{\rm WI}(H) \equiv T_{\rm WI}(H)/T_{\rm WI}(0) = 1 + (H/H^*_{\rm WI})$, $\tau_{\rm SI}(H) \equiv T_{\rm SI}(H)/T_{\rm SI}(0) = 1 - (H/H^*_{\rm SI})$ and $\tau_{\rm P}(H) \equiv T_{\rm P}(H)/T_{\rm P}(0) = 1 - (H/H^*_{\rm P})$ whereas for the quenched sample S3, only the relation $\tau_{\rm SI}(H) \sim H$ is obeyed, $\tau_{\rm P} - H$ irreversibility line does not exist and the weak irreversibility line is described by the expression $T_{\rm WI}(H) = 1 - (H/H^*_{\rm WI})^2$, as is noticed from the data presented in figure 2. In these expressions, the quantity H^* is the field that characterises the magnetic irreversibility and depends on the degree of site disorder present. The mean-field vector-spin models [7], applicable to spin glasses, predict that $\tau_{\rm WI}(H) \sim H^2$ and $\tau_{\rm SI}(H) \sim H^{2/3}$. The theoretically predicted field dependences are at variance (except for the WI line in the case of S3) with the observed ones. In order to ascertain if anisotropy is at the root of the observed irreversibility, coercive field, $H_{\rm C}$, has been determined at different temperatures from the measured hysteresis loops. $H_{\rm C}(T)$ follows the relation $T(H_{\rm C})/T(0) = 1 - (H_{\rm C}/H^*_{\rm C})^{1.5}$ (figure 2) which, again, does not conform to the observed field dependences of $\tau_{\rm WI}$, $\tau_{\rm SI}$ and $\tau_{\rm P}$. Thus, no obvious connection between anisotropy and irreversibility could be established.

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