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Site occupancy of Fe in ternary $Ni_{75-x}Fe_{x+y}Al_{25-y}$ alloys*

B ANNIE D'SANTHOSHINI and S N KAUL
School of Physics, University of Hyderabad, Hyderabad 500 046, India
Email: kaulsp@uohyd.ernet.in

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Abstract. The results of a detailed structural and magnetic study clearly indicate that regardless of the thermal history of the samples, Fe has a strong preference for the Ni sites in Ni-poor (non-stoichiometric) $Ni_{75}Al_{25}$ alloys. Fe substitution has a profound effect on the nature of magnetism in $Ni_{75}Al_{25}$.

Keywords. Itinerant ferromagnet; Ni–Fe–Al; site and bond disorder; site preference of Fe; magnetisation; structure.

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1. Introduction

The parent alloy Ni_3Al is an ordered intermetallic compound in which Ni and Al atoms respectively occupy face centres (A sites) and corners (B sites) of the face-centred-cubic (fcc) unit cell. The Ni and Al atoms thus constitute the so-called A and B sublattices. A substitutional ternary species may prefer A or B site or may not have any preference at all. Site selectivity/preference of the alloying species has a profound effect on the physical properties of the parent system. As far as the magnetic properties are concerned, they can get significantly altered if the alloying species is a magnetic one. While the Ni sublattice preference is well established in the case of Co and Pd substitution, no definite site substitution pattern of Fe has emerged from the theoretical [1,2] and experimental [3,4] studies so far. To resolve this issue, an in-depth study of the structure and magnetic property correlation in the alloys with nominal composition $Ni_{75}Al_{25}$, $Ni_{70}Fe_5Al_{25}$, $Ni_{65}Fe_{10}Al_{25}$ and $Ni_{55}Fe_{20}Al_{25}$ has been undertaken. Prior to the measurements, different samples of the alloy of a given composition have been subjected to different heat treatments so as to 'pre-

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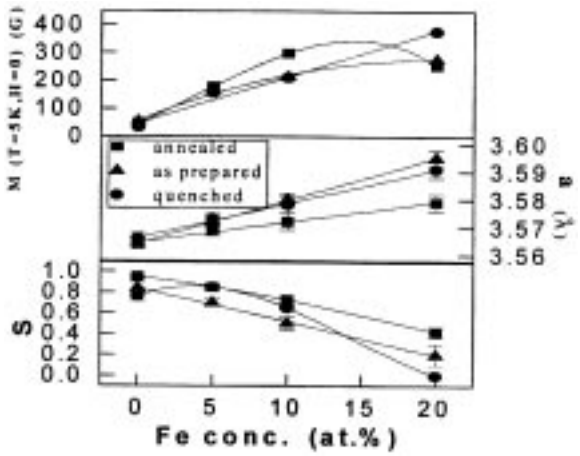

Figure 1. Long-range order parameter *S* (bottom), lattice parameter *a* (middle), spontaneous magnetisation *M* at 5 K (top), as functions of Fe concentration.

pare' them in different states of site disorder. Such a study permits one to ascertain if the thermal history of the sample affects the site preference of Fe.

2. Experimental details

Polycrystalline rods of diameter 10 mm and length 100 mm with the nominal composition stated above were prepared starting from 99.999 pure Ni, Fe and Al by RF induction technique. Several spherical (3 mm in diameter) and disc-shaped (diameter 10 mm and thickness 5 mm) samples were spark-cut from such rods. The spheres and discs that have (have not) undergone annealing at 520°C for 16 days are labelled as the 'annealed' ('as-prepared') samples. A part of the 'as-prepared' rod was melt-quenched to form ribbons of width 2 mm and thickness $\sim$30 μm. Spherical and disc-shaped samples were used for magnetic and X-ray diffraction measurements, respectively. Apart from the (111), (200), (220),... fundamental peaks of the fcc lattice, the diffraction pattern showed a few superstructure peaks such as (100), (110), (210), etc. Magnetisation (M) was measured as a function of external magnetic field (H) in fields up to 70 kOe at $T = 5$ K (a typical example is shown in figure 2) and as a function of temperature in fixed fields $H = 1$ kOe and 10 kOe in the temperature range 5 K $\leq T \leq$ 350 K on SQUID magnetometer.

3. Results, discussion and conclusion

An elaborate analysis of the X-ray diffraction patterns yields accurate values for the atomic long-range order parameter, S (which is a measure of atomic order on Ni or Al sublattice), and the interatomic spacing, a, which are displayed in figure 1. As expected, irrespective of sample history, S decreases with increasing Fe concentration. Effect of annealing is to increase the number of Ni (Al) atoms on the Ni (Al) sublattice and hence for a given Fe

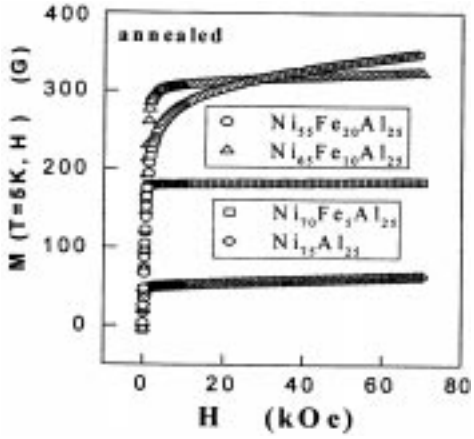

Figure 2. Magnetisation M at 5 K as a function of field H.

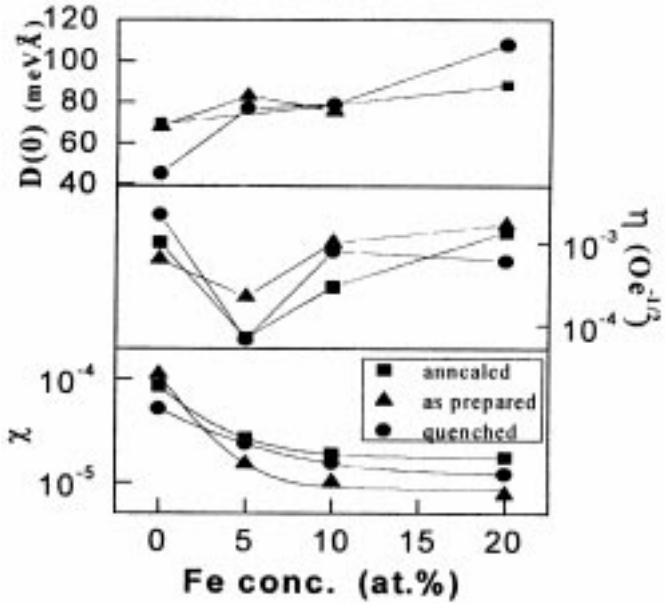

Figure 3. High-field magnetic susceptibility χ (bottom), spin wave suppression parameter η (middle), spin wave stiffness constant at 0 K $D(0)$ (top) as functions of Fe concentration.

concentration, S increases. Considering that ionic radii for Fe, Ni and Al in closed-packed structures are 1.27 Å, 1.25 Å and 1.43 Å, respectively, a linear relation between lattice parameter a and Fe concentration, regardless of sample history (figure 1) strongly indicates that Fe preferentially occupies Ni sites. A substantial reduction in the rate of increase of a with x for the annealed samples compared to that in the 'as-prepared' or 'quenched' states

is thus a consequence of some of the Fe (Al) atoms migrating from Ni (Al) sites to Al (Ni) sites upon annealing.

M versus H isotherms taken at $T = 5$ K have been analysed using the expression $M\ (T = 5\ \mathrm{K}, H) = M\ (T = 5\ \mathrm{K}, 0)\ [1 + \eta\sqrt{H}] + \chi H$, where $M\ (T = 5\ \mathrm{K}, 0)$ is the spontaneous magnetisation at 5 K, η accounts for the suppression of spin waves by H, the high-field susceptibility at 0 K,

$$\chi = N\mu_\beta^2 N(E_\mathrm{F})/(IN(E_\mathrm{F}) - 1),$$

N is the number of spins per unit volume, I is the Stoner parameter and $N(E_\mathrm{F})$ is the density of states at the Fermi level, E_F. The fits based on this expression are shown as continuous curves in figure 2 while the values of $M\ (5\ \mathrm{K}, 0)$, η and χ are displayed in figures 1 and 3. The decline (enhancement) in η with Fe concentration is a clear indication of the decreased (increased) sensitivity of spin waves to H. This inference is consistent with the observed Fe concentration dependence of the spin-wave stiffness at 0 K, $D(0)$ (accurately determined [5] from the $M(T)$ data) (figure 3). Fe substitution leads to the suppression of local spin-density fluctuations (prominent at intermediate temperatures and for temperatures close to Curie point, T_C) and this accounts for the observed enhancement in T_C. Regardless of the thermal history, we find that (figure 3) χ follows the empirical relation $\chi(x) = \chi_0 + A\mathrm{e}^{-\alpha x}$, where x denotes the Fe concentration. This finding implies that both I and $N(E_\mathrm{F})$ increase with x when more and more Fe atoms preferentially occupy the empty Ni sites. The increase in $IN(E_\mathrm{F})$ is, in turn, responsible for the increase in $M\ (5\ \mathrm{K},\ 0)$ with x (figure 1).

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References

[1] M H F Sluiter and Y Kawazoe, *Phys. Rev.* **B51**, 4062 (1995) and references cited therein
[2] A V Ruban and H L Skriver, *Phys. Rev.* **B55**, 856 (1997) and references cited therein
[3] *Ternary alloys, a compendium of evaluated constitutional data and phase diagrams* edited by G Petzow and G Effenberg (VCH Publishers, New York, 1993) vols 4–8
[4] S Pascarelli, F Boscherini, S Mobilio, K Lawniczak-Jablonska and R Kozubski, *Phys. Rev.* **B49**, 14984 (1994) and references cited therein
[5] A Semwal and S N Kaul, *Phys. Rev.* **B60**, 12799 (1999)