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Superparamagnetic behaviour in melt-spun Ni_2FeAl ribbons

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Abstract
Heusler alloy Ni_2FeAl has been synthesized by the melt-spinning technique. The dc magnetization and frequency-dependent ac susceptibility measurements reveal that this alloy exhibits the characteristic feature of superparamagnetism. This behaviour may be associated with a structural disorder stemming from the fast quenching after the heat treatment. The small frequency-dependent ac susceptibility shifts in the blocking temperature and the existence of a pronounced peak in FC magnetization as well as Vogel–Fulcher activation processes indicate that intergranular interactions dominate in the melt-spun ribbons of Ni_2FeAl.

1. Introduction

Heusler alloys are ternary intermetallic compounds of the form X_2YZ, where X and Y are usually two different transition metals and Z is a nonmagnetic metal or nonmetallic element. Heusler alloys have attracted much attention due to their diverse physical properties as well as potential application in the fields of smart materials and spin-electronics. In particular, Ni-based Heusler alloy has been a subject of intense research ever since the observation of a martensitic transition in Ni_2MnGa by Webster [1]. It has been reported that NiMnAl and Ni_2FeGa undergo similar transition and show magnetic shape memory behaviour [2–5]. On the other hand, other Ni-based Heusler alloys, such as Ni_2MnIn, exhibit useful properties

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for applications and are regard as promising materials for the injection of spin-polarized currents into semiconductors. Interestingly, Kurfiß *et al* synthesized an Ni_2MnIn film by the co-evaporation process and observed superparamagnetic behaviour below the Curie temperature [6]. Although superparamagnetism is commonly seen in CuCo series [7, 8], it is rare in the family of Heusler alloys. Up to now, this behaviour has been found only in a few Heusler alloys, including Fe_2VAl, Fe_2VGa, Cu_2CrAl and Fe_2TiSn [9–12]. These materials possess nonmagnetic ground state, while local disorder creates a magnetic moment on Fe sites and leads to the formation of small grains. As the size of the magnetic grains reduces to the nanometre scale, the anisotropy energy E_a of particles can be overcome thermally, leading to the superparamagnetic state. This anisotropy energy has been empirically defined by Stoner and Wohlfarth for non-interacting nanoparticles as

$$E_a = KV\sin^2\theta \tag{1}$$

where K is the anisotropy energy constant, V is the volume of the nanoparticle, and θ is the angle between the magnetization and the easy axis of nanoparticles. Factors such as inter-particle interactions and the non-uniform particle size always make magnetic properties complicated.

Recently, we have substituted Al atoms for the Ga atoms of Ni_2FeGa and successfully synthesized a new Heusler compound Ni_2FeAl by the melt-spinning technique. The static and dynamic properties of Ni_2FeAl ribbon were investigated in detail. Our experimental results reveal the superparamagnetic behaviour associated with a structural disorder in this alloy. However, the small frequency-dependent ac susceptibility shifts in the blocking temperature and the differences between FC and ZFC magnetization imply that intergranular interactions should be considered.

2. Experiment

The sample was prepared by repeated melting of a properly composed mixture of high-purity Ni, Fe and Al metals in an arc furnace with argon atmosphere. Subsequently, the ingots were homogenized by annealing in vacuum at 800 °C for 3 days, and then quenched by spinning onto a copper wheel with an Ar atmosphere protection. The Cu wheel substrate velocity is about 25 $m\,s^{-1}$. X-ray power diffraction has been carried out at room temperature to identify the crystallographic structure. Magnetization and ac susceptibilities were measured by a physical property measurement system (Quantum Design, PPMS). For zero field cooled (ZFC) magnetization measurement, the sample was first cooled from room temperature to 5 K without magnetic field, then a magnetic field was applied during the warm-up process. The magnetization was measured as a function of temperature. Meanwhile, for the field cooled (FC) magnetization measurement the same magnetic field was applied during both cooling and heating processes and the magnetization measurement was carried out on warming up from 5 K to room temperature. Ac susceptibility data were recorded with an excitation field of amplitude 5 Oe at frequencies from 11 to 9999 Hz.

3. Results and discussion

A typical pattern of the x-ray diffraction for the Ni_2FeAl ribbon is shown in figure 1. All diffraction peaks can be indexed to a cubic structure. The ordering of the Ni sublattice is indicated by the presence of the (200) superlattice diffraction peak. But the (111) superlattice peak that indicates the ordering of the Fe and Al is not pronounced, implying that there may

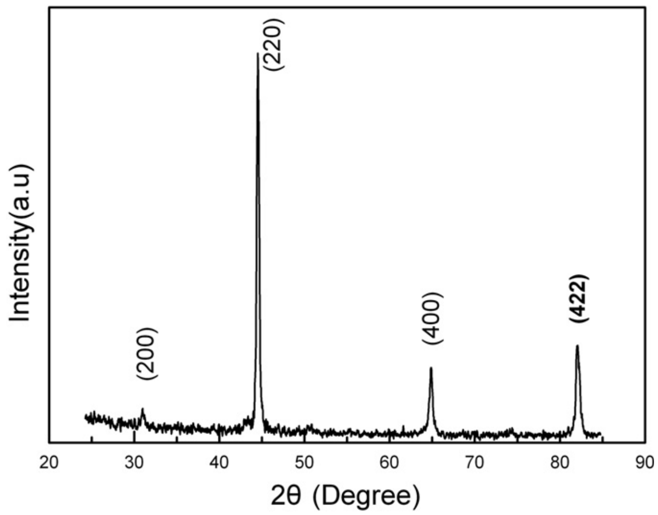

Figure 1. X-ray diffraction spectra for the melt-spun ribbon Ni_2FeAl.

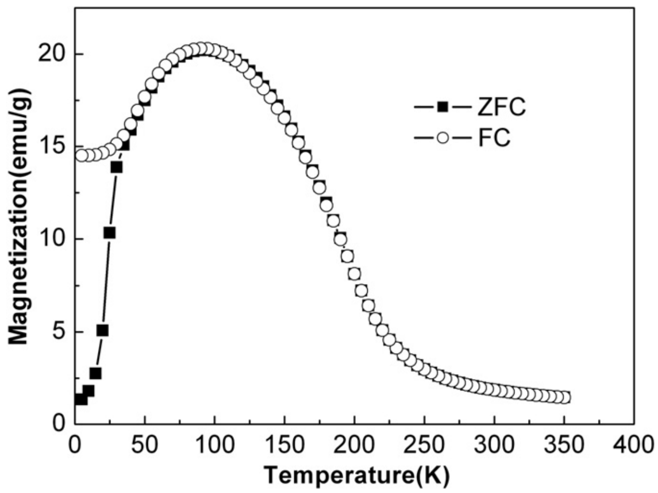

Figure 2. ZFC and FC magnetization curves for Ni_2FeAl measured under the applied field of 100 Oe.

exist some site disorder in the Ni_2FeAl lattice. Indexing all characteristic diffraction peaks, the calculated lattice parameter is 0.547 nm, which is smaller than that of the Ni_2FeGa [4].

Zero field cooled (ZFC) and field cooled (FC) magnetizations were recorded for applied magnetic fields of 100 Oe. As shown in figure 2, the magnetic behaviour of the N_2FeAl ribbons shows a ZFC magnetization maximum at about 95 K. Below 95 K, ZFC magnetization continuously decreases to the lowest measurement temperature, ∼5 K, whereas the FC magnetization exhibits similar behaviour and overlaps with the ZFC magnetization at a certain temperature. The observed maximum and irreversibility suggest the blocking of superparamagnetic moments below the blocking temperature, which is the usual phenomenon for a fine magnetic particle system [13]. However, spin-glass systems also display features similar to the ones described above and it seems necessary first to classify our Ni_2FeAl ribbons.

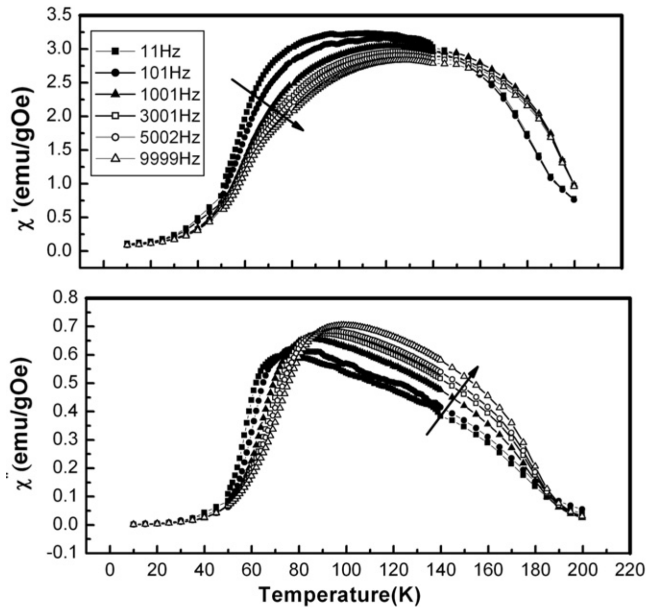

Figure 3. Temperature dependence of the in-phase (χ') and out-of-phase (χ'') ac susceptibilities for Ni_2FeAl ribbons, at different excitation frequencies. Arrows indicate increasing frequencies.

The temperature dependences of ac susceptibilities of the Ni_2FeAl ribbons were then measured for different frequencies f ranging from 11 to 9999 Hz at an excitation field of 5 Oe. As one can see from figure 3, a broad peak with an 11 Hz maximum is present at $T_{max} \approx 112$ K for the in-phase component (χ') and at $T_{max} \approx 75$ K for the out-of-phase component (χ'') and the peak position shifts towards higher temperatures with increasing frequency. A criterion which can be used to distinguish the freezing/blocking process is the relative shift of the temperature of the maximum in $\chi'(T)$, T_m, with the measuring frequency f, as

$$\Phi = \frac{\Delta T_m}{T_m \Delta \log_{10}(f)} \tag{2}$$

where ΔT_m is the difference between T_m measured in the $\Delta \log_{10}(f)$ frequency interval. Experimentally, the Φ values found for non-interacting superparamagnetic systems are in the range ~0.10–0.13, whereas a much smaller dependence of T_m on f is observed in spin glasses ($\Phi \sim 0.005$–0.05) [14, 15]. In our case, we obtained about 0.04 for Ni_2FeAl. The value is of the same order of magnitude as that of insulating spin glass $Eu_{0.4}Sr_{0.6}S$ [16]. However, a relatively low Φ value can also be accounted for within the superparamagnetism by allowing a certain degree of interaction between the clusters [17].

In order to further ascertain the occurrence of a real thermodynamic transition in Ni_2FeAl ribbons, we analysed the temperature dependence of the nonlinear susceptibility χ_{nl} according to the Sherrington–Kirkpatrick mean-field model [18]. Owing to a possible non-Curie–Weiss behaviour of χ_0, it is necessary to consider a development in terms of $\chi_0 H$ instead of H to avoid an overestimate of the temperature dependence of the non-linear susceptibility [19]. The divergence disappears for temperature below $T \approx 1.5T_{max}$. Accordingly,

$$M = \chi_0 H - b_3(\chi_0 H)^3 + b_5(\chi_0 H)^5 + \cdots. \tag{3}$$

The initial nonlinear susceptibility χ_0 could be obtained from the slope $(\mathrm{d}M/\mathrm{d}H)_{H\to 0}$ of the isothermal magnetization curves deduced from the field-cooled data (figure 4). The

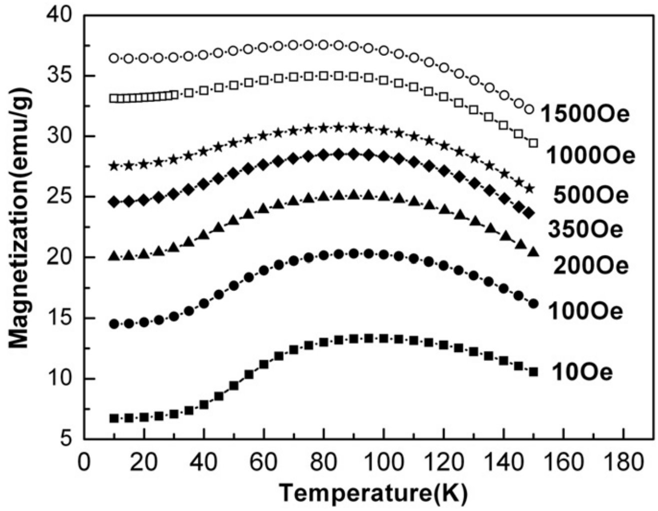

Figure 4. The field-cooled magnetization as a function of the temperature for Ni_2FeAl.

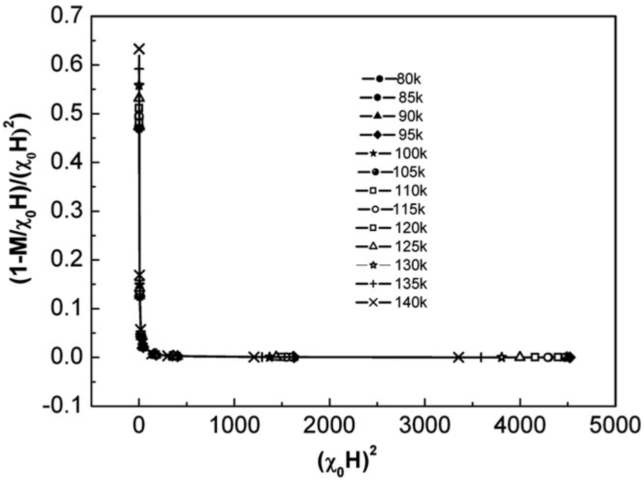

Figure 5. $(1 - M/\chi_0 H)/(\chi_0 H)^2$ as a function of $(\chi_0 H)^2$ for different temperatures.

coefficients b_3 and b_5 are calculated from the intercept with the ordinate axis and initial slope of $(1 - M/\chi_0 H)/(\chi_0 H)^2$ versus $(\chi_0 H)^2$ plots, respectively. As can be seen from figure 5, these curves superimpose for temperature ranging from 80 to 140 K, implying that b_3 and b_5 are temperature independent. This observation eliminates the existence of a spin-glass transition.

The origins of the susceptibility maximum can also be related to mixed ferromagnetic and antiferromagnetic interactions. In such a case, a clear shift in the field-cooled hysteresis loop below T_{max} disappears at temperatures above T_{max}. However, no shift at all was observed in the hysteresis (figure 6), therefore we ruled out this possibility.

The small value of Φ for Ni_2FeAl ribbons is then most likely to be due to magnetic interactions between superparamagnetic blocking moments. The relaxation process of grain moments for Ni_2FeAl provides further support for this point. The linear fit of the data to the Arrhenius law $\ln(\tau/\tau_0) = E_a/(k_B T_{max})$ for Ni_2FeAl ribbons gives an unrealistically large

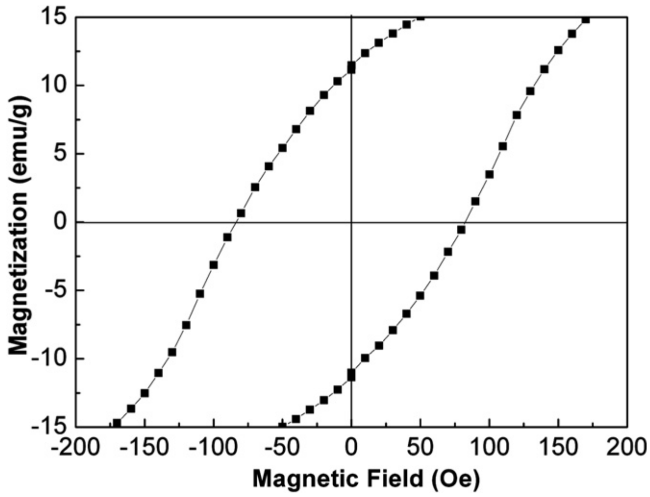

Figure 6. Magnetization versus applied field at 20 K after field cooling.

value for E_a/k_B (7119 K) as well as unphysical pre-exponential factors (3.8×10^{-29} s), suggesting that the relaxation process is not governed by the simple blocking of each magnetic moment but is rather strongly influenced by a cooperative mechanism related to the presence of intergranular magnetic interactions [20]. Another manifestation of the existence of intergranular interactions emerges from the aforementioned thermomagnetic measurements. A peak in the FC curve can occur as intergranular interactions (essentially of dipolar type) dominate over the magnetocrystalline anisotropy, as predicted by Chantrell *et al* [21]. This argument has been confirmed experimentally by Greaves *et al* [22]. Our results appear to fit the model very well.

To quantify the effect of the interactions, it is possible to apply the phenomenological Vogel–Fulcher model

$$\tau = \tau_0 \exp\left[\frac{E_a}{k_B(T - T_0)}\right] \tag{4}$$

where T_0 has been introduced as an additional parameter with respect to the Arrhenius law representing the temperature for which the relaxation diverges and can be the measure of the intergranular interactions. When a physically realistic value of $\tau_0 = 10^{-11}$ s is selected a reasonable fit is obtained with a realistic value of $E_a/k_B \approx 742$ K and $T_0 \approx 81$ K, as shown in figure 7. This behaviour clearly confirms that the intergranular interactions significantly influence the variation of τ against T.

As discussed above, the magnetic properties of melt-spun Ni_2FeAl are significantly different from those of Ni_2FeGa [5]. The possible origin of superparamagnetic behaviour in Ni_2FeAl lies in the presence of a high level of disorder in the samples resulting from the fast quenching. Accordingly, the structural disorder seems to be the main reason behind the anomalous behaviour observed in Ni_2FeAl.

4. Conclusions

The granular Heusler alloy Ni_2FeAl has been successfully synthesized by the melt-spinning technique. The static and dynamic properties of Ni_2FeAl ribbon having a cubic structure resemble those observed in canonical spin glasses. However, the non-divergence of the

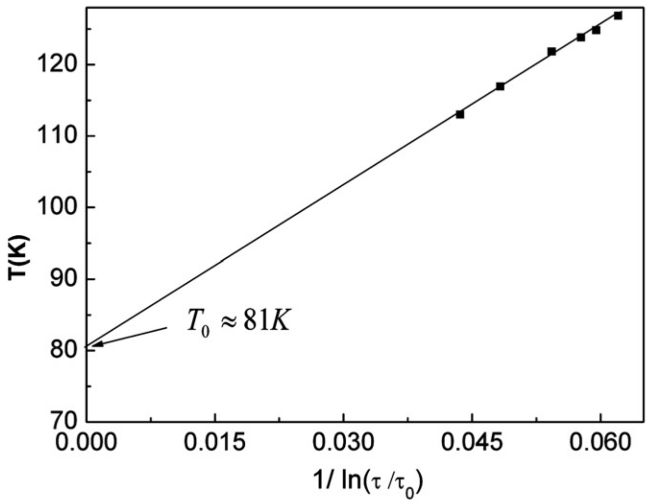

Figure 7. The plot of T_{max} versus $1/\ln(\tau/\tau_0)$ with $\tau_0 = 10^{-11}$ s; the solid line represents the fit to the Vogel–Fulcher model.

nonlinear magnetization at T_{max} excludes the existence of a thermodynamic phase transition. The observed features were proved to stem from a progressive blocking of the moments of superparamagnetic grains. The appearance of this behaviour may be associated with the presence of structural disorder. Comparison with the typical superparamagnetic behaviour, however, highlights that intergranular interactions are not negligible. This point is confirmed by observing the dynamic behaviour of the susceptibility peak corresponding to a Vogel–Fulcher model and a pronounced peak in the field-cooled magnetization.

Acknowledgments

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