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Premartensite transition in Ni_2FeGa Heusler alloy

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ABSTRACT

Martensitic phase transformation of Ni_2FeGa Heusler alloy was studied by differential scanning calorimetry. Atomic ordering induced in the austenite structure by quenching from high temperature plays a significant role on martensitic phase transformation. Higher magnetization and larger magneto-crystalline anisotropy of martensite phase than that of austenite phase are noticed. Tweed contrast regions observed in the transmission electron microscopy were correlated to premartensite phenomena. A shift in premartensitic transition temperature prior to martensitic transformation as measured by differential scanning calorimetry is being reported for the first time in this system.

1. Introduction

In the last decade, many literatures have been published on Ni–Fe–Ga Heusler alloys. They show promising characteristics for magnetic shape memory applications. The presence of γ-phase along with the austenite phase has been reported in Ni_2FeGa Heusler alloys [1–3]. The γ-phase improves ductility and hot deformation behavior in Ni–Fe–Ga Heusler alloys [2]. Qian et al. [3] showed that the traces of γ-phase embedded in the matrix of single crystal Ni_2FeGa generate residual stress and result in anisotropic two way shape memory effect. HRTEM images of typical modulated structures (5 M and 7 M) were reported elsewhere [4,5]. Modulated martensite structures generate large amount of magnetic field induced strain in Heusler alloys. Liu et al. [6] reported microtwins within martensite lamellae which are induced due to high internal stress in the vicinity of grain boundaries. High stacking fault energy coupled with low transformational activation energy leads to the martensite transformation with little irreversibility [7]. Martensite transformation and shape memory properties have been studied by Huang et al. [8]. Complete pseudoelastic recovery of 5% was obtained at 0° intersection angle between loading direction and grain boundary of highly oriented polycrystalline Ni–Fe–Ga Heusler alloys. The homogeneous strain exhibited by low temperature martensite phase was assumed to be linearly coupled with micromodulation of the phase [9]. Thus, Ni–Fe–Ga system is chosen as it has the potential for future shape memory application. This article focuses on martensite transformation and related phenomena in Ni_2FeGa Heusler alloy.

Though several authors have studied the origin of modulated structures in Ni–Mn–Ga system [9–11], the literatures on the origin of modulated structures in Ni_2FeGa Heusler alloys are limited. The dynamic instability towards low temperature transition generates modulation in the structure due to lattice shuffling and is associated with premartensitic transition [12]. Premartensitic transformation in $Ni_{51.5}Fe_{21.5}Ga_{27}$ single crystal has been analyzed using phonon dispersion, elastic diffusion scattering and TEM [5,12]. Premartensite transition prior to martensite transformation exhibits the lattice dynamic effect in Ni_2FeGa Heusler alloy. The coupling of vibrational and magnetic degrees of freedom is quite essential to generate large magnetic field induced strain. In this paper we discuss the appearance of premartensitic transition in Ni_2FeGa Heusler alloy using differential scanning calorimetry (DSC) and transmission electron microscopy. Both DSC and TEM results have been correlated to explain the premartensitic transition. The effect of multiple thermal cycling on premartensitic transition and martensite transformation has been also studied.

2. Materials and methods

Ni_2FeGa Heusler alloy was prepared from high purity (99.99%) Ni, Fe and Ga elements by choosing the alloy composition close to stoichiometry X_2YZ, but with higher Ni-content. This was done hoping that Ni substituting Ga would increase the transition temperature to near room temperature. The alloy sample preparation is similar to an earlier study [13]. Initially the furnace was evacuated up to 3×10^{-5} mbar and backfilled with argon to maintain inert atmosphere. The alloy was cast into button shape (approximately 4 g) using chilled copper hearth. It was melted four times, each time flipping the sample, to promote compositional homogeneity. The alloy is then annealed at 1273 K for 1 h and quenched in water. Phase identification and microstructural characterization were

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carried out using X-ray diffraction (XRD), scanning electron microscopy (SEM) and TEM. XRD was used as the primary technique for phase identification. The microstructure and compositions of different regions were analyzed using SEM and its energy dispersive spectroscopy attachment. The structure of the phases present in the alloy was confirmed independently using electron diffraction in the TEM studies. Structural phase transition was characterized by DSC at heating/cooling rate of 10 °C/min in the temperature range of −125 °C to +100 °C (148 K–373 K). Magnetic properties were studied by VSM both below and above transition temperature.

Table 1
Elemental compositions of Ni_2FeGa Heusler alloy and of the respective phases present in the microstructure.

Composition	Ni	Fe	Ga
Ni_2FeGa Heusler alloy	52	25	23
γ-Phase	52	32	16
Austenite	52	21	27

All the elemental compositions are given in at.% taken from SEM–EDX analysis.

3. Results and discussion

The microstructure of Ni_2FeGa Heusler alloy studied by SEM is shown in Fig. 1c. It consists of two phases, the dark contrast gamma phase is dendritic and the light contrast austenite is the interdendritic phase. The composition of the alloy and the corresponding phases are given in Table 1. The phase separation and their evolution were discussed in an earlier study on the same alloy system [1]. The structures are confirmed by XRD (Fig. 1a, b) as fcc (γ-phase) and $L2_1$ (chemically ordered) austenite phases. The γ-phase has disordered fcc structure and its presence in microstructure is known to improve the ductility [2] — a desirable property of Ni_2FeGa Heusler alloys. After annealing and quenching from 1273 K, the high angle peaks of austenite are visible. This confirms the improvement in chemical ordering in $L2_1$ structure. Annealing does not affect the morphologies of the phases present in the microstructure. The as-cast alloy did not undergo martensitic transformation (see Fig. 1a). Martensite transformation in Heusler like ferromagnetic shape memory alloys depends on the ability of high temperature austenite phase to undergo structural transition. The evolution of chemical ordering in $L2_1$ structure after annealing and quenching from 1273 K (Fig. 1b) modifies the electronic and magnetic properties of austenite phase aiding in a martensitic transformation. The increase in martensitic transformation temperature due to evolution of atomic ordering has also been reported elsewhere [14–16].

TEM bright-field image of both the phases along with their phase interface and the corresponding diffraction patterns are shown in Fig. 2. The electron diffraction patterns were taken from the respective phases on either sides of the phase boundary and confirm the phases to be fcc and $L2_1$ ordered structures, respectively. The TEM images captured only from the austenite grain (Figs. 2b and 3) show the presence of localized microtwins as tweed structure. In the magnified view, the pattern of the tweed structures is clearly visible (Fig. 3d) as a grid-like network of strain fields. Tweed patterns observed in TEM (Figs. 2, 3) are regions of distorted lattice due to localized atomic displacement. These tweed regions are inhomogeneous, highly anisotropic and could act as elastic scattering centers [17]. The presence of tweed patterns around dislocations and stacking faults (Figs. 2 and 3) confirms the role of these defect interactions in formation of precursor effect prior to martensitic transformation. The diffuse spots around the main Bragg reflecting spots are quite prominent (Fig. 3b) and are attributed to the tweed.

The structural phase transformation was studied by differential scanning calorimetry (DSC) and shown in Fig. 4. Different cooling and heating cycles were performed (up to 8 in number) to study the reproducibility of the martensitic transformation. DSC curves in each cycle show two reversible peaks, one of them appears while cooling and the other while heating the sample that corresponds to the first order martensitic transition. While cooling it shows exothermic peak corresponding to the release of energy due to the transformation from high temperature austenite phase to low temperature phase martensite. The reversible transformation takes place while heating and the endothermic peak is at a higher temperature indicating transformation hysteresis. The hysteresis is due to the elastic strain energy stored during forward structural transformation. Additionally, two small peaks at $T_{p1} = 273$ K and $T_{p2} = 265$ K prior to martensite transformation were also observed (Fig. 4) while cooling the sample in DSC experiment in the 1st cycle in the temperature range of 148 K–373 K and these disappear during the heating portion of the cycle.

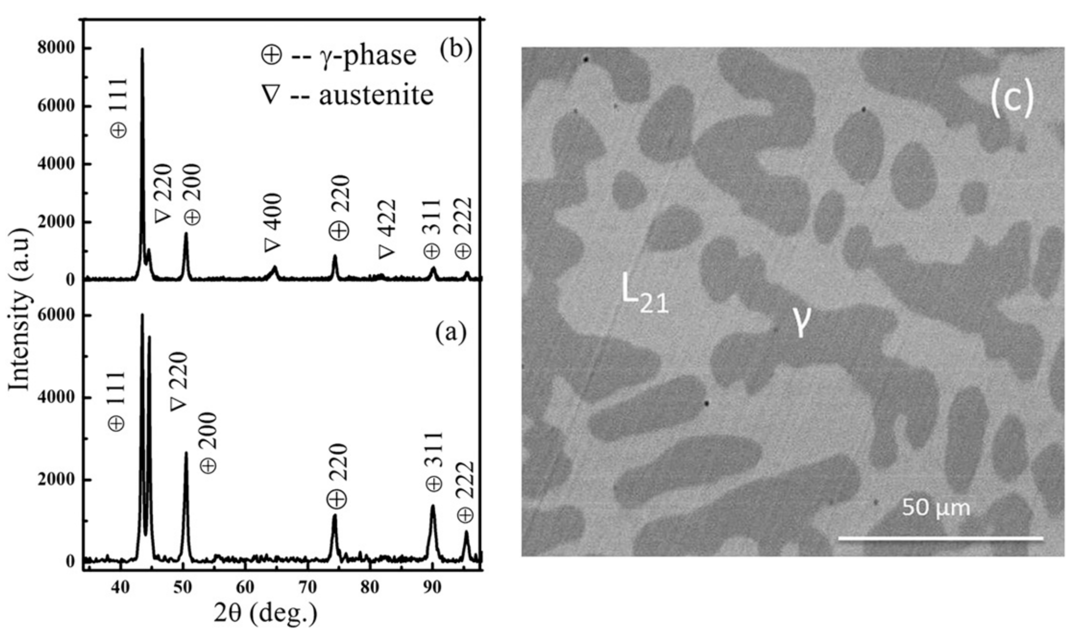

Fig. 1. X-ray diffraction pattern of Ni_2FeGa Heusler alloy (a) as cast, (b) after heat treatment at 1273 K, (c) SEM back scattered electron image shows the morphologies of both γ-phase and austenite (marked as $L2_1$).