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Ferromagnetic Resonance Observation of Martensitic Phase Transformation in Ni-Mn-Ga Ferromagnetic Shape Memory Films

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Polycrystalline Ni-Mn-Ga films have been deposited onto mica substrates held at 720 K by flash-evaporation method. At room temperature the films have a tetragonal structure with $a = b = 0.598$ and $c = 0.576$ nm typical for bulk Ni_2MnGa below a martensitic transformation. Temperature measurements of ferromagnetic resonance reveal a martensitic phase transformation at 310 K. The transformation brings about a substantial decrease in the effective magnetization and a drastic increase in the ferromagnetic resonance linewidth due to a strong increase in the magnetic anisotropy in the martensitic phase.

Key words: ferromagnetic shape memory alloys, thin films, ferromagnetic resonance

1. Introduction

Heusler alloy Ni_2MnGa is a typical ferromagnetic shape memory (FSM) alloy showing a large strain actuated by a magnetic field [1]. Since thin films of FSM alloys are promising candidates for magnetic field driven microactuators, much effort has been put on the single-crystalline Ni_2MnGa films grown using molecular beam epitaxy (MBE) on carefully prepared single-crystalline substrates [2, 3]. On the other hand, there is an interest in achieving a large strain-induced actuation in textured polycrystals [4], and an extension of the results to the polycrystalline films would markedly increase the possible application capabilities of FSM thin films as microactuators. We have already reported the magnetic properties of Ni_2MnGa films deposited on glass substrates [5], but to realize FSM effect in polycrystalline thin films it is essential to deposit them on flexible substrates [2] and to set up a high magnetic anisotropy below the martensitic transformation (MT) [6]. In this paper we show that it is possible to produce on flexible substrates the polycrystalline Ni_2MnGa films which exhibit a clear MT and a substantial magnetic anisotropy in the martensitic phase.

The 100~200 nm thick Ni_2MnGa films were deposited by flash-evaporation of the alloy powders onto heated up to 720 K mica substrates in a vacuum better than 5×10^{-5} Pa. For evaporation we used the powders obtained from a carefully prepared $Ni_{0.493}Mn_{0.247}Ga_{0.26}$ ingot. The structural characterization of the Ni_2MnGa was carried out by x-ray diffraction (XRD) using Cu Kα radiation using grazing-incidence and out-of-plane Bragg Θ-2Θ scans. The grazing-incidence XRD patterns consist of a strong and split (220) reflections and the (210), (321) and (400) smaller peaks. The (220) reflection is the most intensive. All other peaks in the complete grazing-incidence x-ray scan suggest a random orientation of crystallites in the film plane and some (110) texture around the film normal. Figure 1 shows that the splitting of the (220) reflection in the normal Θ-2Θ Bragg scan corresponds to the (202) and (220) reflections of the tetragonal [7] martensite phase with $a = b = 0.598$ nm and $c = 0.576$ nm. In conclusion, our films with a tetragonal structure ($c/a = 0.96$) are different from the single-crystalline films deposited by MBE [2] on GaAs ($c/a = 1.08$) and similar to Ni_2MnGa films grown on the silicon substrates by ion-beam sputtering [8] with broadly distributed (110) axes around the film normal.

Using a simple experimental setup (Fig. 2-inset), we measured a temperature variation of the deflection angle of a laser beam from a partially released Ni_2MnGa film. During a transition, stress-induced actuation of the partially released film results in an abrupt change in the

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*Corresponding author: e-mail: dubowik@ifmpan.poznan.pl angle of deflection. We confirmed a clear sign of thermoelastic anomaly at temperature of ≈ 315 K, a characteristic of the austenite/martensite transformation in some bulk off-stoichiometric Ni-Mn-Ga alloys (see, e.g., Ref. [9]). However, we did not observed any FSM effect in the martensitic phase in fields up to 10 kOe. This might be due to the polycrystalline microstructure of our films with broadly oriented crystallites.

The magnetic properties of our Ni_2MnGa films were investigated using a ferromagnetic resonance (FMR) spectrometer operating at 9.08 GHz at temperatures from 78 K to 400 K in a magnetic field applied perpendicular and parallel to the film plane. Figure 3 shows the temperature dependence of the saturation magnetization $4\pi M_S$ measured in a field of 10 kOe, with a Faraday balance and a vibration sample magnetometer (above 280 K). The Curie temperature, estimated to 390-400 K is slightly higher than the temperature reported [7] for the bulk stoichiometric cubic $L2_1$ phase (376 K). The extrapolated (to 0 K) $4\pi M_S \approx 6000$ G is comparable to our recent data [5] for polycrystalline Ni_2MnGa films deposited on glass substrates and to the single-crystalline films [2].

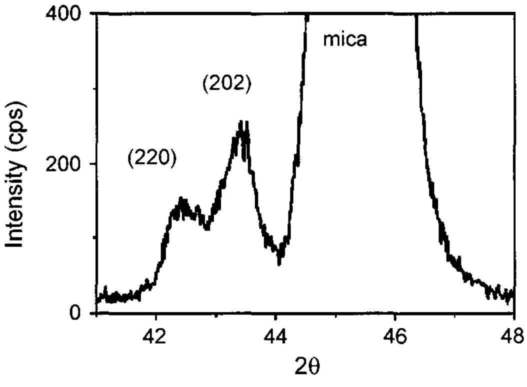

Fig. 1. XRD Θ-2Θ Bragg scan of the Ni_2MnGa film deposited on (001) mica substrate held at 720 K.

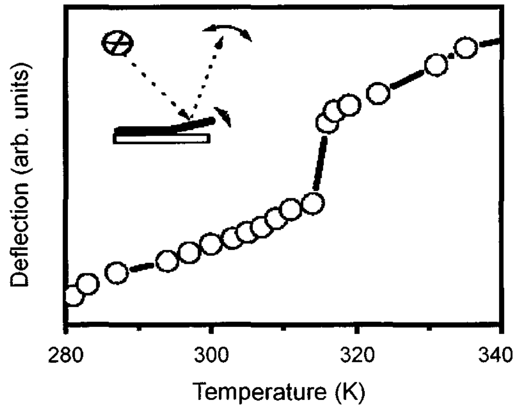

Fig. 2. Thermoelastic anomaly detected in a partially released film of Ni_2MnGa. Inset is a sketch of an experimental setup.

The values of $4\pi M_{eff}$ were determined from the resonance fields H_{perp} taken in the perpendicular configuration at various temperatures; $\omega/\gamma = H_{perp} - 4\pi M_{eff}$, where $\omega = 2\pi f$ is a microwave angular frequency and γ is the gyromagnetic ratio corresponding to a g-factor of 2.2 [10]. It is clearly seen from Fig. 3 that below 310 K $4\pi M_{eff}$ is substantially smaller than $4\pi M_S$ with a characteristic anomaly in $4\pi M_{eff}$ around 300-310 K. The anomaly resembles an anomaly in the magnetic-field-induced strain for the bulk polycrystalline Ni_2MnGa [11]. The difference between $4\pi M_S$ and $4\pi M_{eff}$ of 2.2 kG at 310 K results from a strong effective anisotropy field in the martensitic phase: $H_{eff} = 2\ K_{eff}/M_S$ and $4\pi M_{eff} = 4\pi M_S - H_{eff}$, where K_{eff} is the effective anisotropy energy. The estimated K_{eff} at 310 K is about 4×10^5 erg/cm^3 that is 5 times smaller than that reported for a single crystal of Ni_2MnGa (1.5×10^6 erg/cm^3 at 317 K) [12]. We interpret this weaker magnetic anisotropy to be due to a specific microstructure of our films. As inferred from the XRD measurements, the Ni_2MnGa films are polycrystalline with a (110) texture. Therefore, the easy c-axes of crystallites are broadly distributed around the film normal. Additionally, below MT the crystallites are divided into twin variants what makes the distribution of the c-axes even broader. Hence, in accordance with the observed magnetoelastic effects in bulk Ni_2MnGa polycrystals [11] and by assuming that the in-plane strains are fixed owing to a strong film-substrate interaction, we argue that the anomaly in $4\pi M_{eff}$ is due to a spontaneous stress accompanying MT in polycrystalline films with a random

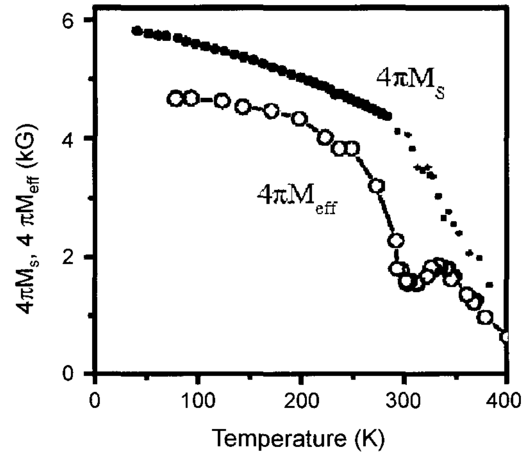

Fig. 3. Temperature dependence of the saturation magnetization $4\pi M_S$ (full dots) and the effective magnetization $4\pi M_{eff}$ (open circles) of Ni_2MnGa films deposited on mica.

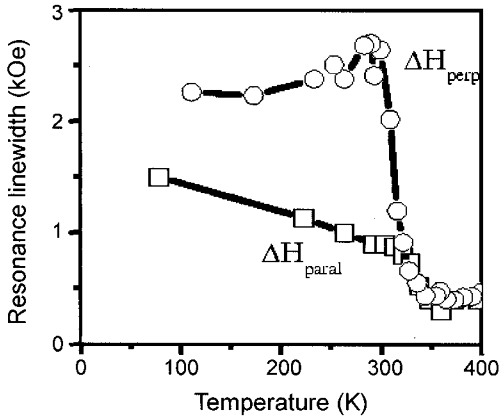

Fig. 4. Temperature dependence of the FMR linewidth for the in-plane ($\Delta H_{\mathrm{paral}}$) and the out-of-plane ($\Delta H_{\mathrm{perp}}$) configurations.

microstructure. A simple estimation for the stress-induced anisotropy $3/2\lambda_S\Delta\sigma$ with $\lambda_S \approx 200\times10^{-6}$ [11] and a reasonable spontaneous stress $\Delta\sigma$ of 100 MPa gives a similar value of K_{eff} of $\approx 3\times10^5$ erg/cm^3.

The effect of MT in the FMR linewidth (defined as a peak- to-peak linewidth of the first derivative of FMR absorption) is even more pronounced than the anomaly in $4\pi M_{\mathrm{eff}}$ (Fig. 4). The linewidth ΔH_{perp}, determined in the perpendicular configuration, experiences a similar jump of about 2 kOe at the transformation temperature T_M = 310 K, as reported recently in a single crystal Ni_2MnGa [13]. However, in contrary to Ref. [13], we relate the increase in ΔH_{perp} and $\Delta H_{\mathrm{paral}}$ to an increase in the magnetic anisotropy rather than a difference in the magnetization between martensite and austenite. As was shown in our detailed analysis [14] of the angular dependencies of the linewidth, the anomalous broadening of the FMR spectra below MT can be interpreted in terms of a spread of the anisotropy easy axes and a broad distribution of the internal effective anisotropy fields. It supports the presented interpretation that the anomalous FMR response below MT results from the interplay between the microstructure of the Ni_2MnGa and the strong magnetic anisotropy.

In summary, the polycrystalline ferromagnetic Ni_2MnGa films grown on heated mica substrates show the tetragonal structure below the MT at ≈ 310 K. The transformation leads to a specific anomaly in the effective magnetization $4\pi M_{\mathrm{eff}}$ and in the FMR linewidth.

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