Ferromagnetic resonance in Ni–Mn–Ga films

S. I. Patil,[a] Deng Tan,[b] S. E. Lofland,[c] and S. M. Bhagat
Ferromagnetic Resonance Group, Department of Physics, University of Maryland, College Park, Maryland 20742

I. Takeuchi,[d] O. Famodu, J. C. Read, K.-S. Chang, C. Craciunescu, and M. Wuttig
Department of Materials Science and Engineering, University of Maryland, College Park, Maryland 20742

(Received 3 May 2002; accepted for publication 25 June 2002)

Textured thin films of nominal composition $Ni_{0.50}(MnGa)_{0.50}$ were sputter deposited on Si substrates and studied by x-ray diffraction, micromechanical displacement, dc magnetization, and ferromagnetic resonance (FMR). We report the observation of spin wave resonances in this alloy, yielding a spin wave stiffness of $D=200$ meV Å^2 at 300 K. A marked thermal hysteresis is observed in the temperature-dependent FMR data arising from the reversible martensitic transition. [DOI: 10.1063/1.1501161]

Ni_2MnGa and related alloys are magnetic shape memory alloys (MSMAs) which exhibit extremely large magnetic-field induced strains.[1] Ferromagnetic domains in these materials are identical with the tetragonal variants in the martensitic state, and the field-induced twin boundary motion in the martensite can give rise to strains as large as 6% in applied fields of ~5 kOe.[1] One important direction of research in MSMAs is the fabrication of thin films, which hold potential for sensor and actuator devices with applications in microelectromechanical systems. In general, these thin films display properties which are rather different from those of bulk because their mechanical properties are greatly affected by their geometry, microstructure, and the constraint from the substrate.[2] There is a subtle interplay between the magnetic and structural properties that dictates their overall mechanical behavior, and understanding these relationships is of paramount importance in realizing the potential of these materials.

Ferromagnetic resonance (FMR) is one of the most powerful methods for probing the magnetic quality of thin films. In particular, narrow FMR lines and the observation of well resolved spin wave resonances (SWR) imply a high degree of magnetic homogeneity. In this letter, we present a FMR study of Ni–Mn–Ga alloy films grown by magnetron sputtering on Si substrates. In the perpendicular geometry (magnetic field $H\perp$ film plane), we have observed FMR linewidths (full width at half maximum) as narrow as 70 Oe, about an order of magnitude smaller than those reported for bulk polycrystalline samples of Ni–Mn–Ga.[3] In addition, in the films with thickness ≈200 nm, we have observed up to eight SWR modes and made the first determination of the temperature dependence of the spin wave stiffness. In thicker films (>500 nm), the temperature dependent FMR displays pronounced thermal hysteresis associated with the reversible martensitic transition.

Ni–Mn–Ga films were deposited by rf magnetron sputtering on Si (100) substrates in a high-vacuum chamber with a base pressure of $\sim 5\times 10^{-9}$ Torr.[4] During the deposition, Ar gas at $\sim 5\times 10^{-3}$ Torr was introduced in the chamber and controlled via throttling. The nominal starting composition of the target was Ni_2MnGa. Stoichiometric deviation from the target composition is expected in the deposited films, and wavelength dispersive spectroscopy was performed on individual films to obtain the exact composition. Measured chemical compositions are given in Table I. The films were either deposited at elevated temperature up to 500 °C or they were deposited at room temperature followed by an *in situ* annealing in the chamber at temperatures up to 500 °C. The film thickness ranged from ~150 to ~1000 nm. We also deposited relatively thick films (~1 μm) on micromachined Si cantilevers for detection of martensitic transitions. X-ray diffraction of the films at room temperature indicated that the films were textured with the peak from the (110) orientation of the austenite phase being the predominant feature.[4] The ferromagnetic transition temperature was determined to be near 370 K for all the films; which is the Curie temperature for bulk Ni_2MnGa.[3]

FMR was measured using a conventional homodyne spectrometer operating at 9.87 GHz, in the temperature range of 77–400 K. The applied field H was rotated in the plane perpendicular to the film.

We first discuss the room temperature data. Referring to Table I, films A and B are comparatively thin, <200 nm, and

TABLE I. Room temperature characteristics of thin films ($g=2$), Eqs. (1) and (2).

Sample ID	$H_{\parallel}$ (kOe)	$H_{\perp}$ (kOe)	H_{an} (kOe)	$4\pi M_{eff}$ (kOe)	$\Delta H_{\perp}$ (Oe)	Composition
A	1.64	8.70	0.10	5.40	100	—
B	1.28	9.58	0.27	6.35	70	$Ni_{0.50}Mn_{0.33}Ga_{0.17}$
C	1.68	7.04	0.40	3.90	600	$Ni_{0.54}Mn_{0.31}Ga_{0.15}$
D	1.85	7.17	0.20	3.90	300	$Ni_{0.50}Mn_{0.30}Ga_{0.20}$
E	1.27	9.82	0.24	6.60	300	$Ni_{0.49}Mn_{0.36}Ga_{0.15}$

[a] Also at: Dept. of Physics, University of Pune 411007, India; electronic mail: patil@glue.umd.edu
[b] Intern from: Eleanor Roosevelt High School, Greenbelt, MD 20770.
[c] Also at: Center for Materials Research and Education, Dept. of Chemistry and Physics, Rowan University, Glassboro, NJ 08028.
[d] Also at: Center for Superconductivity Research, University of Maryland, College Park, MD 20742; electronic mail: takeuchi@squid.umd.edu

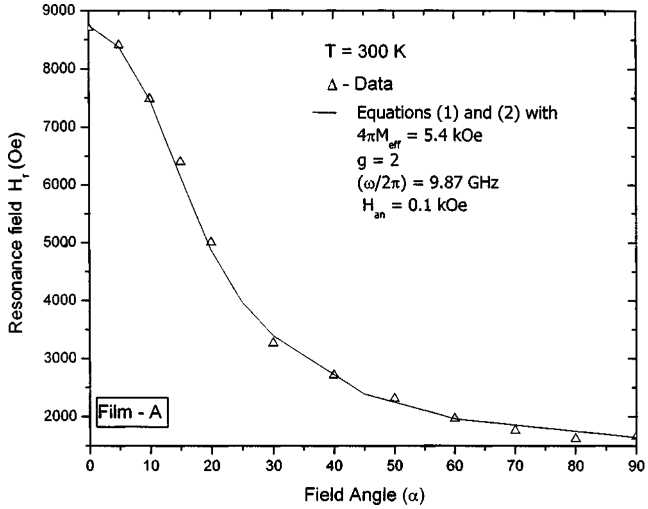

FIG. 1. Angular dependence of resonance field at $T = 300$ K in film A. The full line was obtained using Eqs. (1) and (2) with the parameter values given in Table I.

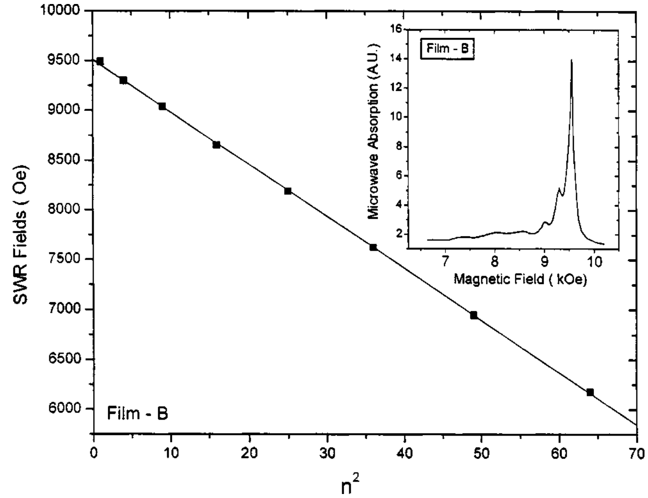

FIG. 2. Spin wave resonance fields for film B at $T = 300$ K vs n^2. The inset shows the SWR pattern for the same film.

show a narrow FMR line with $\Delta H_{\perp} < 100$ Oe in the perpendicular geometry ($H \perp$ film plane). The other three films are ~ 1 μm thick and $\Delta H_{\perp} \sim 300-600$ Oe. All the films studied were found to show significantly wider resonance lines (>700 Oe) for $H \|$ to the film plane. Next, the angular dependence of the resonance field (H_r) was used to establish that the magnetization was uniform and that the strain-induced anisotropy field had a symmetry axis along the film normal. Namely, H_r is given by the equation

$$\left(\frac{\omega}{\gamma}\right)^2 = (H_r \cos\alpha - 4\pi M_{\mathrm{eff}} \cos\theta)^2 + H_r \sin\alpha (H_r \sin\alpha + 4\pi M_{\mathrm{eff}} \sin\theta) \quad (1)$$

combined with the equilibrium condition

$$\frac{\sin(\theta - \alpha)}{\sin\theta \cos\theta} = \frac{4\pi M_{\mathrm{eff}}}{H_r}, \quad (2)$$

where $\omega = 2\pi f$, f is the frequency, γ is the gyromagnetic ratio, and θ (α) measures the angle between the $\mathbf{M}$ ($\mathbf{H}_r$) and the film normal.

In order to understand the data for all the films using a single value of g (2.00), it was necessary to introduce a small anisotropy field H_{an} to augment H_r. As before,[5] this is a field-induced anisotropy with the easy axis aligned parallel to H. As seen in Fig. 1, Eqs. (1) and (2) provide an excellent fit to the data, showing that the film is a fairly homogeneous ferromagnet. The room temperature parameter values for different films are listed in Table I, and it is notable that the lines are among the narrowest observed in Ni_2MnGa, again attesting to the high "magnetic quality" of the film. While vibrating sample magnetometer measurements yield $4\pi M(300\text{ K}) \cong 8 \pm 0.5$ kOe, the values for $4\pi M_{\mathrm{eff}} = 4\pi M + 2K_u/M$ are significantly lower, thereby indicating that the strain-induced anisotropy field $2K_u/M$ is negative and 1–2 kOe in magnitude, i.e., the "easy" axis is out of plane.

Note that H_{an} is necessary to keep $g = 2$ for every film. Otherwise, the calculated g values show unphysically large changes from film to film. As usual, $2K_u/M$ is needed to account for the difference between $4\pi M_{\mathrm{eff}}$ and the dc magnetization ($4\pi M$).

In the thinner films (films A and B), several spin wave resonances were observed (inset of Fig. 2). The resonance fields follow the n^2 dependence expected for Kittel modes (albeit with both even and odd n) in a homogeneous ferromagnetic film, namely

$$\frac{\omega}{\gamma} = H + \frac{D}{\gamma\hbar}\left(\frac{n\pi}{L}\right)^2 - 4\pi M_{\mathrm{eff}} + H_{\mathrm{an}}, \quad (3)$$

yielding a spin wave stiffness of $D(300\text{ K}) = 200$ meV Å^2 (full line in Fig. 2) for $L = 1800$ Å. Although the temperature interval is rather narrow (250–350 K) it is notable that the temperature dependence of D follows $D = D_0\ (1 - CT^{5/2})$, with $D_0 = 300$ meV Å^2 and $C = 3 \times 10^{-7}$ K$^{-5/2}$, as shown in Fig. 3. Thus we have demonstrated that the low-lying excited states are spin waves and that the stiffness renormalizes with temperature in accord with the simple spin wave theory. There are no neutron scattering data available for the present alloys. In the Heusler alloys D_0 values of 100–200 meV Å^2 have been reported and the temperature dependence follows

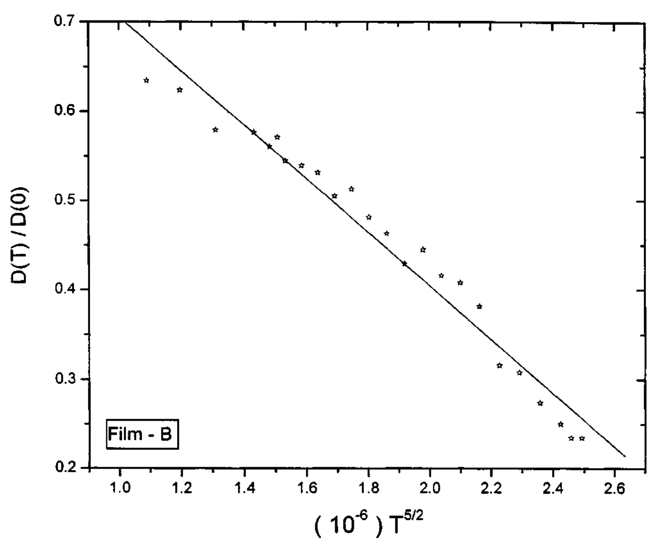

FIG. 3. Normalized spin wave stiffness constant as a function of $T^{5/2}$.

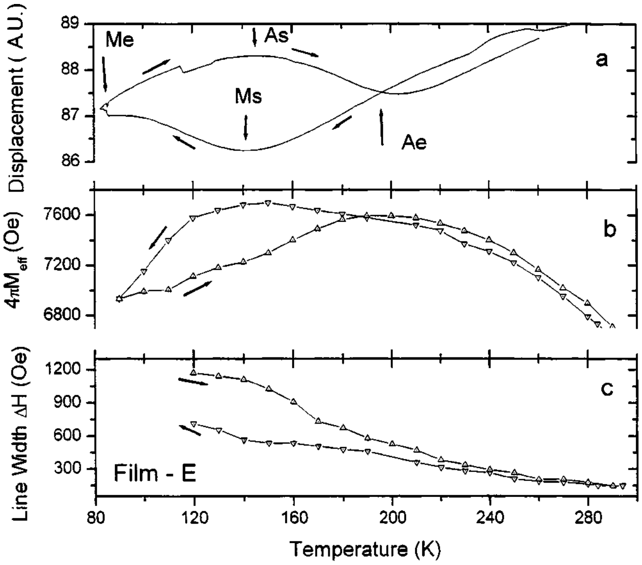

FIG. 4. The dependence of (a) micromechanical displacement, (b) effective magnetization, and (c) FMR linewidth on temperature for film E.

the spin wave theory.[6] However, the present D values are exceptionally large for a material with $T_C \sim 370$ K.

Figures 4(a)–4(c) show the temperature dependence of the micromechanical displacement and the FMR characteristics. Figure 4(a) represents the result of a thermomechanical measurement of a ~ 1 μm film (film E) deposited on a micromachined Si cantilever. Thermally activated displacement of the cantilever is monitored using a capacitance formed between the cantilever and a separate electrode.[7] A clear thermal hysteresis loop is observed indicating the occurrence of a reversible martensitic transformation in the film. Martensite start and end and austenite start and end temperatures are marked in the figure as M_s, M_e, A_s, and A_e, respectively. The observed transition behavior is very similar to that of previously reported Ni–Mn–Ga films.[8] Figures 4(b) and 4(c) show the temperature dependence of the resonance parameters $4\pi M_{\mathrm{eff}}$ (derived from $H_\perp$ using $g=2$) and $\Delta H_\perp$ for the same sample. The hysteresis is also very apparent in the FMR data, and the hysteresis end points agree with the points of the multiphase region [M_e and A_e in Fig. 4(a)] quite well. In addition, when the martensite phase is dominant, the linewidth is much larger; as would be expected from a state with many martensitic variants. Indeed, for $T < 130$ K the FMR line becomes severely distorted [hence, the arrows in Fig. 4(c)]. Also, during warming, $4\pi M_{\mathrm{eff}}$ is significantly lower, indicating that the (negative) strain-induced field $(2K_u/M)$ is enhanced by several hundred oersted. However, the onsets at A_s, M_s are not too obvious in the FMR data except that, on cooling, $4\pi M_{\mathrm{eff}}$ begins to drop rapidly for $T \leqslant M_s$.

To conclude, using FMR we have demonstrated that one can grow thin films of $Ni_{1-x-y}Mn_xGa_y$ that are magnetically very homogeneous. We have determined the spin wave stiffness ($D_0 = 300$meV Å^2) and shown that it renormalizes as $T^{5/2}$. The martensite transition is well delineated by the resonance data in agreement with micromechanical experiments.

This work was supported by ONR N000140110761 and N00014010503, NSF under the MRSEC Grant Nos. DMR-00-80008, DMR0114176, and the New Jersey Commission on Higher Education.

[1] S. J. Murray, M. Marioni, S. M. Allen, and R. C. O'Handley, Appl. Phys. Lett. **77**, 886 (2000).
[2] A. Ishida and V. Martynov, MRS Bull. **27**, 111 (2002).
[3] B. D. Shanina, A. A. Konchits, S. P. Kolesnik, V. G. Gavriljuk, I. N. Glavatskij, O. Soderberg, V. K. Lindroos, and J. Foct, J. Magn. Magn. Mater. **237**, 309 (2001).
[4] I. Takeuchi, O. Famodu, and J. C. Read (unpublished).
[5] D. J. Webb, and S. M. Bhagat, J. Magn. Magn. Mater. **42**, 109 (1984).
[6] U. Stuhr, P. Vorderwisch, and V. V. Kokorin, Physica B **234**, 135 (1997).
[7] M. Wuttig, C. Craciunescu, and J. Li, Mater. Trans., JIM **41**, 933 (2000).
[8] C. Craciunescu, Y. Kishi, L. Saraf, R. Ramesh and M. Wuttig (unpublished).