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Epitaxial Growths and Magnetization Dynamics of Ni_2MnSn Heusler Alloy Films

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Ferromagnetic resonance has been investigated in Ni_2MnSn Heusler alloy films. The films were deposited at $673 < T < 723$ K on MgO(001) substrates by means of magnetron sputtering. X-ray diffraction confirmed that the films were epitaxial. The films had magnetic parameters typical of bulk Ni_2MnSn with $L2_1$ structural order. From angular dependences of the resonance field and the ferromagnetic resonance linewidth the intrinsic Gilbert contribution and extrinsic contribution to the ferromagnetic resonance linewidth due to two-magnon scattering were separated. Our data indicate a room temperature Gilbert damping of 0.007, which is comparable to that of half-metallic Heusler alloy. Surface roughness is responsible for a substantial extrinsic two-magnon contribution to the linewidth of the order of 50-300 Oe in these epitaxial soft magnetic Heusler alloy films. The spin wave stiffness of $D = 70-100$ meVÅ^2 is one of the lowest for the known Heusler alloy.

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1. Introduction

The Heusler alloys (HA) are regarded as attractive materials for spintronic devices since some of them exhibit half-metallic behavior [1]. In particular, half-metallicity has stimulated great interest aiming at achieving a high tunnel magnetoresistance (TMR) in magnetic tunnel junction (MTJ). For the best epitaxial electrodes in all HA Co_2MnSi, a record value of TMR of ≈ 1000% at 4.2 K has been demonstrated [2]. On the other hand studies on spin dynamics in half-metallic materials are also current topics since small magnetization precessional damping may be an advantage for reduction in critical current density in current-induced magnetization switching [3]. Usually the Gilbert damping constant α describing the intrinsic damping is low in HA [4]. Recently a damping constant of 0.0023 has been reported for NiMnSb films [5]. A very low value of α of 0.003 has been reported and explained in terms of half-metallic behavior of $Co_2Fe_xMn_{1-x}Si$ with $x = 0.4$ [3]. Assuming that, according to Kambersky [6], the spin-orbit coupling ξ and the density of states (DOS) at the Fermi level $D(E_F)$ are low, the intrinsic damping $\alpha \propto \xi^2 D(E_F)$ is expected to be small in half-metallic HA with zero or negligible DOS for the minority spin band. However, even in not half-metallic HA, such as Co_2FeAl the intrinsic damping is very low (of 0.001) [7] and therefore half-metallicity may not be the cause of a low intrinsic damping in some HA.

The aim of the present contribution is to determine both intrinsic and extrinsic contributions to the ferromagnetic resonance (FMR) linewidth and to evaluate the Gilbert damping constant in Ni_2MnSn epitaxial films, which exhibits no half-metallic behavior according to several first-principle calculations (e.g. [8]).

2. Experimental and results

Thin films were prepared by magnetron sputtering on single crystalline Cr-buffered MgO(001) substrates by means of co-sputtering from Ni, MnNi, and MnSn targets at 623–673 K. Base pressure was better than $\approx 10^{-10}$ mbar and Ar pressure was 2×10^{-4} mbar. The composition of the films — close to stoichiometric — was controlled by means of the choice of optimal deposition rates from individual targets and confirmed with X-ray fluorescence analysis. Structural characterization of the films was performed by X-ray diffraction (XRD) with Co K_α radiation. Magnetization measurements were carried out using a home-made ferromagnetic resonance spectrometer and broad frequency FMR-VNA equipment with Agilent Vector Network Analyzer.

The results of structural characterization are shown in Fig. 1. Figure 1a shows $\Theta-2\Theta$ scan of XRD for a film MgO(100)/Cr(20 nm)/Ni_2MnSn(40 nm). In addition to peaks from MgO substrate, only the (002) Cr and the (002) and (004) reflections of Ni_2MnSn were detected indicating a good (001) epitaxial growth. In addition, the ratio of the intensities of the (200) and (400) reflections, which can be the measure of the degree of structural order [9], is of 0.28. This implies a good site ordering of Ni and Mn atoms. Lattice constant of $a = 0.600$ nm estimated from these peaks was nearly equal to $a = 0.605$ nm for a bulk alloy. A ϕ-scan (not shown) of the reflection from the (200) planes for the epitaxial Ni–Mn–Sn film reveals shift of 45° from those for (200) peaks of the MgO substrate. From these data, the epitaxial relationship for the films was confirmed to be $Ni_{50}Mn_{25}Sn_{25}$(001)[100]/MgO(001)[110]. On the other hand, our epitaxial films deposited at elevated tempera-

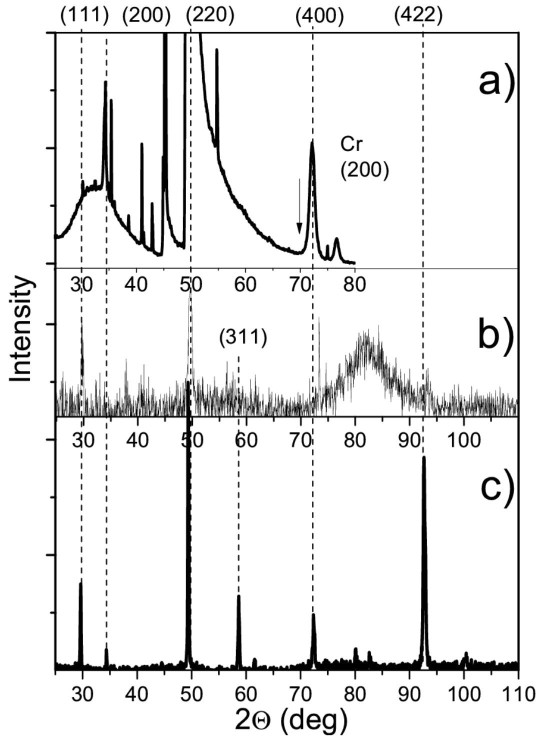

Fig. 1. X-ray diffraction patterns of a $Ni_{50}Mn_{25}Sn_{25}$ epitaxial film on a MgO substrate with Cr (20 nm) buffer (a), and a $Ni_{50}Mn_{25}Sn_{25}$ on a Si substrate (b). Part (c) shows XRD pattern of bulk Ni_2MnSn with $L2_1$ ordering.

tures are rather rough with a roughness of 1–3 nm estimated by grazing incidence X-ray diffraction. Figure 1b shows XRD of the polycrystalline film B deposited on Si(100) at 623 K with the presence of the reflections (111), ((200), (311), (220) and (422) from B2 (or $L2_1$) Heusler alloy structure. Both (111) and (200) reflections present in the case of fully ordered $L2_1$ suggest that the polycrystalline NiMnSn films - deposited on Si substrates at the same conditions as those on MgO - are well ordered. For XRD $\Theta-2\Theta$ scans the samples deposited on Si were tilted of 1–15° with respect to a diffractometer stage to get rid of a very strong Si(400) reflection but some diffused reflection between 70–80° is still seen. XRD of a reference bulk Ni_2MnSn with typical $L2_1$ HA ordering is shown in Fig. 1c for comparison.

The out-of-plane angular dependence of the ferromagnetic resonance linewidth ΔH in field-swept (with the microwave frequency ω kept constant and a varied applied field H) FMR experiments has often been analyzed in order to obtain some additional information on the intrinsic contribution to the linewidth ΔH^{Gilb} due to the Gilbert damping and on the extrinsic one $\Delta H^{2-\mathrm{mag}}$ resulting from two-magnon scattering due to structural inhomogeneity and defects present in thin magnetic films. We have recently shown that these two contributions to the field-swept FMR linewidth of thin films can be separated [10].

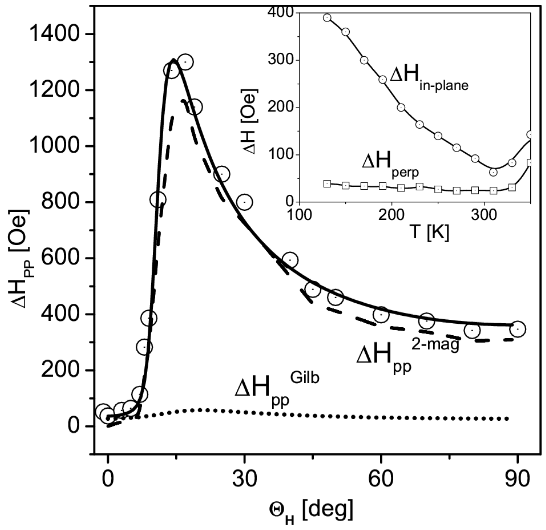

Fig. 2. Peak-to-peak FMR linewidth ΔH_{pp} as a function of the angle Θ_H of the epitaxial Ni_2MnSn. The data were taken at 160 K. The open circles show the data and the solid curve show the results of computations. The dotted and dashed curves show the intrinsic Gilbert $\Delta H_{\mathrm{pp}}^{\mathrm{Gilb}}$ and the extrinsic $\Delta H_{\mathrm{pp}}^{2-\mathrm{mag}}$ contributions to the linewidth, respectively. The inset shows temperature dependence of ΔH_{perp} and $\Delta H_{\mathrm{in-plane}}$ measured at $\Theta_H = 0°$ and $\Theta_H = 90°$, respectively.

Figure 2 shows the peak-to-peak FMR linewidth ΔH_{pp} (at $T = 160$ K) as a function of the angle Θ_H between the applied field and film normal Θ_H (open circles) of an epitaxial Ni_2MnSn film 180 nm in thickness and a fit (full line) according to our model [10] with the following fitting parameters: the microwave frequency $f = 9.2$ GHz, the spectroscopic splitting factor $g = 2.06$, the effective magnetization $M_{\mathrm{eff}} = 590$ G, the magnetocrystalline anisotropy field $H_K = 2K_4/M = -150$ Oe and the magnetocrystalline anisotropy $K_4 = -4.4 \times 10^4$ erg/cm^3. The Gilbert damping constant $\alpha = 0.009$ and the Gilbert relaxation rate $G = \alpha\gamma M = 106$ MHz values are comparable to that of permalloy.

The most characteristic feature seen in Fig. 2 is that at 160 K the intrinsic Gilbert contribution to the linewidth $\Delta H_{\mathrm{pp}}^{\mathrm{Gilb}}$ is of the order of magnitude lower than the extrinsic two-magnon contribution $\Delta H_{\mathrm{pp}}^{2-\mathrm{mag}}$. Since in our NiMnSn films the out-of plane $\Delta H_{\mathrm{pp}}(\Theta_H = 0°)$ is mainly determined by the Gilbert damping (two-magnon scattering is inoperative at the perpendicular configuration) and the in-plane linewidth $\Delta H_{\mathrm{pp}}(\Theta_H = 90°)$ is mostly governed by two-magnon contribution the inset in Fig. 2 clearly shows that the intrinsic damping weakly depends on temperature (e.g. at 300 K $\Delta H_{\mathrm{pp}}(\Theta_H = 0°) = 25$ Oe and $\alpha = 0.0075$ or $G = 50$ MHz) while the extrinsic damping substantially decreases with the increase in temperature (e.g. at 300 K $\Delta H_{\mathrm{pp}}(\Theta_H = 90°) = 80$ Oe). Since roughness may add significant linewidth in thin films [11] due to two-magnon scattering, we argue that the high roughness of 1-3 nm of our expitaxial films is

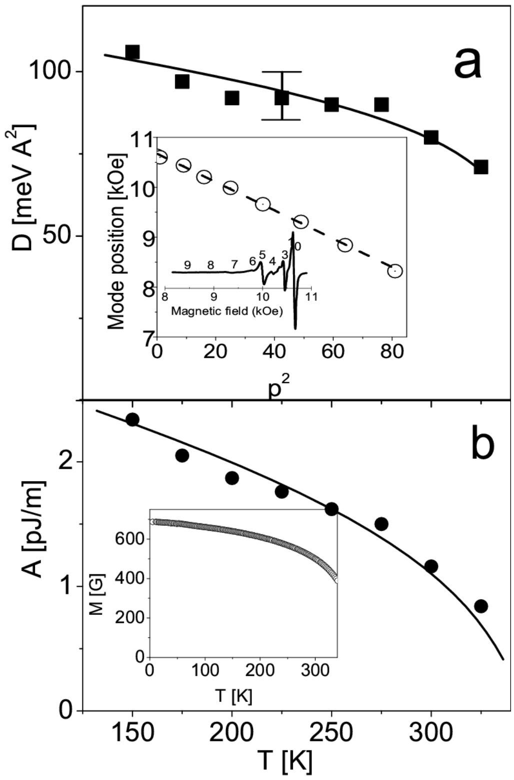

Fig. 3. (a) Temperature dependence of the spin-wave stiffness D of the 180 nm Ni_2MnSn film. The inset in (a) shows mode positions of standing spin waves taken at 9.2 GHz with the applied field perpendicular to the sample plane. (b) Temperature dependence of the exchange constant A for the same film. The inset in (b) shows temperature dependence of magnetization M measured at 20 kOe of the same film.

the main source of such a high extrinsic contribution to the observed linewidth. To sum up, our epitaxial Ni_2MnSn films are magnetically soft (the magnetocrystalline anisotropy K_4 is of -1.5×10^4 erg/cm^3 at room temperature) and have the intrinsic damping comparable to other HA films [5], while the two-magnon contribution is several times higher than the intrinsic one.

All the films thicker than 30 nm displayed multiple modes due to spin wave resonance (see the inset in Fig. 3a). This feature allows us to determine the exchange constant A and the spin wave stiffness $D = 2A/M$ and their temperature dependences. As it is shown in Fig. 3a D relatively weakly depends on T in agreement with neutron spectroscopy studies of a similar bulk Ni-Mn-Ga HA [12]. D values of about 70 to 100 meVÅ^2 are in agreement with theoretical calculations (150 eV/Å^2 at 0 K) [13] and are low in comparison with that of the known HA [4]. Figure 3b shows temperature dependence of A which roughly scales as $M(T)$ (see the inset in Fig. 3b and attains 50 pJ/m, which is one of the lowest value for the known HA [5].

3. Conclusions

We have investigated Ni_2MnSn Heusler alloy films. Structural characterization of the films was performed by XRD. XRD confirmed that the films were epitaxial with the lattice parameter $a = 0.600$ nm and Ni_2MnSn(001)[100]||MgO(001)[110] relationship. The films had the saturation magnetization $M_S(4\,K) = 690$ G (4 μ_B per formula unit) typical of bulk Ni_2MnSn with $L2_1$ structural order. Our FMR study of these films indicates that they have at room temperature a low intrinsic magnetization damping of 0.007 and a low magnetocrystalline anisotropy of K_4 of -1.5×10^4 erg/cm^3 both comparable to that of other HA films with half-metallic behavior. However, due to the high roughness the extrinsic two-magnon contribution to the linewidth is several times higher than the intrinsic contribution. The spin wave stiffness values of about 70–100 meVÅ^2 are consistent with the values evaluated from the first-principle calculations.

Acknowledgments

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