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FREQUENCY DEPENDENCE OF FMR AND THE STRUCTURE OF COMPOSITIONALLY MODULATED COBALT MANGANESE FILMS

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FMR absorption measurements have been carried out on compositionally modulated films prepared by electron beam evaporation in $\approx 5\times10^{-10}$ Torr and nominally consisting of 20 layers of pure cobalt interleaved with 20 layers of pure manganese. Detailed FMR results are reported at 9, 24, 37 and 73 GHz using a particular film for which the variation of composition with depth was investigated by means of a scanning Auger microprobe. Each cobalt layer had a thickness of (24 ± 5) Å, and each manganese layer had a thickness of (36 ± 5) Å: the total thickness of the film was estimated to be 1200 Å. A well-defined compositional periodicity was resolved for the 8–10 layers near the glass substrate: this periodicity gradually disappeared and was unobservable in the top 5–6 layers.

The magnetic measurements indicated an isotropic homogeneous magnetic material characterized by a saturation magnetization $4\pi M_s = (11.80\pm0.05)$ kOe, a value $g = 2.12\pm0.01$, and a Landau–Lifshitz damping parameter $G = (3.4\pm0.5)\times10^8$ Hz. This magnetic material is believed to be a fcc cobalt alloy containing 13–14% manganese.

1. Introduction

Several laboratories [1,2[are studying compositionally modulated magnetic films in order to investigate the magnetic excitations which such films can support. Of particular interest are the modes which periodic structures can support when the magnetic layers are coupled by dipole interactions [3] and/or by exchange interactions [4]. Such modes have been studied both by means of Brillouin light scattering experiments [3] and by means of ferromagnetic resonance experiments [5] (FMR).

In this note we report the results of FMR studies, and of structural studies using Auger spectroscopy, on a specimen consisting of 20 layers of cobalt, nominally 24 Å thick, alternating with 20 layers of manganese, nominally 36 Å thick. FMR measurements were carried out at 9.16, 23.83, 36.95 and at 73.03 GHz and for various orientations of the static magnetic field with respect to the plane of the samples. The use of several frequencies enabled us to check the consistency of the results and also to obtain a good measure for the Landau–Lifshitz magnetic damping parameter from the variation of FMR linewidth with frequency.

2. Experimental details

The specimens were prepared by sequential evaporation of cobalt and manganese metals using two electron guns installed in an ultrahigh vacuum system ($\approx 10^{-10}$ Torr). The metals were alternately evaporated, beginning with cobalt, onto a 1 cm diameter Corning glass substrate which had been pre-coated with 200 Å of manganese. The substrate temperature was maintained at ap-

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proximately 20°C by means of water cooling. During evaporation the pressure in the system increased typically to $4–8 \times 10^{-9}$ Torr. Thicknesses of the metal layers were estimated by means of a quartz crystal thickness monitor: the cobalt layers were approximately 24 Å thick and the manganese layers were approximately 36 Å thick. Absolute thicknesses were known within ±5 Å, but relative thicknesses were known within ±2 Å. The total thickness of the specimen was approximately 1200 Å. After the FMR measurements had been completed, the depth profile [6] of the film was measured in ultra-high vacuum ($\approx 10^{-10}$ Torr) using a 3 keV xenon ion beam nearly at oblique incidence ($\approx 12°$ to the plane of the specimen). The composition of the film was continuously monitored using the LMM Auger lines for cobalt, manganese, and oxygen *. Detailed depth profile experiments were carried out for only one film – a typical film. Only one film was measured because:

(1) the depth-profiling experiment is very tedious – a great deal of effort and multiple trials were required to properly align the electron beam probe within the sputtered crater;
(2) xenon ions were used in order to achieve maximum depth resolution – however, xenon ions tend to poison the vacuum pumps on the scanning Auger microprobe system.

All the evidence we have, including FMR measurements, magnetization measurements, and NMR measurements, indicate that the results reported here are typical.

FMR absorption measurements were carried out at 9, 24, 37 and 73 GHz using a conventional field modulation technique in which the signal obtained was proportional to the field derivative of the microwave absorption in the specimen [7]. The specimens were lightly clamped over a suitable orifice in the microwave cavities so that the microwave energy was incident on the metal film from only one side (single-sided excitation). Absorption derivatives were measured as a function of angle as the static magnetic field orientation was varied from parallel to perpendicular to the plane of the specimens.

* This analysis was carried out by means of a PHI Model 595 Scanning Auger Microprobe. Physical Electronics Division, Perkin-Elmer Corp., Eden Prairie, MN 55343, USA.

Magnetic fields were obtained using a Varian V-3800 electromagnet equipped with a Varian V-7900 regulated power supply *. Field values were obtained from the dial readings of the power supply: they are known to be accurate to ±100 Oe from previous calibration experiments using proton NMR.

Frequencies were measured using standard, calibrated, frequency meters which are accurate to better than 1 part in 1000 except for the highest frequency for which the OKI CFM-753 LA frequency meter is stated by the manufacturer to be accurate to 1 part in 500.

All FMR measurements were carried out at room temperature, approximately 20°C.

3. Results

3.1. Depth profile

The variation with sputtering time, and hence with depth into the specimen of the strength of the cobalt, manganese and cobalt Auger lines is shown in fig. 1. For the first 80 min of the depth-profiling, the Auger lines were generated by rastering the ≈ 0.3 μm diameter beam spot over a 20 μm by 20 μm area using a 0.02 μA electron beam current. For the last 60 min the electron beam current was increased to 0.06 μA, and rastering was stopped so that the Auger lines were generated from an area approximately 0.6 μm in diameter. Sputtering conditions were maintained constant during the depth-profiling measurements: only the operating conditions of the electron beam Auger probe were altered to improve the signal to noise ratio in the Auger signals. Several of the observed features are of interest:

1. The average strength of the cobalt Auger signal is nearly three times greater than the average strength of the manganese signal within the bulk of the specimen. According to the tables supplied by the manufacturer of the scanning Auger microprobe, the Auger sensitivities of manganese and cobalt are the same to within a few percent. We interpret the relatively weak

* Varian Associates, Palo Alto, California, USA.

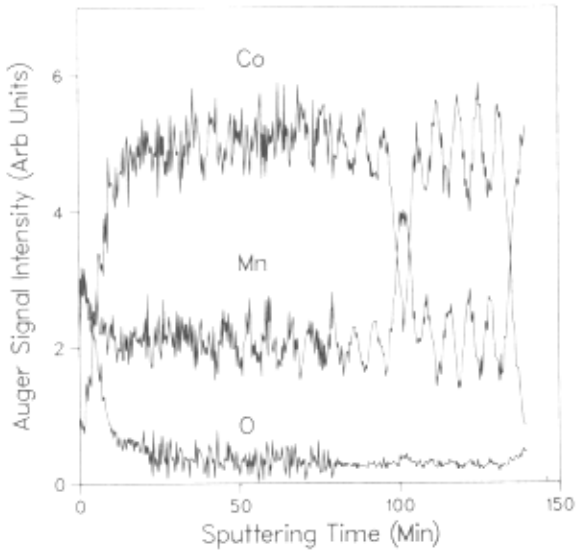

Fig. 1. Variation of the intensity of Auger LMM lines for cobalt, manganese and oxygen vs. sputtering time for the compositionally modulated film reported in this work. This depth profile was obtained using a 3 keV beam of xenon ions incident upon the specimen at approximately 12° to the plane. For the first 80 min the auger spectra were obtained using an electron beam current of 0.02 μA rastered over an area 20 μm × 20 μm. For the last 60 min the electron beam was not rastered, and the electron beam current was increased to 0.06 μA to improve the signal to noise (the sputtering conditions were maintained constant). Nominal thicknesses of Co and Mn were 24 and 36 Å, from which a sputtering rate of 8.9 Å per minute was deduced. The amplitude of the oxygen line in the interior of the specimen corresponds to zero oxygen within experimental error. Note the break in the periodicity of the signals near 100 min where the manganese deposition was continued longer than was planned!

strength of the manganese signal to mean that the manganese sputtering rate was 2–3 times larger than the cobalt sputtering rate so that the material exposed in the sputtered crater was cobalt rich. This interpretation is supported by the relatively large amplitude of the manganese signal near the substrate which was covered with a thick layer of manganese.

2. Near the front surface of the specimen the Auger signals do not show any compositional periodicity. However, near the substrate the compositional periodicity is well-resolved, and one can estimate from that periodicity an average sputtering rate of 8.9 Å per minute.
3. It is estimated that the depth resolution was approximately 30 Å because of the nearly sinusoidal variation of the strength of the cobalt and manganese Auger lines. If the resolution depth had been small compared with the layer thicknesses (24 and 36 Å), the Auger amplitude should have exhibited a more nearly square wave amplitude with time. It is known that depth-profile resolution is limited to 20–40 Å by ion-beam induced cascade mixing [6].
4. The amplitude of the oxygen Auger signal fell to a low level after sputtering for 12 min. Within experimental uncertainty, this low level corresponded to no oxygen within the bulk of the specimen.

The Auger scans of fig. 1 were supplemented by Auger scans across the depth-profiling crater and by images of the crater taken using the instrument in the scanning electron microscope mode. The layered structure of the specimen was clearly revealed in the image of the 1 mm diameter crater, and spatial Auger scans across the walls of the crater resembled very closely the time variation of the Auger amplitudes shown in fig. 1: i.e. oscillating amplitudes with the Co and Mn signals 180° out of phase. Moreover, the contrast between the various layers was very clear in the deepest part of the crater, but the contrast gradually weakened towards the outer edges of the crater corresponding to layers near the surface of the specimen. There can be little doubt that the gradual decrease of the modulation envelope towards earlier times (i.e. towards the surface) shown in fig. 1 means that the composition modulation, when averaged over a beam spot diameter – approximately 0.6 μm – was very well defined near the specimen substrate but became undetectably small in the upper 1/3 of the specimen thickness.

3.2. FMR studies

The absorption spectra generally consisted of a strong peak plus a weaker satellite peak shifted approximately 700 Oe to lower field values. Additional smaller peaks were observed at 9.16 GHz with the field normal to the specimen plane. At 9.16 GHz and at 24 GHz both the main and subsidiary peak could be observed over the entire

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angular range as the field was rotated from a direction perpendicular to the specimen plane to a direction parallel with the specimen plane, except for a narrow angular interval between approximately 10° and 20° from the normal where line broadening due to magnetic dragging effects made it difficult to observe any but the main absorption peak. The sensitivities of our 37 and 73 GHz systems were less than those for the lower two frequencies, and the FMR linewidth increases with frequency, so that the subsidiary resonance peak could not be observed with the dc field parallel with the specimen plane. The angular variation of the resonance fields as the external field was rotated in the specimen plane was found to be less than 100 Oe. This specimen therefore exhibited very little anisotropy in the plane.

The variation of the resonance fields with angle

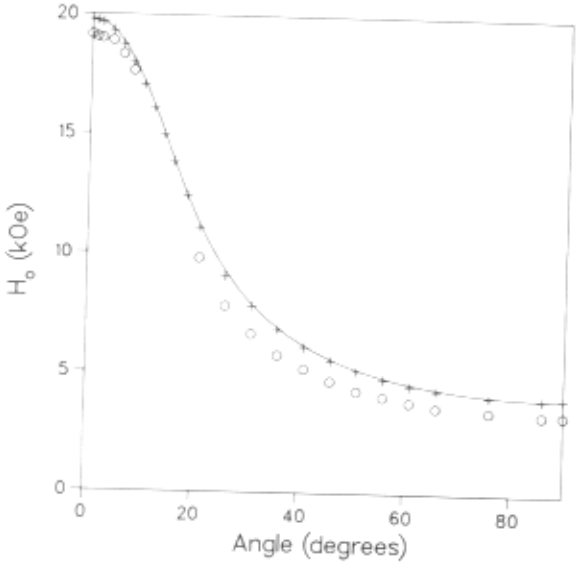

Fig. 2. The variation of the FMR resonance field with angle at 23.86 GHz as the static magnetic field was rotated from a direction parallel to the specimen normal to a direction parallel with the plane of the specimen. Note the subsidiary peak displaced approximately 700 Oe towards lower fields: it is thought to be due to a standing spin-wave resonance associated with the total thickness of the film (1200 Å). The solid line was calculated from $(\omega/\gamma)^2 = H(H + 4\pi M_s \sin^2\theta)$, using $4\pi M_s = 11.80$ kOe and $\gamma = 1.871 \times 10^7$ Hz/Oe, where θ is the angle between the magnetization direction and the specimen normal: it was calculated from the magnetic field angle using the condition for static equilibrium.

out of the plane at 24 GHz is shown in fig. 2. The solid line was calculated using $4\pi M_s = 11.80$ kOe and $\gamma = 1.871 \times 10^7$/Oe along with the simple formula

$$(\omega/\gamma)^2 = H(H + 4\pi M_s \sin^2\theta) \qquad (1)$$

appropriate for a non-conducting film having no exchange stiffness. In eq. (1) θ is the angle between the magnetization and the specimen normal: it was calculated from the angle between the applied field and the specimen normal using the condition for static equilibrium. We do not know either the exchange stiffness or the resistivity for the films used in this work because the magnetic material is the cubic form of cobalt containing approximately 14 at% of dissolved manganese (see below). Fortunately, the field at which the uniform resonance occurs in a conducting film whose thickness is less than the rf skin-depth is very insensitive to the values of exchange stiffness and resistivity. For example, a value of exchange stiffness as large as 2×10^{-6} erg/cm, a value of resistivity as small as 5×10^{-6} Ω cm, and a damping parameter as small as $G = 2.0 \times 10^8$ Hz, lowers the calculated resonance * field only by 53 Oe. The agreement between calculated and observed resonance fields at 37 GHz was similar to that shown in fig. 2; the agreement was less satisfactory for 9.16 GHz where the observed resonance fields tended to be larger than the calculated values as the static field became more parallel with the specimen plane. The resonance fields and linewidths observed for the various frequencies, and for fields parallel and perpendicular to the specimen plane, are listed in table 1 along with values calculated from eq. (1) for a range of $4\pi M_s$ and γ. The values $4\pi M_s = 11.80$ kOe and $\gamma = 1.87 \times 10^7$ ($g = 2.13$) provide a satisfactory description of the data. We might also mention that the angular variation of the linewidth was in satisfactory agreement with that calculated assuming a homogeneous magnetization. The value $4\pi M_s = 11.80$ kOe is in good agreement with magnetization measurements made on a similar specimen

* Calculations were carried out using the formalism of Ament and Rado [8], and no spin pinning [9]. The specific formalism is described by Cochran et al. [10].

Table 1
FMR fields and linewidths observed for a specimen consisting of 20 layers of alternating cobalt and manganese deposited on a glass substrate. Each cobalt layer had a nominal thickness of 24 Å and each manganese layer had a nominal thickness of 36 Å: the total thickness of the specimen was 1200 Å. Calculations were carried out using the simple formula $(\omega/\gamma)^2 = (H)(H + 4\pi M_s \sin^2\theta)$. All measurements were made at room temperatures, approximately 293° C

Frequency (GHz)	Expt. H (kOe)	Expt. ΔH (Oe)	Calc. $4\pi M_s = 11.80$ (kOe) $g = 2.128$	Calc. $4\pi M_s = 11.80$ (kOe) $g = 2.116$	Calc. $4\pi M_s = 11.95$ (kOe) $g = 2.128$
H parallel to the specimen plane					
9.16	0.93 ± 0.05	190 ± 20	0.75	0.76	0.75
23.83	4.06 ± 0.10	303 ± 30	4.04	4.08	4.01
36.95	7.80 ± 0.15	400 ± 40	7.84	7.90	7.80
73.03	19.26 ± 0.15	746 ± 80	19.33	19.45	19.27
H perpendicular to the specimen plane					
9.16	14.76 ± 0.10	220 ± 20	14.88	14.89	15.03
23.83	19.67 ± 0.10	303 ± 30	19.80	19.85	19.95
36.95	24.28 ± 0.15	340 ± 40	24.21	24.28	24.36

[20 layers of 24 Å thick Co alternating with 36 Å thick Mn] reported by Sakakima et al. [11]. They obtained a value $4\pi M_s = 12.80$ kOe calculated using a thickness of 24 Å for the cobalt layers: their magnetization value is therefore subject to the same 20% uncertainty as the thickness of the cobalt layers.

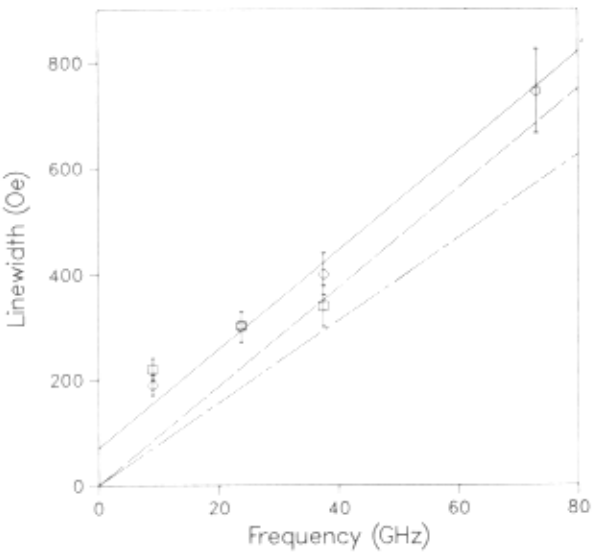

Fig. 3. FMR linewidth plotted as a function of frequency at room temperature. The solid line was calculated for a 1200 Å thick film using $4\pi M_s = 11.80$ kOe, $g = 2.12$, exchange stiffness $A = 2.0 \times 10^{-6}$ erg/cm, a resistivity of 5.0×10^{-6} Ω cm, and a Landau–Lifshitz damping parameter $G = 3.4 \times 10^8$ Hz. The dashed line and the dash-dot line were calculated for negligible exchange broadening and for $G = 3.3 \times 10^8$ and 2.8×10^8 Hz.

The experimental linewidths were found to depend linearly on frequency, within the experimental uncertainty, fig. 3. In fig. 3, the data have been compared with calculated linewidths for a 1200 Å thick film using $G = 3.3 \times 10^8$ Hz, and $G = 2.8 \times 10^8$ Hz, neglecting exchange conductivity damping (dashed and dashed-dot lines), and with linewidths calculated using $G = 3.4 \times 10^8$ Hz, an exchange stiffness $A = 2 \times 10^{-6}$ erg/cm, and a resistivity of $R = 5 \times 10^{-6}$ Ω cm. There can be little doubt that the material of our compositionally modulated films is characterized by a Landau–Lifshitz damping parameter $G = (3.4 \pm 0.5) \times 10^8$ Hz: most of the uncertainty in this value can be ascribed to the uncertainty in the measured linewidths. The zero frequency off-set is consistent with that expected for exchange conductivity broadening in a thin film: there is no evidence of two-magnon scattering such as might have been expected for an inhomogeneous magnetic material [12]. The value $G = 3.4 \times 10^8$ Hz is very reasonable; for example, $G = 2.45 \times 10^8$ Hz for pure nickel [13].

4. Discussion and conclusions

The observation that Co–Mn films behave like a soft, homogeneous, magnetic material characterized by a value $4\pi M_s = 11.8$ kOe, a value much smaller than the estimated room temperature value for pure fcc cobalt, $4\pi M_s = 18.3$ kOe, can be most simply explained on the assumption that the material in the individual cobalt-rich layers is a fcc alloy of 13–14 at% of manganese in cobalt. This estimate is based upon the concentration dependence of the Curie temperature and of the saturation magnetization at 0 K for fcc Co–Mn alloys as measured by Crangle [14]. That we are dealing here with the cubic phase of a cobalt alloy is supported by X-ray studies [11] and by ^{59}Co NMR spin echo studies [15,16] carried out on similar cobalt–manganese layered specimens. The magnetizations reported by Crangle [14] were corrected for the difference between 0 and 293 K using the assumption that the magnetization follows the same reduced temperature dependence as nickel [17]: the correction varied from 2 to 4% depending upon manganese concentration. Crangle's data in emu/g were converted to Oe/cm^3 using a density of 8.84 g/cm^3, the value for pure cobalt at room temperature reported by Myers and Sucksmith [17]. We can also conclude from the lack of inhomogeneous line broadening of the FMR lines that the average compositional variation of the Co–Mn alloy from layer to layer could not have been much greater than 2 at%. This estimate is based upon the formula quoted by Sparks [18] which takes into account dipole–dipole narrowing: $\Delta H \approx [\Delta(4\pi M_s)]^2/4\pi M_s$ where $\Delta(4\pi M_s)$ is the variation in magnetization. For $\Delta H = 50$ Oe and $4\pi M_s = 11.8$ kOe, the above formula gives a variation of 0.8 kOe in $4\pi M_s$. According to Crangle's data, $4\pi M_s$ varies by approximately 0.4 kOe per atomic percent of manganese for an alloy of 14 at% of manganese in cobalt. This argument does not exclude the possibility of a composition gradient normal to the plane of the film within a single cobalt-rich layer. It has been shown by White and Herring [19] that all of the spins within a strongly exchange coupled layer tend to process together with an effective magnetization given by $M_{\text{eff}} = \langle M^2 \rangle / \langle M \rangle$. It is interesting to note that a linear variation of magnetization across the layer from zero at each side to a maximum of 18.3 kOe at the center would give an effective magnetization $4\pi M_{\text{eff}} = 2/3(18.3) = 12.2$ kOe in good agreement with the value required by the FMR experiments. However, the corresponding average value measured by the vibrating sample magnetometer experiments, would be $(1/2)(18.3) = 9.2$ kOe – a value much smaller than the (12.8 ± 2.5) kOe required by the data.

The layers of cobalt-rich and manganese-rich material nearest the substrate were very well defined over an area at least as large as the 0.6 μm beam spot, but the layers near the surface of the sample exhibited no observable compositional modulation, and appeared to be a homogeneous mixture of cobalt and manganese. The loss of compositional modulation appeared to occur gradually during the growth of the film. This observation suggests an island growth habit in which small variations in layer thicknesses from island to island due to slight irregularities in growth would accumulate to result in a loss of coherence between the positions of the cobalt-rich and manganese-rich layers.

In conclusion, vapour-deposited multilayer films of cobalt and manganese behave like homogeneous, soft magnetic materials: they exhibit no evidence of FMR line broadening due to concentration gradients or of line-broadening due to two-magnon scattering. The Landau–Lifshitz damping parameter for an alloy of 13–14 at% manganese in cobalt was found to be $G = (3.4 \pm 0.5) \times 10^8$ Hz. The depth profiling data demonstrate that it is possible to achieve a depth resolution of approximately 30 Å in these materials using xenon ions at oblique incidence.

Acknowledgements

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