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Long-range order and magnetic properties of Mn_xPt_{1-x} thin films

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Abstract

Thin films of Mn_xPt_{1-x} (x = 0.18, 0.22, 0.25, 0.30, 0.34) were prepared by magnetron sputtering onto quartz substrates. After annealing in vacuum for 1 h at 850°C, X-ray diffraction indicates that the films have the Cu_3Au cubic structure (a = 3.87 Å) and are highly textured with the (111) axis along the film normal. These samples have a high degree of long-range order, as indicated by the ratio of the intensities of the superlattice (100) and fundamental (200) X-ray diffraction reflections. We report here the results of magnetic and magneto-optical measurements on these samples over a wide range of temperatures.

1. Introduction

The ordered alloys of the form MPt_3 (M = V, Cr, Mn, Fe, Co) have received considerable attention because of their interesting magnetic and magneto-optic properties.[l-4] These alloys form in the cubic Cu_3Au structure, with the transition metal atoms on the cube comers and the Pt atoms occupying the face-centered sites. Typically, the transition metal atoms in these alloys have a magnetic moment of a few Bohr magnetons and there is often a significant induced magnetic moment on the Pt atoms [5]. For example, Co—Pt alloys and multilayers near the $CoPt_3$ composition have perpendicular magnetic anisotropy and large magneto-optic properties in the blue and so have been widely touted as candidates for blue media magneto-optical storage applications [6-8]. Very recently, Kato et al. [2] observed quite large Kerr rotation and ellipticity in thin films of $MnPt_3$, suggesting that it may also be suitable for such applications.

The stoichiometric ordered alloy $MnPt_3$ is ferromagnetic with a Curie temperature T_C of 380 K. The magnetic moment is near $4\mu_B$ per formula unit, with about $3.6\mu_B$ on the Mn atoms and remainder on the Pt atoms. Auwärter and Kussman [9], in their study of bulk samples, showed that T_C increased as the Mn concentration was increased above 25 at%, while the saturation magnetization decreased. Neutron diffraction measurements by Menzinger et al. [10] showed that Mn atoms occupying Pt sites coupled to their neighbors antiferromagnetically, which is consistent with the decrease in magnetization with increasing Mn content. More recently, Kato et al. [2] studied the room-temperature magneto-optic properties of $Mn_{0.253}Pt_{0.747}$ thin film. They found large magneto-optic effects over much of the visible, with a peak Kerr rotation (θ_K) value of –1.18° occurring at a photon energy of 1.2 eV. The band structure calculations of Oppeneer et al. [11] were in quite good agreement with these experimental results.

In the work presented here, we prepared thin films of Mn_xPt_{1-x} ($0.18 < x < 0.34$). After annealing these films have a high degree of long-range order. We have studied their magnetic properties over a wide temperature range, and our results are substantially in agreement with published results for bulk samples of $MnPt_3$. In addition, we have studied the magneto-optic properties using Kerr rotation and ellipticity measurements, and find good agreement with the experimental results of Kato et al. [2] and the band structure calculations of Oppeneer et al. [11].

2. Experimental procedures

Mn/Pt multilayers were prepared by dc magnetron sputtering onto room temperature fused quartz substrates. The base pressure of the sputtering chamber was 4×10^{-7} Torr and the argon sputtering pressure was 2 mTorr. For the series of samples discussed here the individual Pt layer thicknesses were held constant at 7 Å, while the Mn layer thickness was varied to obtain Mn_xPt_{1-x} of average compositions ($x = 0.18$, 0.22, 0.25, 0.30, 0.34). The compositions of the as-deposited films were measured using X-ray fluorescence and were found to be close to the nominal compositions. The total film thickness of each sample was 100 nm and each film was coated with a 100 nm thick SiO_x protective overcoat. The as-deposited samples were subsequently annealed in vacuum (6×10^{-6} Torr) at 850°C for 1 h to form the homogeneous crystalline Mn_xPt_{1-x} alloys.

The saturation magnetization M_s and coercivity H_c of the Mn_xPt_{1-x} films were measured over the 20–300 K temperature range using an alternating gradient force magnetometer. The polar Kerr rotation and ellipticity spectra were measured using a home-built system based on a photoelastic modulator, and the Curie temperatures were obtained from Kerr rotation versus temperature measurements in an applied field of 7.1 kOe.

3, Results and discussion

X-ray diffraction of measurements of the annealed Mn_xPt_{1-x} films are consistent with the cubic Cu_3Au structure. Fig. l(a) shows the X-ray diffraction results for a $MnPt_3$ film. The measured *a*-axis lattice parameter of 3.87 Å for MnPt3 agrees reasonably well with the value of 3.89 Å obtained for bulk $MnPt_3$ samples [5]. The film is highly textured with the (111) direction perpendicular to the film plane. In addition to the dominant (111) diffraction peak, weak (100), (110), (200), (210) and (211) peaks are observed. Of particular interest is the weak (100) peak, which is forbidden for the disordered $MnPt_3$ alloy, but allowed for the ordered alloy. Ignoring the difference in the thermal factors, the intensity ratio of the (100) to (200) diffraction peaks can be written as [12,13]

$$\frac{J_{(100)}}{J_{(200)}} = \frac{\eta_*^2 \gamma^2 \left[(f_m - f_p)^2 P_L \right]_{(100)}}{\left[\left((1-x) f_p + x f_m \right)^2 P_L \right]_{(200)}}, \qquad (1)$$

where x is the concentration of Mn atoms, and $y = 3x$ for $x \leq 0.25$, $y = 1 - x$ for $x \geq 0.25$. The long-range order parameter η describes the deviation of order from the maximum possible value for an alloy of a given composition and varies from zero (completely disordered state) to unity (maximum order possible) [13]. f_p and f_m are the scattering amplitudes for Pt and Mn,

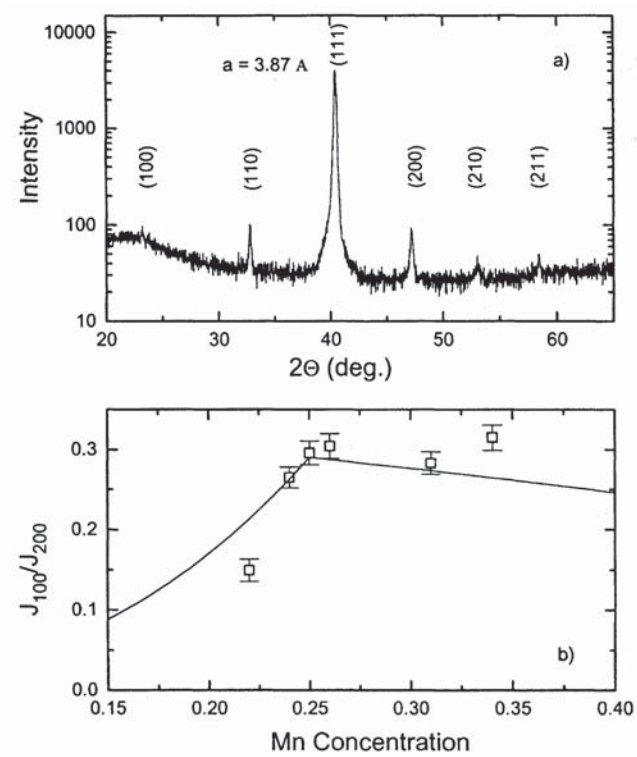

Fig. 1. (a) X-ray diffraction of $Mn_{0.25}Pt_{0.75}$ thin film; (b) XRD intensity ratio of the (100) to (200) diffraction peaks as a function of Mn concentration. □ experimental data; solid curve, calculated ratio as discussed in text.

respectively, P_L is the Lorentz polarization factor [14] given by

$$P_L = \frac{1 + \cos^2 \delta_m \cos^2 2\theta}{\left(1 + \cos^2 2\delta_m\right) \sin^2 \theta \cos \theta}, \qquad (2)$$

where $\delta_m \equiv$ graphite monochromator Bragg angle, and $\theta \equiv$ Bragg angle of the *(hkl)* reflection. Fig. l(b) displays the measured and calculated (Eq. (1)) integrated intensity ratios between the superlattice (100) and fundamental (200) reflections as a function of Mn content. As x is increased to 0.25 this ratio tends to saturate at a value of 0.29. Using the three data points near $x = 0.25$, an estimated value of $\eta_* = 1.03 \pm 0.05$ is obtained using Eq. (1) and the atomic form factors obtained from Ref. [15]. Thus, the solid curve in Fig. l(b) was calculated using Eq. (1) with $\eta_* = 1$. Obviously there is reasonable semi-quantitative agreement between the measured and calculated intensity ratios, indicating the samples have a high degree of long-range order. Eq. (1) assumes random distributions of Mn on the Pt sites above 25% Mn. However, above 38% Mn a face-centered tetragonal (fct) phase appears which is stable in the range from about 38% to 50% Mn [12]. In the fct phase the Mn and Pt atoms form alternating planes perpendicular to the c-axis. The larger measured J_{100}/J_{200} ratio at $x = 0.34$ is suggestive of this Mn site occupational preference at compositions less than necessary for formation of the fct phase.

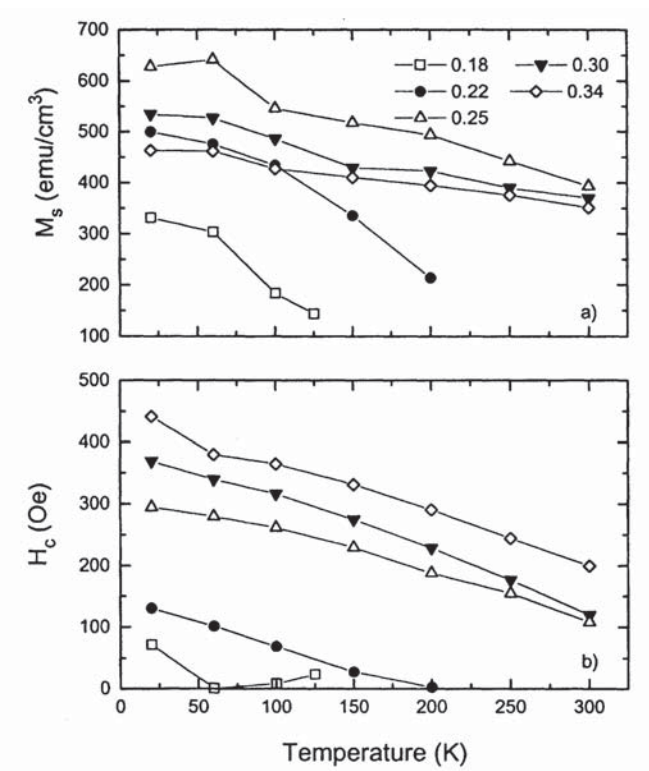

Fig. 2. (a) M_s versus temperature, (b) H_c versus temperature for Mn_xPt_{1-x} ($x = 0.18, 0.22, 0.25, 0.30, 0.34$).

The temperature dependencies of the in-plane saturation magnetizations and the coercivities of each Mn_xPt_{1-x} film are displayed in Fig. 2(a) and (b), respectively. The room-temperature values of M_s range from 0 to 395 emu/cm^3, with the $x = 0.25$ film showing the largest value. At 20 K, the $x = 0.25$ sample has a measured saturation moment of $3.92\mu_B$ per formula unit, which is in good agreement with the value of $4.04 \pm 0.08\mu_B$ reported for bulk [5] and also with the calculated moment of $4.1\mu_B$ calculated for the fully ordered state [11]. These results are summarized in Table 1. Thus, the magnetic properties of our thin films are quite similar to those of bulk samples. The in-plane coercivities of the samples are generally in the range of a few hundred Oe, and they increase with decreasing temperature. Fig. 3 shows the Curie temperature as a function of Mn concentration. The values obtained for the thin films are in excellent agreement with those found by Auwarter and Kussman [9] in bulk Mn_xPt_{1-x}. Both T_C and H_c increase with increasing Mn content. This increase in T_C with x is most likely due to the increased macroscopic exchange stiffness between the Mn atoms as the face-centered sites are occupied.

The room-temperature Kerr rotation (θ_K) and ellipticity (η_K) spectra (measured from the substrate side) for three of the samples are shown in Fig. 4. These spectra show considerable structure. The Kerr rotation spectrum for the $x = 0.25$ samples shows negative and

Table 1
Summary of the magnetic properties of Mn_xPt_{1-x}

x	M_s (emu/cm^3) at 20 K	M_s (emu/cm^3) at 300 K	μ_B /(unit cell)	T_C (K)
0.18	332	0	2.07	188
0.22	500	0	3.12	282
0.25	628	395	3.92	375
0.30	535	370	3.34	465
0.34	465	352	2.91	530

positive peaks in the near uv and a slow (negative) increase to –0.9° at 900 nm. These results are in quite good agreement with the experimental results of Kato et al. [2]. The ellipticity of the $x = 0.25$ sample has a maximum value of – 0.59 at 560 nm. The shape of the ellipticity spectrum is quite similar to that of Kato et al. [2], but it does not show a sign reversal at 345 nm. This is possibly due to different sample preparation and annealing procedures. Similar effects were explicitly demonstrated for disordered and ordered FePt alloys, where Cebollada et al. [16] showed that the details of the θ_K and η_K spectra depend quite strongly on the degree of long-range order.

4. Summary

We have prepared thin films of highly-ordered Mn_x-Pt_{1-x} alloys by magnetron sputtering and shown that the magnetic properties depend strongly on Mn concentration. These films show the same strong dependence of the Curie temperature on Mn content as was observed for bulk samples. We also measured the coercivities and saturation magnetizations of these films over the 20–300 K temperature range, finding that H_c increases with decreasing temperature and increasing Mn content. Our Kerr rotation and ellipticity measurements are in quite good agreement with the earlier results of Kato et al.[2]

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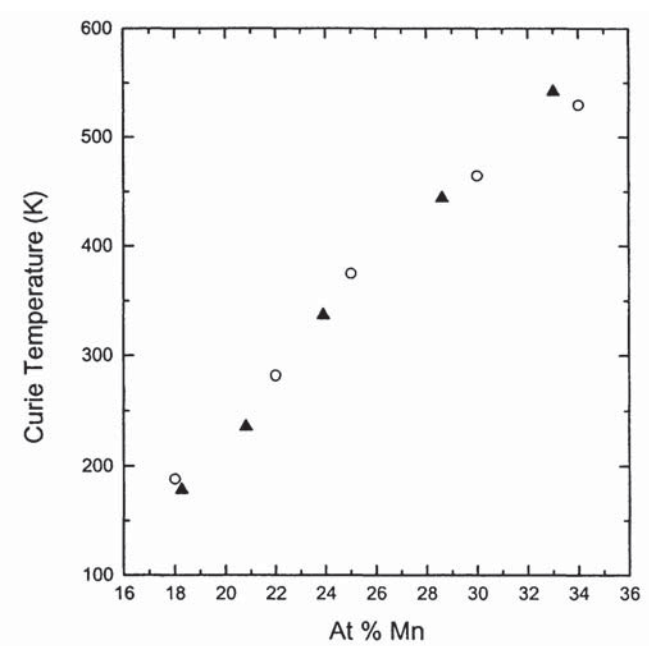

Fig. 3. Curie temperature versus Mn concentration for bulk and thin-film specimens; ○ thin film, ▲ bulk [9].

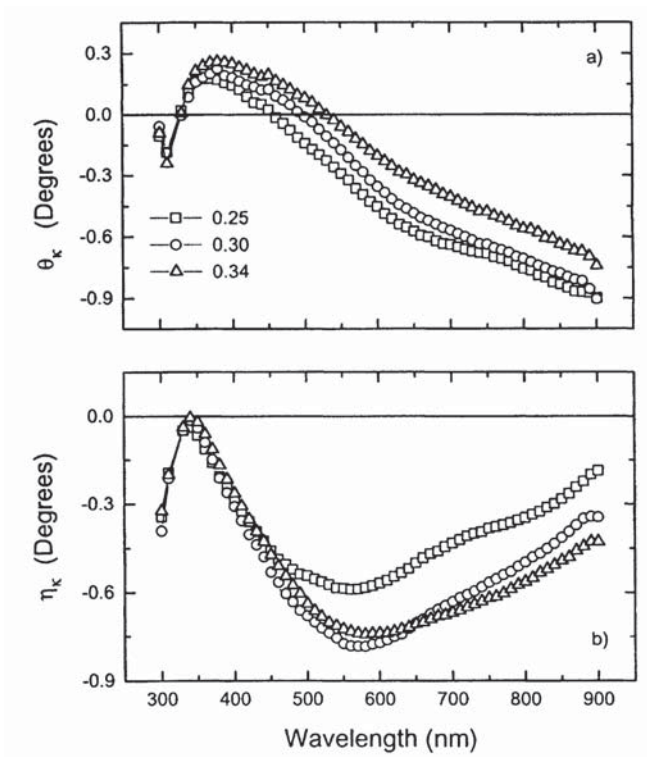

Fig. 4. (a) θ_K and (b) η_K wavelength dependence of Mn_xPt_{1-x} ($x = 0.25, 0.30, 0.34$).

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