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Stoichiometry–anisotropy connections in epitaxial $L1_0$ FePt(001) films

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The order parameters and anisotropy constants of a series of epitaxial $L1_0$ FePt films with compositions in the range of 45–55 at. % Fe and nominal thicknesses of 50 nm have been characterized. The films were made by cosputtering the elements onto single crystal MgO(001) substrates. The substrates were coated with 1 nm Pt/1 nm Fe bilayer seeds prior to alloy deposition. Both the bilayer seed and the alloy film were deposited at 620 °C. Lattice and order parameters were obtained by x-ray diffraction. Film compositions and thicknesses were determined by Rutherford backscattering spectrometry, and room-temperature magnetocrystalline anisotropies were determined with a torque magnetometer. It was found that the order parameter had a maximum for the film composition closest to the equiatomic composition, whereas the magnetocrystalline anisotropy increased as the Fe content increased from below to slightly above the equiatomic composition. These results imply that nonstoichiometric FePt compositions, with a slight excess of Fe, may in fact be preferred for applications that require high anisotropy. © 2004 American Institute of Physics. [DOI: 10.1063/1.1667856]

I. INTRODUCTION

With appropriate easy-axis orientation and microstructure control, high-anisotropy, high-energy-product, chemically ordered FePt films with the $L1_0$ crystal structure are attractive candidates for many advanced magnetic material applications such as high-density recording media or micro- and nanoelectromechanical systems magnetic components.^{1–11} As the chemical ordering characteristics and associated magnetocrystalline anisotropy for nonequiatomic FePt compositions are different from those characterizing the equiatomic composition, it is important to systematically investigate the atomic ordering and the resultant magnetic properties as a function of alloy composition. In this work, we characterize the order parameters and anisotropy constants of a series of epitaxial FePt films with Fe concentrations of 45–55 at. %. As we demonstrate, the highest-order parameter does not correspond to the highest magnetic anisotropy, implying that nonstoichiometric FePt films may be preferable for applications that require high anisotropy.

II. EXPERIMENT

FePt alloy films with nominal thicknesses of 50 nm and compositions in the range of 45–55 at. % Fe were epitaxially grown by cosputtering elemental targets onto seed layers of 1 nm Pt/1 nm Fe deposited on MgO(001) single crystal substrates. The base pressure of the sputtering chamber was below 5×10^{-8} Torr and the sputtering gas was Ar–4% H₂ at a pressure of 3 mTorr and a flow rate of 20 sccm, with deposition rates of 0.1–0.2 Å/s. The bilayer seed and the alloy film were both deposited at a substrate temperature of 620 °C.

The composition and thickness of the films were measured by Rutherford backscattering spectrometry (Table I). Lattice and order parameters were determined by normal and in-plane x-ray diffraction (XRD). Cu $K\alpha$ radiation and a graphite monochromator were used for all the scans. A beam incidence angle of 2°, with respect to the sample plane, was adopted in the case of the in-plane scans.

For the determination of the unit-cell dimensions, the in-plane lattice parameter, a , was determined using the (110) superlattice peak, whereas the lattice parameter perpendicular to the film plane, c , was obtained by using the three (00 l) peaks. The long-range order parameter, S , was calculated as

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TABLE I. Measured composition, thickness, lattice parameters a and c , c/a and order parameter, S , for FePt(001) films. S_{\max} is the maximum order parameter for the given composition. See text for more details. Also listed is whether or not in-plane c -axis variants were present. The substrate temperature, T_s , for seed layer and film deposition was 620 °C. The relatively large error in the actual compositions ($\pm 2.0\%$) and thicknesses (± 5.0 nm) measured by Rutherford backscattering are a result of the presence of the bilayer Fe/Pt seed.

Material	Composition		Thickness (nm)	a	c	c/a	S	S_{\max}	In-plane c -axis variant
	x (at. %)								
Fe _x Pt _{100-x}	46.0		50.6	3.870	3.721	0.961	0.89±0.02	0.92	No
/Pt(1 nm)	51.0		57.7	3.863	3.710	0.960	0.93±0.05	0.98	No
/Fe(1 nm)	51.9		53.6	3.857	3.706	0.961	0.89±0.06	0.96	No
	55.3		47.8	3.839	3.704	0.965	0.72±0.05	0.89	Yes

the average of the two order parameters determined from the ratios of superlattice-to-fundamental (001)/(002) and (003)/(002) total integrated peak intensities.^{9,12} For these calculations, the root-mean-square displacement amplitude σ was taken as 0.14 Å.^{5,13} The long-range order parameter measures the extent of chemical order and is given by $S = r_\alpha + r_\beta - 1 = (r_\alpha - x_A)/y_\beta = (r_\beta - x_B)/y_\alpha$, where, x_A and x_B are the atom fractions of the two components, y_α and y_β are the fraction of lattice site types α and β in the ordered structure, and r_α and r_β are the fraction of each type of lattice site occupied by the correct types of atom (A on α and B on β). The maximum order parameter for the given film composition is given by $S_{\max} = 1 - 2\Delta x$, where Δx is the compositional deviation in atom fraction from 0.5.

The lattice parameters and the order parameters of the films studied here are listed in Table I, along with whether in-plane c variants are present or not. These variants are ordered regions with their c axes, i.e., the [001] unit-cell axes of the ordered phase, in the plane of the film rather than perpendicular to the plane of the film as would be desired, and give rise to a (200)/(020) peak in the normal XRD scans. The errors in the order parameters were calculated as the standard deviation of the two values for (001)/(002) and (001)/003) and were found to be in the range of 0.02–0.06.

Magnetic characterization was carried out both parallel and perpendicular to the film plane using a vibrating sample magnetometer at room temperature in the applied field ranges of -18.5 to $+18.5$ kOe. No corrections for demag-

netization were made. The room-temperature anisotropies were determined with a torque magnetometer operated in the applied field range of 0 to 18.5 kOe. In detail, the intrinsic anisotropy constant is defined as $K_1 = K_1^{\text{eff}} + 2\pi M_s^2$, where K_1^{eff} is the perpendicular anisotropy constant that is measured using the “45° method” and high-field extrapolation.^{5,14}

III. RESULTS AND DISCUSSION

The evolution of the relative intensity of the superlattice and fundamental peaks as a function of FePt stoichiometry is shown in Fig. 1. The order parameter of the films is plotted as a function of film composition in Fig. 2. Table I and Fig. 2 indicate that the highest-order parameter (0.93) is obtained for the film composition closest to the equiatomic (51.0 at. % Fe), as might have been expected for the $L1_0$ structure. The evolution of the FePt magnetic properties resulting from the $L1_0$ chemical ordering of the epitaxial films may be followed by an examination of the $M-H$ curves. These are shown as a function of composition in Figs. 3(a) and 3(b) and reveal a progression in the squareness, remanence, and coercivity. The films exhibit an initially sharp reversal, indicative of a nucleation barrier for the formation of the oppositely magnetized domains. For the film with the lowest Fe content (46.0 at. %), the nucleation of the negatively (i.e., reverse) magnetized domains occurs before the external positive field is reduced to zero, as a result of the demagnetization field of the sample in the perpendicular direction. For the films with higher concentrations of Fe, a higher nucleation barrier is present and the nucleation of reverse domains is delayed un-

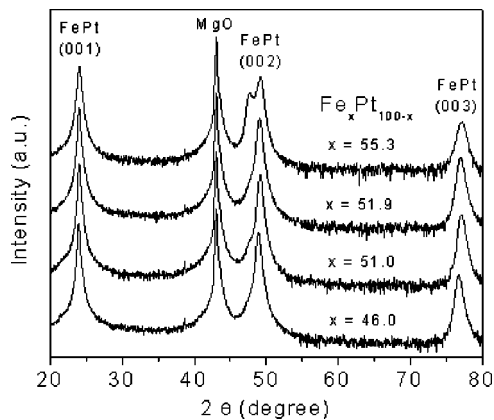


FIG. 1. θ - 2θ XRD scans of Fe_xPt_{100-x} epitaxial films as a function of film composition.

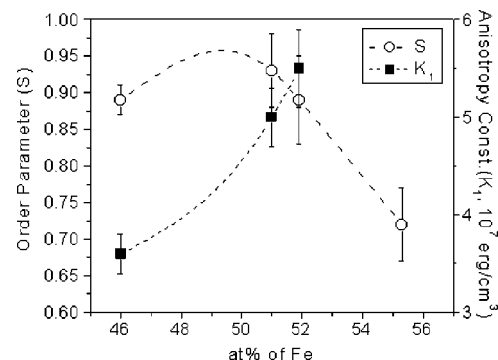


FIG. 2. The order parameter, S , and intrinsic anisotropy constant, K_1 , of FePt(001) epitaxial films as a function of film composition. The lines are drawn to guide the eye.

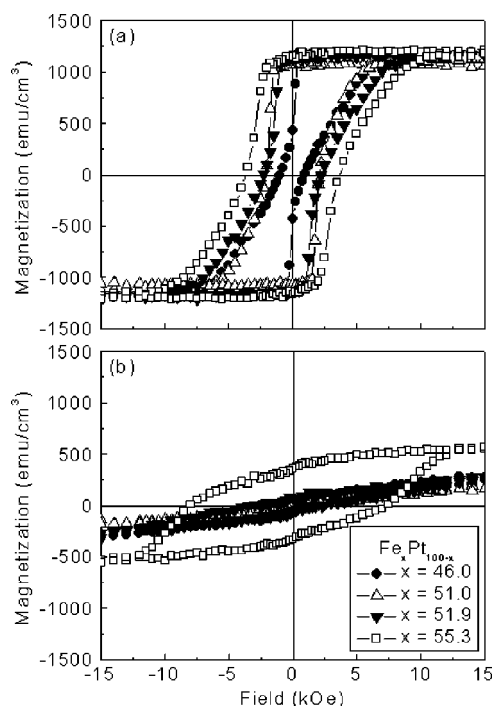


FIG. 3. (M - H) hysteresis loops, (a) perpendicular, (b) parallel, as a function of film composition for FePt(001) epitaxial films.

til the external field has become negative, aiding the demagnetization field of the sample. For the film with the highest Fe content (55.3 at. %), a significantly increased in-plane coercivity was observed likely as a result of the presence of the in-plane c -axis variants (Fig. 1 and Table I).

The saturation magnetization values, M_s , of the films in Table II studied here can be compared with that reported for bulk FePt at 1140 emu/cm³.¹⁵ The computed magnetocrystalline anisotropy constants K_1 are also listed in Table II and are found to be somewhat smaller than those reported by other researchers for similar films.^{3,5,10} The anisotropy constant was not determined for the 55.3 at. % Fe film because of a significant coercivity measured in the film plane as noted above, indicative of the presence of in-plane c -axis variants, as seen in the XRD studies (Fig. 1 and Table I). There appears to be no simple correlation between the measured FePt order parameter and the measured anisotropy constant. The order parameter has a maximum for the film composition (51 at. %) closest to equiatomic, whereas the anisotropy shows a monotonic increase as the Fe content is

TABLE II. Saturation magnetization, coercivity from perpendicular hysteresis loops, and magnetic anisotropies of FePt(001) epitaxial films.

Composition (at. % Fe)	M_s (emu/cm ³)	H_c (Oe)	K_{\perp} (erg/cc) 10 ⁷	K_{\parallel} (erg/cc) 10 ⁷	S
46.0	1175 ± 117	969	2.6 ± 0.2	3.6 ± 0.2	0.89
51.0	1070 ± 117	2105	4.1 ± 0.3	5.0 ± 0.3	0.93
51.9	1176 ± 117	2263	4.5 ± 0.4	5.5 ± 0.4	0.89
55.3	1197 ± 117	3683	0.72

increased from below (46 at. %) to slightly above (52 at. %) the equiatomic composition.

IV. CONCLUSIONS

It is demonstrated that the order parameter in FePt(001) epitaxial films of varied Fe content has a maximum near the equiatomic composition, as is expected for the $L1_0$ structure. Magnetization studies reveal the presence of nucleation barriers for the reversal of the films in the perpendicular direction. The anisotropy of the films is quantified by the determination of the intrinsic anisotropy constant K_1 . Whereas the measured order parameter has a maximum at the composition closest to equiatomic, the magnetocrystalline anisotropy increases as the Fe content is increased from below to slightly above the equiatomic composition. It is believed that a slight excess of Fe increases the magnetization and perhaps also the polarization of the Pt, thereby increasing the intrinsic anisotropy constant of the $L1_0$ phase without appreciably degrading the long-range chemical order. Taken together, the results of our study imply that nonequiatomic compositions of FePt alloy films may in fact be preferred for applications that require high magnetic anisotropy.

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