## Enhanced magneto-optical Kerr effect in spontaneously ordered FePt alloys: Quantitative agreement between theory and experiment

A. Cebollada and D. Weller

IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, California 95120

J. Sticht

Biosym Technologies Inc., 9685 Scranton Road, San Diego, California 92121

G.R. Harp, R.F.C. Farrow, R.F. Marks, R. Savoy, and J.C. Scott

IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, California 95120

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A chemical-ordering-induced peak in the polar magneto-optical Kerr effect spectra of FePt compounds is found to be in quantitative agreement with predictions based on *ab initio* magneto-optical calculations. Experimental spectra of epitaxial molecular-beam-epitaxy grown FePt(001) films reveal a more than twofold enhancement of the Kerr rotation if full chemical ordering is present. A spectral feature at 2-eV photon energy shows a peak Kerr rotation of up to  $\sim 0.8^{\circ}$  which is strongly correlated with the presence of perpendicular magnetic anisotropy in ordered compound films.

The magneto-optical Kerr effect (MOKE) of FePt alloys and multilayers has been studied extensively in the recent literature because of their technological importance as magneto-optic or magnetic recording materials. Examples are arc-melted<sup>1</sup> or sputtered<sup>2</sup>  $Fe_xPt_{1-x}$  alloys and several types of Fe/Pt (Refs. 3, 4) and FePt/Pt sputtered multilayers.<sup>5,6</sup> A new feature in polar Kerr rotation spectra related to the formation of the chemically ordered CuAu(I) crystal structure of FePt near the equiatomic composition has recently been reported by Lairson et al.<sup>7,8</sup> in [001]-oriented, post-annealed sputtered Fe/Pt multilayers. Sugimoto et al.9 confirmed the presence of this chemical ordering feature, but found an overall decrease in the magneto-optical activity in their [111]-oriented, post-annealed sputtered FePt alloys. We have endeavored to grow fully ordered FePt compound films of equiatomic composition, determine quantitatively the chemical ordering parameter, and compare MOKE spectra of such samples with ab initio MOKE calculations<sup>10</sup> of FePt in the AuCu(I) structure. In contrast to previous work, we grow high quality epitaxial compound films by simultaneous deposition of Fe and Pt at equilibrium growth temperatures in ultrahigh vacuum (UHV). The dynamics of such a process is governed by the adatom mobility at the growing film surface<sup>12</sup> rather than by bulk diffusion, exploited in post-annealing experiments. We show here that indeed spontaneous chemical ordering to the almost fully ordered FePt L10 compound phase occurs in molecular-beam-epitaxy (MBE) growth at  $\sim 500$  °C, which is considerably lower than the bulk order-disorder temperature of  ${\sim}1300\,^{\circ}\mathrm{C}.^{11}$  We have specifically exploited the sensitivity of magnetooptical spectra to alloy chemical ordering,<sup>12</sup> structural ordering, and crystallographic orientation<sup>13</sup> to synthesize these compound films.

We restrict the present discussion to two 1000 Å thick FePt alloy films prepared in a VG Semicon 80M MBE system<sup>14</sup> by seeded epitaxial growth at 100 and 500 °C, respectively. Templates were polished MgO(100) substrates onto which 150 Å thick Pt buffer layers were grown at 700 °C. The MgO substrates were dipped in a degreasing solution before being transferred into UHV and heat cleaned for 30 min at 600 °C prior to the deposition of the Pt buffer layers. High quality (001)oriented FePt films were subsequently coevaporated from independently controlled Fe and Pt e-beam sources at base pressures of  $2 \times 10^{-10}$  mbar. Deposition rates were  $\approx 0.1-0.3$  Å/s. Such films presented in-plane square symmetry, as determined in situ by reflection high-energy electron diffraction (RHEED) and low-energy electron diffraction (LEED). To avoid sample deterioration a 20 Å Pt capping layer was deposited at room temperature, before removing the samples from the vacuum system. Compositions and thicknesses were cross-checked ex situ with x-ray fluorescence, electron microprobe, and Rutherford backscattering. Figure 1 shows specular xray diffraction (XRD) scans taken with Cu  $K\alpha$  radiation of two FePt samples grown at 100 and 500 °C, respectively. Both spectra show strong substrate, Pt seed layer, and FePt(002) peaks. In addition, the sample grown at 500 °C shows intense FePt(001) and FePt(003) reflections. These are characteristic of the ordered  $FePt(L1_0)$ compound phase [Fig. 1(b)], which can be viewed as a natural superlattice of alternating Fe and Pt atomic planes along the [001]-growth direction.<sup>15</sup> In the disordered state, Fe atoms are substituted into Pt positions and vice versa, which destroys the superlattice structure [Fig. 1(a)]. The alloy chemical coherence length along the surface normal for the ordered sample is found to be  $\approx 100$  Å [1/HWHM (half width at half maximum) of the FePt(001) peak]. Negligible amounts of FePt(111), due to some misoriented Pt(111) growth, and a very small amount of FePt(200) with the tetragonal c axis in the plane of the film indicate some remaining structural imperfections. Integration of the FePt(00L) peak areas yields a one-dimensional chemical ordering parameter S

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FIG. 1.  $\theta$ -2 $\theta$  XRD scans for two FePt samples taken with Cu  $K\alpha$  radiation. The sample grown at 100 °C (a) shows substrate and seed layer peaks and a strong FePt(002) reflection from the average Fe and Pt spacing. The sharp features at  $2\theta$ =38.6° and 82.8° are due to the MgO(100) substrate. The sample grown at 500 °C (b) shows in addition some minority FePt orientations (see text) and intense FePt(001) and (003) superlattice peaks representative of the tetragonal  $L1_0$ , chemically ordered structure, with the *c* axis in the film normal direction.

which can be defined as<sup>16</sup>

$$S = \frac{r_{\rm Fe} - x_{\rm Fe}}{y_{\rm Pt}} = \frac{r_{\rm Pt} - x_{\rm Pt}}{y_{\rm Fe}}.$$
 (1)

Here  $x_{Fe(Pt)}$  is the atomic fraction of Fe(Pt) in the sample,  $y_{Fe(Pt)}$  the fraction of Fe(Pt) sites, and  $r_{Fe(Pt)}$  the fraction of Fe(Pt) sites occupied by the right atomic species. S reaches unity for perfectly ordered films of the stoichiometric composition Fe<sub>50</sub>Pt<sub>50</sub>, and is zero for a *chemically* disordered film. The area of either a fundamental or a superstructure peak can be expressed as

$$A \propto (LP)FF^*,$$
 (2)

where LP is the Lorentz polarization factor for a single crystal<sup>17</sup> and F and  $F^*$  are the structure factor and its complex conjugate. For the  $L1_0$  structure we have

$$(FF^*)_{\text{fund}} = 16[(x_{\text{Fe}}f_{\text{Fe}}e^{-M_{\text{Fe}}} + x_{\text{Pt}}f_{\text{Pt}}e^{-M_{\text{Pt}}})^2 + (x_{\text{Fe}}\Delta_{\text{Fe}}e^{-M_{\text{Fe}}} + x_{\text{Pt}}\Delta_{\text{Pt}}e^{-M_{\text{Pt}}})^2] \quad (3)$$

TABLE I. Parameters used to estimate the chemical ordering parameter S of FePt(001). We find  $S=0.9\pm0.1$  according to Eqs.(1)-(4).

Peak	Q (Å <sup>-1</sup> )	LP	$M_{ m Fe}$	$f_{ m Fe}$	$\Delta_{\rm Fe}$	$M_{ m Pt}$	$f_{ m Pt}$	$\Delta_{Pt}$
(001)	1.698	2.26	0.005	21.01	3.4	0.005	64.43	8
(002)	3.396	0.94	0.019	16.93	3.3	0.018	55.12	7
(003)	5.094	0.54	0.043	12.86	3.3	0.041	45.81	7

$$(FF^*)_{super} = 4S^2 [(f_{Pt}e^{-M_{Pt}} - f_{Fe}e^{-M_{Fe}})^2 + (\Delta_{Pt}e^{-M_{Pt}} - \Delta_{Fe}e^{-M_{Fe}})^2]$$
(4)

for fundamental and superstructure peaks, respectively.  $e^{-M}$  is the Debye-Waller correction and f and  $\Delta$  are the real and imaginary parts of the atomic scattering factors, respectively. Therefore, the chemical order parameter can be directly extracted from the ratio of the integrated areas of the fundamental (002) peak,  $A_f$ , and one of the superstructure peaks (001) or (003),  $A_s$ . Using the values for Fe and Pt of these parameters for the different diffraction peaks as listed in Table I, we find  $S=0.9\pm0.1$ for the sample grown at 500 °C. Only a small increase in the intensity around the (001) peak region was detected in the sample grown at 100 °C with no indication of (003) within the noise of the XRD measurement. In this case we estimate S < 0.1.

Figure 2 illustrates the effect of chemical ordering on the complex polar MOKE spectra (Kerr rotation  $\theta_K$  and Kerr ellipticity  $\epsilon_K$ ) of FePt. The inset shows two polar



FIG. 2. Complex polar magneto-optical Kerr spectra of the two samples described in Fig. 1. Note that the striking increase in the Kerr rotation for the film grown at 500 °C correlates both with near perfect chemical ordering and with a large perpendicular magnetic anisotropy, as indicated by polar Kerr loops (inset).



FIG. 3. Complex polar Kerr spectrum calculated using *ab initio* theory. The agreement with the experiment is very good; in particular, the 2 eV peak and the strong ultraviolet features are reproduced.

Kerr hysteresis loops taken with a 633 nm (2 eV) HeNe laser. The spectra were measured in the presence of external fields of  $\pm 28$  kOe in the polar geometry with the field normal to the film plane, ensuring magnetic saturation in both the ordered and disordered films. Α more than twofold enhancement of the Kerr rotation at 2 eV photon energy accompanied by a strong out-of-plane anisotropy is observed, as chemical ordering is induced. Both  $\theta_K$  and  $\epsilon_K$  spectra are strongly affected. For a comparison, we find a peak Kerr rotation of  $|\theta_K|=0.79^\circ$ . which is 30% larger than that reported by Lairson *et al.*<sup>8</sup> [annealed Fe/Pt(001) multilayer:  $|\theta_K|=0.61^\circ$ ] and more than a factor of 2 larger than that reported by Sugimoto et al.<sup>9</sup> [annealed FePt(111) alloy:  $|\theta_K|=0.38^\circ$ ]. In agreement with the conclusions of Sugimoto et al.<sup>9</sup> we find no evidence for an optical constant reduction in the vicinity of 2 eV excluding this as a possible cause for the observed strong Kerr effect. Rather we conclude that the Kerr enhancement is indeed due to the geometrical symmetry break introduced by the natural superlattice formation in the ordered FePt  $L1_0$  phase. We attribute the differences between the present results and previous work<sup>9,7,8</sup> to differences in the sample quality, mainly the degree of chemical ordering S. An orientation dependence as explanation of the low Kerr rotation observed by Sugimoto et al.9 can be ruled out, since we also observe large Kerr rotations in excess of  $|\theta_K| = 0.60^\circ$  in (111)-oriented films (not shown). For future reference we quote the presently

TABLE II. Optical and magneto-optical parameters of the present disordered and ordered FePt samples at 2 eV photon energy. Note that the present films were covered with ~20 Å Pt and that uncovered films show approximately 5% larger values in  $\theta_K$  and  $\epsilon_K$ . The real and imaginary parts of the off-diagonal complex tensor  $\tilde{\sigma}_{xy}$  were calculated according to Ref. 18 (10<sup>-13</sup> s<sup>-1</sup> units).

$\overline{S}$	n	k	$\theta_K(^\circ)$	$\epsilon_K(^\circ)$	$\sigma_{1xy}$	$\sigma_{2xy}$
0.0	2.23	3.06	-0.32	-0.14	1.18	8.14
0.9	3.12	3.74	-0.79	-0.19	-9.58	38.84

found Kerr and optical constants in Table II, where we have also calculated the off-diagonal conductivity  $\tilde{\sigma}_{xy}$  at 2 eV photon energy (see, e.g., Ref. 18).

Our results are in good agreement with state-of-theart ab initio MOKE calculations (see also Osterloh et al.<sup>10</sup>). Figure 3 shows a comparison between theory and experiment for ordered FePt in the AuCu(I) structure. The calculations were carried out using experimetally determined lattice parameters c=3.70 Å and a=3.8665 Å (c/a = 0.957). Variations of these parameters within a range of  $\sim \pm 2\%$  did not have a significant effect on the theoretical spectra and it is concluded that subtle changes of the c/a ratio do not explain the large Kerr effect observed here. The two distinct features at 2 and 5 eV are quantitatively reproduced, which is independent evidence for the high sample quality and in particular the presence of chemical ordering. Note that the present ab initio MOKE theory treats interband optical and magneto-optical transitions only. Free electron contributions or intraband transitions, which are dominant in the infrared region of the spectrum  $(E_{ph} < 1.5 \text{ eV})$ , are generally neglected. This, in our opinion, explains the strong deviations in that range. For computational details we refer to Ref. 19.

In conclusion, spontaneously ordered highly (001)oriented FePt compound films can be synthesized in MBE growth at temperatures far below the bulk orderdisorder transition temperature. Fully ordered FePt(001) compounds show unique magnetic and magneto-optic properties including a more than twofold-enhanced Kerr rotation at 2 eV photon energy and strong perpendicular magnetic anisotropy. The experimental MOKE spectra are in quantitative agreement with *ab initio* MOKE calculations.

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