

Influence of hydrogen on the growth of FePt thin films

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FePt films have been prepared by pulsed laser deposition on MgO(100) in a forming gas atmosphere of 5% hydrogen in argon. At a pressure and temperature combination of 0.1 mbar and 800 °C, smooth continuous and well ordered $L1_0$ phase films can be obtained. The anisotropy field is close to the bulk value, while the coercivity is comparatively low at 0.15 T. This is in contrast to films deposited in vacuum, which are granular in nature and show huge coercivity. It is proposed that nucleation centers for island growth introduced by hydrogen on the MgO surface are responsible for this difference in growth mode. The temperature and pressure dependence of the surface morphology are explained by this assumption. © 2006 American Institute of Physics.

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I. INTRODUCTION

Due to its high magnetocrystalline anisotropy, high saturation magnetization and very small critical grain size of around 3 nm, at which magnetization becomes thermally unstable at room temperature, the $L1_0$ ordered equiatomic FePt alloy has been extensively studied and proposed as a promising candidate for ultrahigh density recording media.¹⁻⁷ Since unordered FePt needs a high-temperature treatment either by depositing on heated substrates and/or post-annealing to overcome the energy barrier for formation of the ordered $L1_0$ phase, there have been several attempts to lower the ordering temperature, such as multilayered Fe/Pt,⁸ addition of a third element^{9,10} or introducing Ag underlayers.¹¹ Several studies have shown that the presence of hydrogen gas enhances the ordering of superstructure forming binary alloys.¹²⁻¹⁴ Recently, Leistner and co-workers¹⁵ reported on the influence on ordering of electrodeposited FePt by post annealing in hydrogen, where phase formation is favored and $L1_0$ ordering starts at temperatures as low as 350 °C. Similar results have been obtained by Lai *et al.* for sputtered FePt films, with ordering temperatures as low as 275 °C.¹⁶ In this article, we report on the influence of hydrogen on the growth, microstructure and the magnetic properties of FePt thin films prepared by pulsed laser deposition.

II. EXPERIMENT

FePt films with a thickness of 40 nm have been grown on single crystalline MgO(001) substrates by means of pulsed laser deposition (PLD) under pressure conditions from ultrahigh vacuum (UHV, base pressure less than 10^{-8} mbar) up to 0.3 mbar forming gas atmosphere containing 95% Ar and 5% H_2 ("pressure series") as well as temperatures ranging from room temperature to 950 °C ("temperature series"). A description of the PLD setup can be found in Ref. 17. All films have been deposited with 64 repetitions of 8 pulses Pt and 20 pulses Fe at 5 J/cm² laser fluence. The deposition rate was measured before with an

Inficon XTM/2 rate monitor. For the pressure series, the substrate was heated to 800 ± 50 °C during deposition. (This is the uncertainty for an absolute temperature measurement of a thin, partially transparent film by pyrometer. The relative temperature uncertainty between the films in this series is much lower.) On the other hand, the temperature series was conducted under a constant pressure of 0.1 mbar. Film composition was controlled either after deposition by energy dispersive x-ray spectroscopy or *in situ* by a Staib DESA 100 Auger electron spectrometer in order to arrive at the stoichiometric composition.¹⁸ To evaluate the crystal structure a Siemens D5000 x-ray diffractometer (Co K_{α} radiation) was used, and the surface morphology was determined by a Digital Instruments Dimension 3100 atomic force microscope (AFM) in tapping mode. Magnetic characterizations were done by vibrating sample magnetometry in a Quantum Design PPMS 6000.

III. RESULTS

Figure 1 shows the deposition rate of Fe and Pt versus gas pressure. Starting from vacuum conditions, the rates increase slightly for both elements up to a pressure of 0.01 mbar, where a maximum in the deposition rate is observed. This effect is due to reduced sputtering at the film

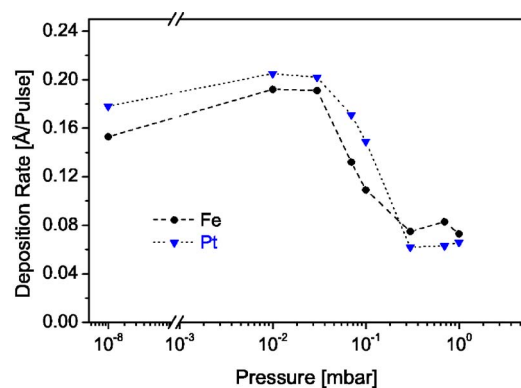


FIG. 1. (Color online) Pressure dependence of the deposition rates of Fe and Pt.

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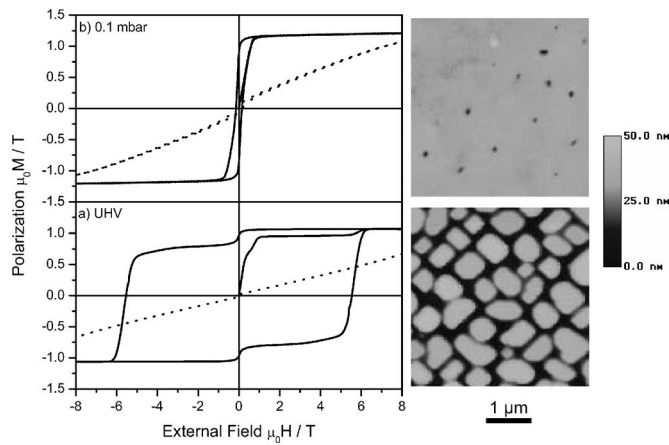


FIG. 2. Out-of-plane (solid line) and in-plane (dashed line) magnetization loops and AFM surface structure of 40 nm thick FePt films deposited under UHV conditions (a) and under a forming gas pressure of 0.1 mbar (b).

surface, as shown by Sturm and Krebs.¹⁹ Above 0.01 mbar, the plasma plume is strongly scattered by the background gas and the rates decrease quickly to one third of the maximum values at pressures ≥ 0.3 mbar.

Figure 2 compares the hysteresis loops and the surface structure of a film prepared at 800 °C under UHV conditions (base pressure 10^{-8} mbar) and under a pressure of 0.1 mbar, where the deposition rate shows the steepest decline (Fig. 1). In the pressure range between UHV and 0.1 mbar the coercivity drops fast with increasing pressure, reaching 0.7 T at 0.01 mbar. The conditions at UHV and 0.1 mbar result both in a magnetic out-of-plane texture. From the extrapolation of the almost linear in-plane magnetization curve to the saturation value, anisotropy fields of $H_A \approx 11(1)$ T and $\approx 9(1)$ T can be inferred for the film deposited under UHV and 0.1 mbar, respectively. These are close to the value for bulk FePt of 11.6 T,²⁰ indicating a well ordered $L1_0$ phase.

This is consistent with x-ray diffraction measurements, where a strong (001) superstructure peak is observed in both cases (Fig. 3). No other orientations besides the (00 l) group are seen, indicating epitaxial growth on the MgO(100) surface. The chemical order parameter S has been estimated

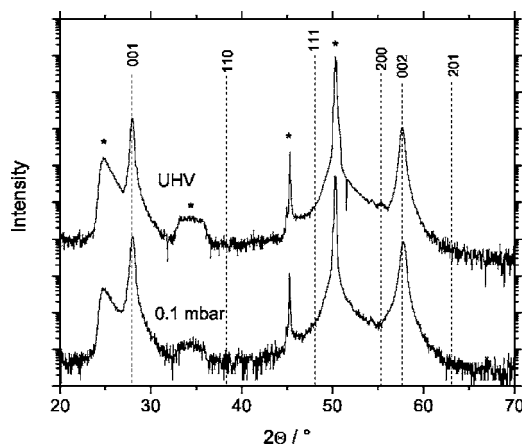


FIG. 3. X-ray diffraction patterns of FePt deposited under UHV conditions and 0.1 mbar, with indices for the $L1_0$ FePt phase. Substrate peaks are marked by an asterisk (*).

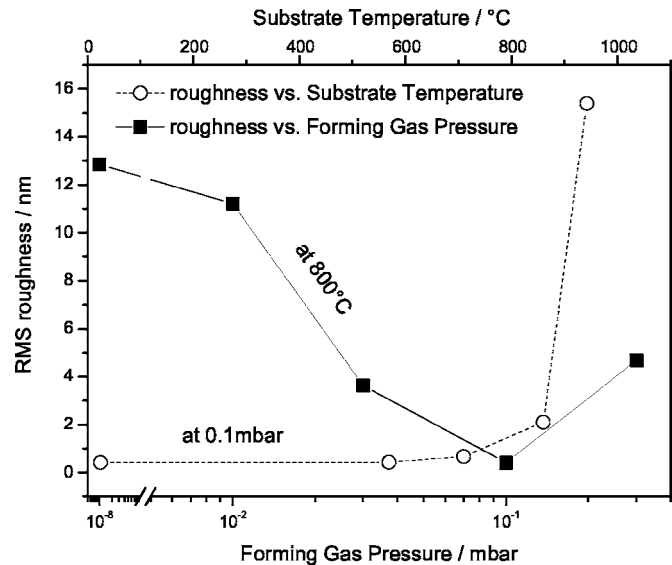


FIG. 4. rms roughness of FePt films as a function of deposition temperature (at 0.1 mbar) and pressure (at 800 °C).

from the intensity ratio of the (001) and (002) peaks according to

$$S^2 = \left(\frac{I_{001}}{I_{002}} \right)_{\text{meas}} / \left(\frac{I_{001}}{I_{002}} \right)_{\text{calc}}, \quad (1)$$

with $(I_{001}/I_{002})_{\text{calc}} = 2$.²¹ $S = 0.88$ for the UHV deposited film and $S = 0.82$ for the film deposited at 0.1 mbar. However, both surface morphology and coercivity are markedly different. Under UHV conditions the film grows in isolated islands and shows a very high coercivity of 5.5 T, whereas in a background pressure of 0.1 mbar, the film is smooth (apart from some holes) and the coercivity is comparatively low at 0.15 T. For higher as well as lower pressure during deposition the film roughness is increased (Fig. 4). Higher temperature during deposition also results in increased roughness. A lower roughness can be achieved by decreasing the deposition temperature below 800 °C. However, chemical order and hard magnetic properties are reduced strongly at the same time.

IV. DISCUSSION

Since the intrinsic properties (H_A and S) of the films deposited under UHV conditions and in 0.1 mbar forming gas pressure are comparable, their strongly differing coercivities are explained solely as a result of the different surface morphologies. As has been shown previously, switching in epitaxial granular FePt occurs by domain nucleation.^{7,22} In agreement with this, a high initial susceptibility is observed in the films in this work [see Fig. 2(a)]. For the smooth film, the coercivity is low, because a reversed domain may form in low field at a defect site anywhere on the film surface and spread over the whole film. In the granular, nonconnected microstructure of the UHV deposited film, domain wall movement is confined within a grain, inhibiting the switching of the whole film. Each grain therefore switches independently of the others and the switching field is determined by

defects in each grain. The strongly increased coercivity compared to the smooth film indicates a low average density of defects per grain.

A similar, drastic increase in coercivity when the nominal film thickness drops below a critical value and the morphology changes from smooth to granular has been observed in earlier work by Ide *et al.*²³ and Shima *et al.*²² However, in this work the film thickness is kept constant and is therefore not the reason for the emerging smoothness at 0.1 mbar pressure. In order to explain the smoothness of the FePt films deposited at a pressure of 0.1 mbar, we propose that a hydrogen atmosphere increases the number of nucleation centers for island growth, preferring the growth of small islands. These nucleation centers most likely are created by the reduction of MgO by hydrogen at the surface, leaving oxygen vacancies behind. Oxygen vacancies on MgO(100) surfaces can act as strong trapping centers. For a Pd atom in such a vacancy, a trapping potential of up to 2.55 eV has been observed.^{24,25} Assuming a similarly high trapping energy for Pt, trapped Pt atoms will act as nucleation centers for the growth of FePt islands. With increasing hydrogen pressure, the number of nucleation sites increases, resulting in lower average island size and therefore lower roughness (Fig. 4). This process competes with another process that creates nucleation sites and which is a typical feature of pulsed laser deposition: nucleation sites are introduced by the impinging particles of the laser plasma, which have kinetic energies above the sputtering/implantation threshold,²⁶ as well as by the extremely high deposition rate during each pulse. These processes are generally made responsible for the very smooth films prepared by pulsed laser deposition. As deposition rate strongly decreases above 0.1 mbar (Fig. 1), which is accompanied by a decrease of kinetic energy,²⁷ creation of nucleation sites from this process ceases and the roughness increases again (Fig. 4).

Figure 4 also shows the temperature dependence of the root mean square (rms) roughness at a constant pressure of 0.1 mbar. With increasing temperature and thermal mobility, roughness increases slowly at first, as it is predominantly determined by the constant nucleation center density. However, even for higher temperatures of up to 1000 °C that have been applied in these experiments, the trapping potential is too deep to allow dissociation of the Pt atoms from a vacancy. In order to explain the sudden increase of roughness above 800 °C, it is argued that the critical grain size for nucleation increases from one Pt atom to three or more Pt atoms, as has been suggested by Haas *et al.*²⁸ for Pd on the MgO(100) surface. Pt islands of three or fewer atoms are not stable any more at these temperatures. They dissociate and agglomerate at larger islands that can act as nucleation sites. This leads to increased roughness.

Although hydrogen aids the formation of continuous and out-of-plane ordered FePt films at 800 °C, at lower temperatures the degree of ordering is strongly reduced. No influence of hydrogen on the ordering temperature has been observed compared to UHV deposition. This is in contrast to the effects of hydrogen during post-annealing of FePt disordered films, where a significant decrease of the ordering temperature has been observed.^{15,16} Films prepared under pure argon

support the conclusion of the important role of hydrogen to achieve a continuous and smooth film. At pressures of 0.1 mbar pure argon as process gas, a granular and irregular surface morphology was observed, with rms roughness of 4.7 nm.

V. CONCLUSIONS

Pulsed laser deposition allowed us to examine the influence of a hydrogen atmosphere in a broad pressure and temperature range on the growth of FePt thin films. Smooth, *c*-axis oriented $L1_0$ phase FePt was obtained on MgO(100) in a narrow parameter window at 800 °C substrate temperature and 0.1 mbar forming gas pressure. In contrast to granular, highly coercive films obtained under UHV conditions, such films are well suited for structuring and may be useful for microelectromechanical systems devices. In agreement with the concept of nucleation controlled switching, the coercivity in these continuous, unpatterned films is quite low. The microstructural change is attributed to a change of the island nucleation behavior at hydrogen induced defects on the MgO surface. No change in the ordering temperature has been observed due to the presence of hydrogen.

ACKNOWLEDGMENT

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