Columnar grain growth of FePt($L1_0$) thin films

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An experimental approach for obtaining perpendicular FePt-SiOx thin films with a large height to diameter ratio FePt($L1_0$) columnar grains is presented in this work. The microstructure for FePt-SiOx composite thin films as a function of oxide volume fraction, substrate temperature, and film thickness is studied by plan view and cross section TEM. The relations between processing, microstructure, epitaxial texture, and magnetic properties are discussed. By tuning the thickness of the magnetic layer and the volume fraction of oxide in the film at a sputtering temperature of 410 °C, a 16 nm thick perpendicular FePt film with ∼8 nm diameter of FePt grains was obtained. The height to diameter ratio of the FePt grains was as large as 2. Ordering at lower temperature can be achieved by introducing a Ag sacrificial layer. © *2012 American Institute of Physics*. [doi:10.1063/1.3679463]

FePt($L1_0$) is considered to be one of the most promising materials for ultrahigh density recording applications.[1–4] For achieving high density, the film media needs to be granular in nature, of small and uniform sized grains with a non-magnetic phase at the grain boundaries.[5–7] Well defined carbon regions at the grain boundaries with small magnetic grain sizes have been achieved in FePt($L1_0$)-C films. However, non-columnar film microstructures in these films leads to a layer with new secondary grains, leading to undesired magnetic properties.[8] The difficulty in obtaining columnar growth of $L1_0$ FePt grains with large height to diameter ratios lies in perhaps the high surface energy associated with the material choice.

In this work, an experimental method for obtaining perpendicular FePt-SiOx thin films with large height to diameter ratio FePt($L1_0$) columnar grains is presented. It was found that the microstructure of composite films can be influenced by not only the deposition pressure, temperature, and oxide volume fractions, but also the choice of matrix material and thickness of the thin film.

FePt/oxide multilayers and MgO/Ta underlayers were deposited on Si substrates by RF sputtering. The base pressure was approximately 5×10^{-7} Torr and the argon pressure during deposition was maintained at 10 mTorr. The MgO/Ta underlayers were deposited at room temperature; the FePt/oxide multilayers were fabricated by the alternating sputtering of the $Fe_{55}Pt_{45}$ alloy and the SiOx targets on a heated substrate. The substrate temperature was varied between 370 °C to 475 °C during the multilayer deposition. X-ray diffraction and transmission electron microscopy were used to study the texture and microstructure of the films. An alternating gradient force magnetometer (AGFM) and a polar magneto-optical Kerr effect looper (MOKE) were used to investigate the magnetic properties.

The cross section TEM images in Fig. 1 show the microstructure of FePt-SiOx composite films as a function of oxide volume fraction (OVF). For this set of samples, the FePt/SiOx multilayers deposition temperature was maintained at 410 °C and the FePt film thickness was fixed at 16 nm. As shown in the figure, in the low OVF films, the FePt grains have a smaller height to diameter ratio. Increasing the OVF produces FePt grains with smaller diameter and more columnar shaped FePt grains. When the OVF reaches 37% (Fig. 1(c)), ∼8 nm diameter columnar FePt grains are obtained with 16 nm height. Further increasing the OVF leads to a separation in some of the FePt columnar grains followed by the formation of a secondary layer of FePt grains, as shown in Fig. 1(d).

To study the microstructure of FePt-SiOx composite films as a function of deposition substrate temperature, samples with the same OVF (37%) and the same film thickness (16 nm) were deposited at different substrate temperatures. As shown in Fig. 2(a), lowering the deposition temperature from 410 °C to 370 °C leads to a double layer structure of FePt grains. The grains in the top layer are tapered with a dome like shape due to the limited surface diffusion and self-shadowing effects. When the deposition temperature is raised from 410 °C to 475 °C, bulk diffusion starts to dominate. A second layer of equiaxial spherical FePt grains is formed. In order to obtain a single layer of columnar FePt grains with well-defined grain boundaries, the deposition temperature has to be moderate so that the microstructure is mainly surface diffusion controlled.

The film microstructure as a function of FePt layer thickness is presented in Fig. 3. Here the OVF was fixed at 45% and the deposition temperature was fixed at 410 °C. The film thickness was varied from 9 nm to 16 nm and 12 nm thick films exhibited columnar grains with grain diameter of about 7 nm. Thinner FePt layers also formed columnar grains, as shown in Fig. 3(a). The samples in both Fig. 3(a) and Fig. 3(b) have excellent perpendicular texture, but those in Fig. 3(b) have a larger order parameter, possibly due to

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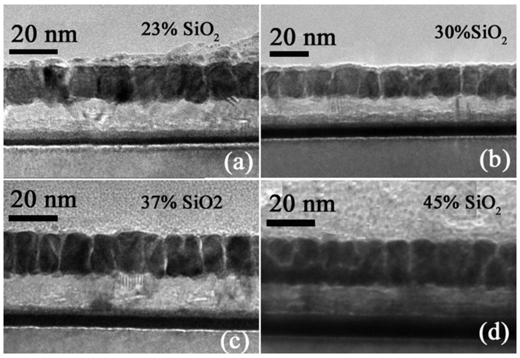

FIG. 1. Cross section TEM images of 16 nm composite FePt-SiOx thin films deposited on MgO/Ta underlayers at 410 °C with different oxide volume fractions in the film.

the larger volume of the grains. Further increasing the thickness of the magnetic layer again leads to a separation in some of the FePt columnar grains. In Fig. 3(c), a secondary FePt layer of grains forms. With 55% SiOx in the film, ~5.4 nm diameter columnar FePt grains in 9 nm thick films have been obtained.

Yuan has modified the well-known Thornton structure zone model[9] to include OVF as a variable in thin film microstructures at a fixed temperature.[10] Here we include the plan view TEM images to better classify the microstructures. Figure 4 confirms that the grain diameters decrease as the OVF increases. At low OVF (23%), large FePt grains with ill-defined grain boundaries are formed. Increasing the OVF (30%) results in elongated worm shape FePt grains, which form a maze-like microstructure. Further increasing the OVF reduces the interconnections between the FePt grains; a

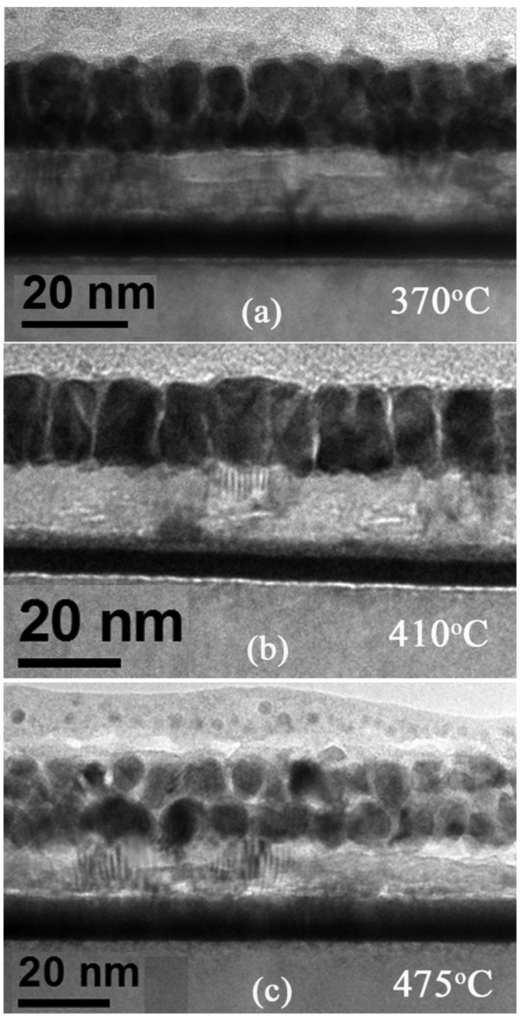

FIG. 2. Cross section TEM images of 16 nm composite FePt-37%SiOx thin films deposited on MgO/Ta underlayers at different substrate temperatures.

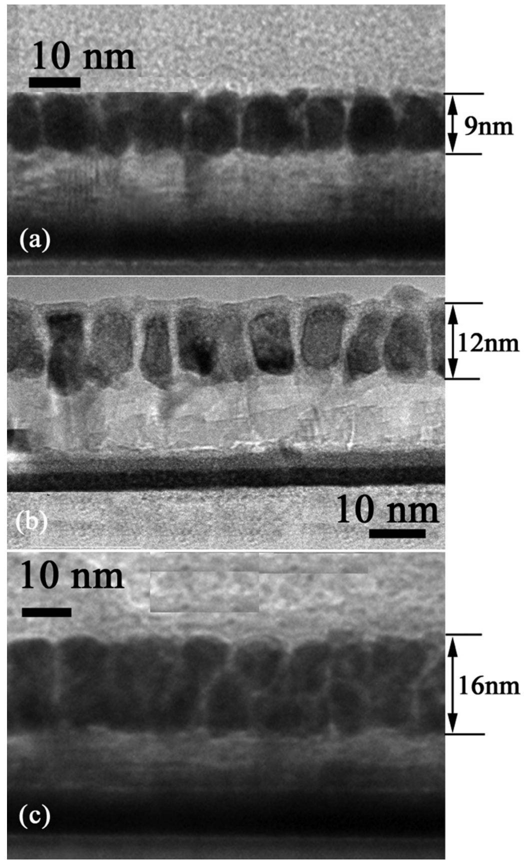

FIG. 3. Cross section TEM images of composite FePt-45%SiOx thin films deposited on MgO/Ta underlayers at 410 °C with different thicknesses of FePt layer.

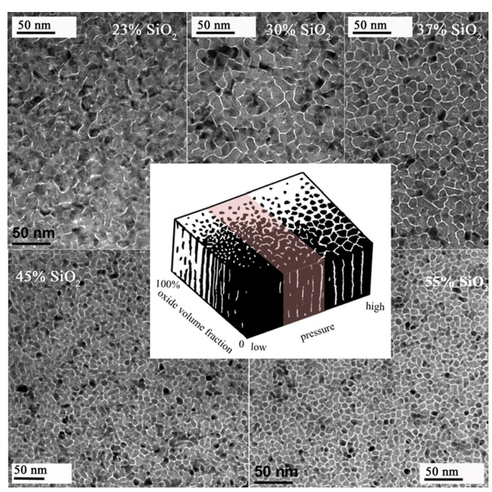

FIG. 4. (Color online) plan view TEM images of composite FePt-SiOx thin films deposited on MgO/Ta underlayers at 410 °C with different oxide volume fractions in the film.

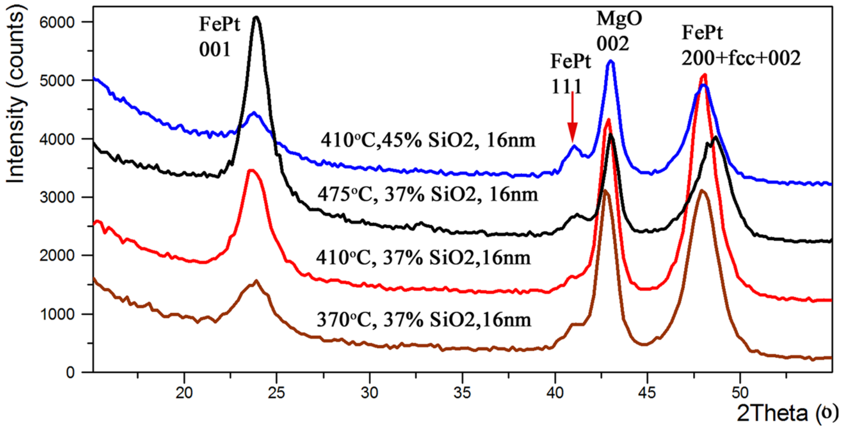

FIG. 5. (Color online) X-ray diffraction patterns of composite FePt-SiOx thin films deposited on MgO/Ta underlayers.

granular microstructure gradually forms with well-defined oxide phases at the grain boundaries. From the composite thin film structure zone model shown in the insert of Fig. 4, the growth of FePt-SiOx composite films deposited at 10 mT, 410 °C here, falls in the shaded middle zone, where the self-shadowing effects and surface diffusion play more or less an equal role. From this zone model, we see that grains completely isolated by the second phase oxide can be obtained not only by increasing the OVF, but also by increasing the Ar deposition pressure. Other than Ar pressure and OVF, variables such as temperature, choice of oxide materials, and the choice of underlayer materials will result in a shift of the zone boundaries and therefore have great influence on the microstructures. For instance, at a slightly increased deposition temperature, the middle maze-like zone will shift along the pressure direction to a higher value. However, because diffusion increases dramatically with the increasing of deposition temperature, it will dominate the self-shadowing effect. Therefore, the pressure has much less influence on the microstructures at higher temperatures.

In order to study the relations between the microstructure and the film texture, x-ray diffraction patterns for samples with the same film thickness (16 nm) but different microstructures are presented in Fig. 5. As shown in the figure, the formation of the secondary FePt grains (by varying temperature or OVF or thickness) increases the (111) FePt peak intensity. Therefore, we must suppress the formation of secondary FePt crystals and form columnar FePt grains to obtain excellent perpendicular texture of the film.

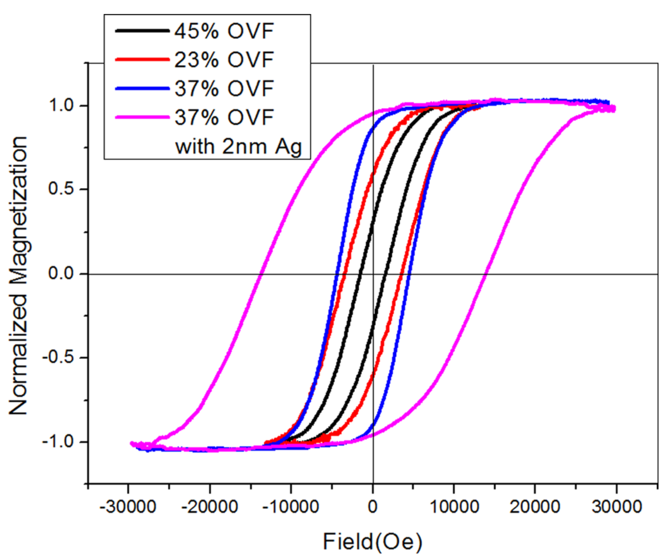

FIG. 6. (Color online) Out of plane hysteresis loops for 16 nm FePt-SiOx thin films deposited at 410 °C on MgO/Ta underlayers.

Figure 6 shows the magnetic hysteresis loops for 16 nm FePt films deposited at 410 °C with different OVF, corresponding to the microstructures shown in Fig. 1. From Fig. 6, it can be seen that with the same deposition temperature and film thickness, high OVF results in low atomic ordering of $L1_0$ FePt. Columnar growth of FePt (37% OVF) results in better orientation and better magnetic decoupling of the FePt grains, therefore better magnetic properties. By introducing a 2 nm Ag sacrificial layer[11] underneath the FePt-SiOx composite film, the coercivity was improved from 4.5 kOe to 13 kOe while maintaining the columnar mircrostructure.

Producing FePt films with thickness to grain ratios of about 2 has been demonstrated. Factors such as temperature, pressure, and oxide volume fraction (OVF) must be carefully controlled to obtain these columnar grains. A microstructure zone model with OVF and pressure variables is used to understand the role of the above variables.

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[1]T. J. Klemmer, N. Shukla, C. Liu, X. W. Wu, E. B. Svedberg, O. Mryasov, R. W. Chantrell, D. Weller, M. Tanase, and D. E. Laughlin, Appl. Phys. Lett. **81**, 2220 (2002).
[2]Y. N. Hsu, S. Jeong, D. E. Laughlin, and D. N. Lambeth,. Magn. Magn. Mater. **260**, 282 (2003).
[3]M. H. Hong, K. Hono, and M. Watanabe, J. Appl. Phys. **84**, 4403 (1998).
[4]T. Ichitsubo, S. Tojo, T. Uchihara, E. Matsubara, A. Fujita, K. Takahashi, and K. Watanabe, Phys. Rev. B **77**, 094114 (2008).
[5]K. Aimuta, K. Nishimura, S. Hashi, and M. Inoue, IEEE Trans. Magn. **41**, 3898 (2005).
[6]Y. F. Xu, J. S. Chen, and J. P. Wang, Appl. Phys. Lett. **80**, 3325 (2002).
[7]E. Yang and D. E. Laughlin, J. Appl. Phys. **104**, 023904 (2008).
[8]A. Perumal, Y. K. Takahashi, and K. Hono, J. Appl. Phys., **105**, 07A304 (2009).
[9]H. Yuan and D. Laughlin, in *Nanoparticle Materials and Nanostructured Films*, edited by K. Lu et al. (Wiley, Hoboken, NJ, 2010), Vol. 223.
[10]H. Yuan, PhD dissertation, Carnegie Mellon University, 2009.
[11]E. Yang, D. E. Laughlin, and J. G. Zhu, IEEE Trans. Magn. **46**, 2446 (2010).