9-1-2002

Permanent-Magnet Properties of Thermally Processed FePt and FePt–Fe Multilayer Films

Jian Zhou
University of Nebraska - Lincoln

Ralph Skomski
University of Nebraska-Lincoln, rskomski2@unl.edu

XINGZHONG LI
Nebraska Center for Materials and Nanoscience, xli2@unl.edu

Wei Tang
Department of Physics and Astronomy, University of Delaware, Newark, DE

George C. Hadjipanayis
University of Delaware, hadji@udel.edu

See next page for additional authors

Permanent-Magnet Properties of Thermally Processed FePt and FePt–Fe Multilayer Films

Jian Zhou, Ralph Skomski, Xingzhong Li, Wei Tang, George C. Hadjipanayis, and David J. Sellmyer

***Abstract*—FePt single-layer and FePt–Fe multilayer thin films are prepared by magnetron sputtering. By varying the Pt content, FePt, Fe_3Pt, or a mixture of FePt and Fe_3Pt can be obtained in Fe–Pt single layers. In annealed FePt–Fe multilayers, the coercivities decrease with the introduction of Fe layers compared to FePt single layers, while the magnetization increases. The single-phase behavior of the hysteresis loops of FePt–Fe multilayers indicates the existence of exchange coupling in these materials. For one FePt–Fe sample with optimized exchange coupling, the intrinsic properties correspond to an energy product of 19 MGOe. The texture of the magnets is determined by the (111) orientation of the crystallites. This means that the easy magnetization directions of the FePt grains form an angle of 54.7° with the film normal, but are randomly oriented in the film plane. It is analyzed how this easy-axis distribution affects the magnetic hysteresis.**

***Index Terms*—Annealing, permanent magnets, Pt–Fe, texture.**

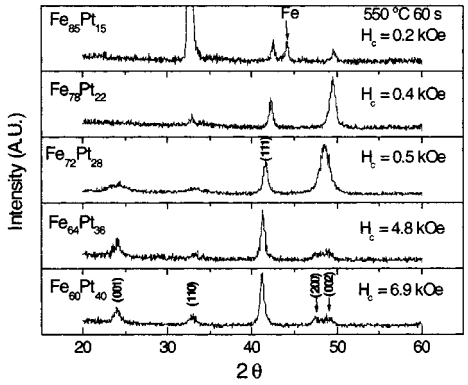

Fig. 1. XRD for Fe–Pt single layer films with different composition. All the samples are annealed at 550 °C for 60 s.

I. Introduction

SINCE THE concept of exchange-spring magnets was introduced by Kneller and Hawig [1], FePt and CoPt emerged as candidates for hard–soft exchange-coupled nanocomposites with appreciable hard-magnetic properties [2]–[4]. Yung *et al.* investigated the microstructure of Fe–Pt thin films and obtained an energy product of 15 MGOe [2], whereas Liu *et al.* developed a rapid annealing process and obtained an energy product of more than 50 MGOe in an Fe–Pt nanocomposite thin film [3]. Exchange-spring bilayer films of CoPt/Co were investigated by Crew *et al.* [5].

Previous work has shown that the as-deposited cubic-phase Fe–Pt may form a tetragonal phase after heat treatment if the Fe concentration is between 50%–75% [2]. When more Fe is added, the Fe_3Pt phase will form. Liu *et al.* found that after proper annealing, hard–soft exchange-coupled nanocomposite is realized in the Fe–Pt multilayer thin film with Fe concentration around 64% [3].

An important aspect of exchange-spring magnets is the degree of crystalline alignment or *texture*. Nanostructured permanent magnets exhibit a broad range of textures, from isotropic [6] to partially aligned [3], whereas the theoretical predictions made in [7] imply perfect alignment. The FePt structures discussed in this paper exhibit a particular (111) texture with random in-plane anisotropy.

In this paper, we report the magnetic properties of thermally processed FePt and FePt–Fe multilayer thin films and discuss the micromagnetism of the (111)-textured thin films.

II. Sample Preparation and Characterization

We use three methods to prepare the Fe–Pt thin films. Method I is to sputter the Fe target with Pt pellets on it. This gives a homogenous Fe–Pt single layer film. The approximate composition is determined by comparing the lattice parameters with those known Fe–Pt thin films. FePt–Fe multilayer is prepared by sputtering FePt and Fe targets. Method II is to sputter Fe and Pt targets to get Fe–Pt multilayers. Method III is similar to Method II, except that some Fe layers are thicker than the remaining Fe layers, to create regions with enhanced Fe concentration. A typical structure is $(Fe8Pt10Fe8Pt10Fe30Pt10Fe8Pt10) \times 5$ (Å). All the samples are sputtered on an Si (100) substrate. The total thickness of the films is between 500–1000 Å. As-deposited samples are heat-treated at 550 °C by rapid thermal annealing for 10–300 s or oven annealed in vacuum for 30 min. The crystal structure is determined by X-ray diffraction (XRD), whereas superconducting quantum interference device (SQUID) magnetometer is used to measure the magnetic properties.

III. Magnetic Properties

Fig. 1 shows the XRD patterns of Fe–Pt single layer thin films rapidly annealed at 550 °C for 60 s (Method I). For Fe concentrations of more than 80%, a two-phase mixture of Fe and Fe_3Pt is obtained. When the Fe concentration is between 70%–80%,

Manuscript received February 14, 2001. This work was supported by the Department of Energy, by the Air Force Office of Scientific Research, and CMRA.

J. Zhou, R. Skomski, X. Li, and D. J. Sellmyer are with the Center for Materials Research and Analysis (CMRA), University of Nebraska, Lincoln, NE 68588 USA (e-mail: jzhou@unlserve.unl.edu; rskomski@unlserve.unl.edu; xli2@unl.edu; dsellmyer1@unl.edu).

W. Tang and G. C. Hadjipanayis are with the Department of Physics and Astronomy, University of Delaware, Newark, DE 19716 USA (e-mail: weitang@udel.edu; hadji@udel.edu).

Digital Object Identifier 10.1109/TMAG.2002.803109.

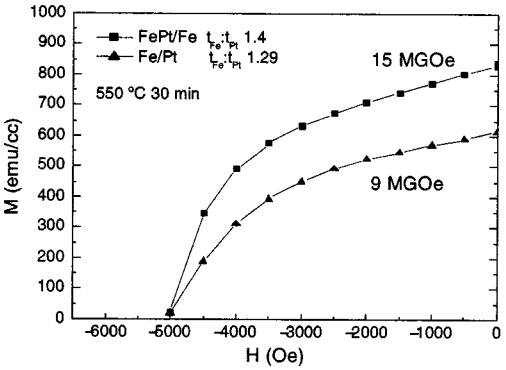

Fig. 2. Demagnetization curves for annealed FePt–Fe and Fe–Pt films.

TABLE I
MAGNETIC PROPERTIES OF FePt MULTILAYER THIN FILMS PREPARED BY METHOD III. THE SAMPLES ARE RAPIDLY ANNEALED AT 550 °C FOR 60 s

$t_{Fe}:t_{Pt}$	M_s (emu/cc)	M_r (emu/cc)	$(BH)_{max}$ (MGOe)	Hc (kOe)	Hc (methodII) (kOe)
1.04	830	720	17	10.9	10.0
1.11	900	740	17	8.6	8.3
1.16	920	760	18	7.8	7.5
1.23	980	810	19	7.8	6.3
1.25	1000	800	18.5	7.3	6.0
1.34	1080	850	15.5	5.3	4.4
1.35	1060	850	16.2	5.3	4.2
1.4	1090	860	14.3	4.6	3.3

the only phase observed is Fe_3Pt. For Fe less than 70%, the appearance of (200) and (002) peaks indicate the existence of tetragonal FePt phase (with a small amount of Fe_3Pt). The corresponding hysteresis loops of these samples show single-phase behavior with in-plane anisotropy. The observed coercivities confirm the results of the X-ray phase analysis. Since Fe_3Pt is a relatively soft magnetic phase, the H_c of Fe_3Pt is less than 0.5 kOe. For the $Fe_{60}Pt_{40}$ sample, a coercivity of larger than 10 kOe can be reached when annealing at 600 °C for 60 s.

To obtain regions with locally enhanced Fe concentrations, we prepared FePt–Fe multilayers (Method III). Fig. 2 shows the comparison of the demagnetization curves of annealed FePt–Fe and Fe–Pt multilayers. It can be seen that the additional Fe increases the magnetization, while the coercivity remains largely unchanged.

Table I shows that magnetic properties of Fe–Pt thin films prepared by Method III. The coercivities are about 1–2 kOe larger than that of the films with same Fe : Pt ratio made by Method II. This indicates that a hard phase exists in samples made by Method III, while those made by Method II have lower H_c because they are more homogenous and rather soft magnetic. Furthermore, the saturation magnetizations M_s and remanences M_r of the samples made by Method III are larger than those made by Method II. A corresponding maximum energy product of 19 MGOe is obtained with the Fe–Pt ratio 1.23, while the corresponding Fe–Pt layer gives 15 MGOe. This proves that our way of creating soft-magnetic regions yields good exchange coupling and fairly good magnetic properties. The dc demagnetization measurement (recoil measurement) also indicates the exchange-spring behavior of hard–soft phase nanocomposite.

Fig. 3 shows the plan-view TEM micrograph for the sample with energy product of 19 MGOe. It can be seen that the

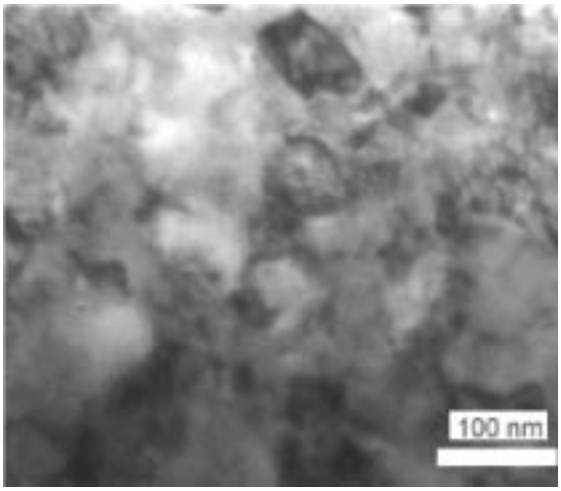

Fig. 3. TEM micrograph of Fe–Pt film with Fe : Pt thickness ratio of 1.25.

nonocrystallites form with a large grain size distribution from less than 10–70 nm. Electron diffraction Debye ring also confirms the existence of both tetragonal and fcc structures; the latter may correspond to soft phase.

Theoretical calculations have shown that for good exchange coupling, the dimension of the soft phase should not be larger than about twice the domain wall thickness of the hard phase [7]. Kim *et al.* measured the CoPt/Co bilayer and found that an annealed CoPt/Co(6.3 nm) shows exchange-coupled behavior [8], while CoPt has domain wall width of 4.5 nm. For tetragonal FePt, the domain wall thickness is around 4 nm [4]. In our experiment, we prepared the thin film using Method III with an Fe layer as thick as 8.5 nm. After rapid annealing at 550 °C for 0–60 s, the XRD patterns show that the Fe peak shown in as-deposit film disappears and the hysteresis loops show single-phase behavior. This indicates: 1) that free Fe is difficult to form in FePt—due to pronounced diffusion, the soft phase is Fe-rich Fe_3Pt and 2) that 8.5-nm-thick Fe can be coupled by the tetragonal FePt hard phase after annealing.

Fig. 4(a) shows the XRD patterns of Fe–Pt–Fe films made by Method III. The accumulative thickness ratio of Fe–Pt is 1.23. It can be seen that short time annealing gives almost (111) texture and the longer annealing time leads to the development of other peaks. Loop measured parallel to the film plane shows $M_r/M_s > 0.8$, whereas perpendicular loop gives $M_r/M_s > 0.7$ with a hard-axis shape [see Fig. 4(b)]. The enhancement of the M_r/M_s ratio shows the exchange coupling and the easy–hard axis behavior shows the effect of partial texture.

Since the magnetic anisotropy of tetragonal FePt reflects the layered structure of the $L1_0$ phase rather than the distortion of the unit cell, the magnetic behavior of the present structure differs from that of cubic (111) crystallites. The angle θ between the local easy axis and the film normal has the common value $\theta_o = \arccos(1/\sqrt{3})$, but the angle φ describing the in-plane orientation of the crystallites is a random quantity. The uniaxial texture is described by the probability $P(\theta)$, where the angle θ describes the c-axis orientation with respect to the film normal. (In terms of P, unit-sphere integration is realized by

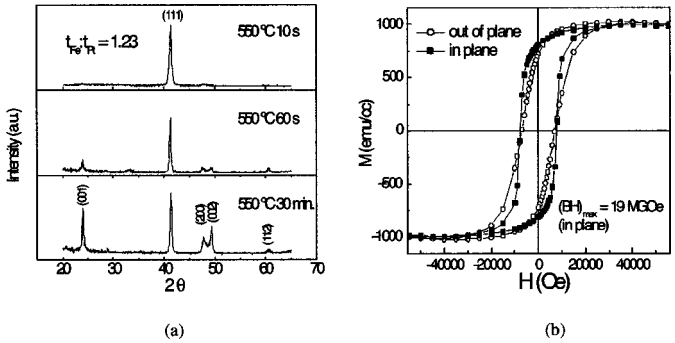

Fig. 4. (a) XRD of Fe–Pt made by Method III (the total thickness ratio of Fe to Pt is 1.23) and (b) the corresponding loop for annealing for 60 s.

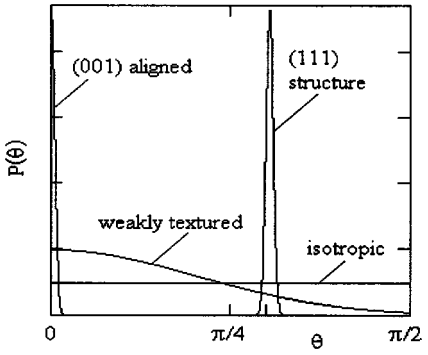

Fig. 5. Calculated texture of thin-film magnets. $P(x)$ describes the angular distribution of the easy-axis direction.

$2\pi \int P(\theta) \sin\theta d\theta = 4\pi$.) Fig. 5 shows that the distribution $P(\theta)$ is peaked at θ_o.

Depending on the size of the crystallites, there are two hysteretic regimes. Crystallites larger than the Bloch-wall width of the hard phase, about 5 nm, can in fair approximation be considered as interaction-free Stoner–Wohlfarth particles. Due to random in-plane orientation of the easy axes averaging, the in-plane remanence is close to zero. The perpendicular remanence reflects a competition between the uniaxial type shape anisotropy of the film and the (111)-type magnetocrystalline anisotropy. Starting from the general expression for the micromagnetic free energy [7] and taking into account that $\nabla \cdot \mathbf{M} \approx 0$ inside the grains, yields the magnetization angle

$$\tan(2\theta_r) = \frac{\sin(2\theta_o)}{\cos(2\theta_o) - \frac{\mu_o M_s^2}{2K_1}}. \quad (1)$$

Fig. 6 shows the corresponding remanence ratio $M_r/M_s = \cos(\theta_r)$ as a function of the anisotropy K_1 of the crystallites. We see that the remanence and, consequently, the coercivity, are nonzero, except in the limit $K_1 = 0$. This is confirmed by the experiment that the remanence is above 0.6 in both in-plane and normal to plane measurements.

In the case of very small grains, which is less relevant in the present context, the hysteretic behavior is governed by random-anisotropy effects. In the limit of strong exchange coupling, the magnetization is uniform on a macroscopic scale, and the net anisotropy is equal to the "coherent" [9] anisotropy obtained after structural averaging. Interestingly, this structural averaging over all angles θ yields a *zero* net magnetocrystalline anisotropy,

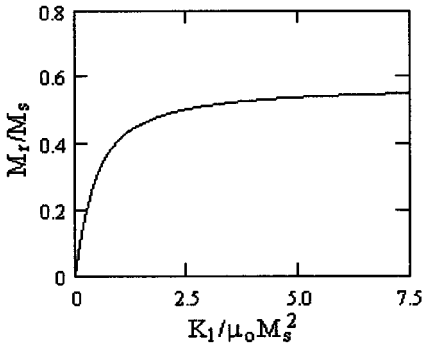

Fig. 6. Remanence of polycrystalline Pt–Fe films as a function of the fraction of anisotropy of the crystallite. Depending on the degree of $L1_0$-type ordering of the Pt atoms in (001) planes, the anisotropy constant K_1 varies from nearly 0 to 6.6 MJ/m^3. The latter value corresponds to $K_1/\mu_o M_s^2 = 4.1$.

and the magnetization is determined by the shape anisotropy. Note that this zero average is a specific consequence of the (111) texture; other textures yield nonzero magnetocrystalline anisotropy contributions.

IV. Conclusion

We have investigated how the fabrication process affects the structural and magnetic properties of the Fe–Pt thin films. The first method co-sputters Fe and Pt with Fe concentrations of 60% to 85% and yields FePt or Fe_3Pt or a mixture of them, depending on the composition. By comparison, Methods II and III make normal or Fe-rich multilayers. After being rapidly thermal annealed, an improved energy product of as much as 19 MGOe is obtained. The hysteretic behavior of the magnets reflects the competition between shape and magnetocrystalline anisotropy. The (111) texture means that the easy magnetization directions of the crystallites form a common angle with the film normal. This gives rise to perpendicular hysteresis, except in the limit of strong exchange coupling, where both the in-plane and perpendicular magnetocrystalline anisotropies average to zero.

References

[1] E. F. Kneller and R. Hawig, "The exchange-spring magnet: A new material principle for permanent magnets," *IEEE Trans. Magn.*, vol. 27, pp. 3588–3600, July 1991.

[2] S. W. Yung, Y. H. Chang, T. J. Lin, and M. P. Huang, "Magnetic properties and microstructure of iron–platinum thin films," *J. Magn. Magn. Mater.*, vol. 116, pp. 411–418, 1992.

[3] J. P. Liu, C. P. Luo, Y. Liu, and D. J. Sellmyer, "High energy products in rapidly annealed nanoscale Fe/Pt multilayers," *Appl. Phys. Lett.*, vol. 72, pp. 483–485, 1998.

[4] R. Skomski and J. M. D. Coey, *Permanent Magnetism*. London, U.K.: Inst. Phys., 1999.

[5] D. C. Crew, L. H. Lewis, J. Kim, and K. Barmak, "Magnetic signature of compositional gradient in exchange-spring bilayer films of CoPt/Co," *J. Appl. Phys.*, vol. 89, pp. 7528–7530, 2001.

[6] R. Coehoorn, D. B. de Mooij, J. P. W. B. Duchateau, and K. H. J. Buschow, *J. Phys.*, vol. 49, no. C-8, p. 669, 1988.

[7] R. Skomski and J. M. D. Coey, "Giant energy product in nanostructured two-phase magnets," *Phys. Rev. B, Condens. Matter*, vol. 48, pp. 15 812–15816, 1993.

[8] J. Kim, K. Barmak, M. De Graef, L. H. Lewis, and D. C. Crew, "Effect of annealing on magnetic exchange coupling in CoPt/Co bilayer thin films," *J. Appl. Phys*, vol. 87, pp. 6140–6142, 2000.

[9] E. M. Chudnovsky, W. M. Saslow, and R. A. Serota, "Ordering in ferromagnets with random anisotropy," *Phys. Rev. B, Condens. Matter*, vol. 33, pp. 251–261, 1986.