An X-Ray Diffraction-Based Method for Evaluating Inhomogeneous Ordering at the Grain Level of $L1_0 - FePt$ Media

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A technique based on x-ray diffraction (XRD) is introduced to understand the variation of the grain-to-grain $L1_0$ ordering in FePt recording media. We classify the possible ways that the grains may be ordered into three types: homogeneous ordering, inhomogeneous ordering, and bipolar ordering. We study the possible impact of each type of ordering on the XRD spectra in terms of the relative angular positions of the FePt (001) and (002) peaks. XRD peak profile fitting is carried out to identify the grain ordering type in $FePt - SiO_x$ thin films sputtered with *in-situ* heating.

***Index Terms*—Grain, $L1_0$ FePt, ordering variation, thin film media, x-ray diffraction.**

I. Introduction

$L1_0$ FePt based thin film media with C or SiO_x grain boundaries are considered to be the future recording media for hard disk drive applications [1]–[5]. One of the important correlations between the magnetic properties and the film crystalline microstructure is that the anisotropy field value of individual grains strongly depends on the degree of atomic ordering of the $L1_0$ superstructure, and varies as a quadratic function from 0 T for a disordered A1 phase to ~11.5 T for a fully ordered $L1_0$ phase [6]. Any variation in $L1_0$ ordering from grain to grain will result in a corresponding switching field distribution, which can be one of the main sources of medium transition noise [7], [8]. Assessment of the ordering at the grain level can be carried out by using nano-beam electron diffraction techniques [9]–[11]. However, this requires intensive experimental effort and can be performed only within a very localized region of the sample. In this paper, we present a novel approach using an x-ray diffraction (XRD) technique to investigate the atomic ordering variations in $L1_0$ film media.

II. Experimental Methods

CrystalDiffract [12] was employed to generate simulated XRD patterns for $L1_0$ FePt crystals with various long-range order parameters, S. The lattice constants, namely a and c, and site occupancies of the $Fe_{50}Pt_{50}$ unit cell with respect to S are defined as follows [13]–[15]

$$S = \sqrt{\frac{1 - c/a}{1 - \left(\frac{c_0}{a_0}\right)^{S=1}}}, \tag{1}$$

$$S = P_\alpha^{Fe} - P_\beta^{Fe} = P_\beta^{Pt} - P_\alpha^{Pt}, \tag{2}$$

where a_0 and c_0 are the lattice constants of a fully ordered $L1_0$ FePt crystal ($a_0 = 3.852$ Å, $c_0 = 3.713$ Å [16]), P_α^{Fe}, P_β^{Fe}, P_α^{Pt}, and P_β^{Pt} are the fraction of α (β) sites occupied by Fe and Pt atoms (Fe on α and Pt on β), respectively. Fig. 1(a) shows the dependence of the lattice constants on S. This plot was made with the assumption that the volume change of the FePt unit cell at varying chemical order is negligible. In fact, the a-axis value of FePt FCC phase measured from our sample, which is shown later, is only ~0.4% larger than the calculated value in Fig. 1(a). The patterns created by CrystalDiffract are for x-ray powder diffraction. As far as perpendicular media is concerned, only FePt (00k) peaks were extracted for analysis. A Gaussian function was chosen for the peak profile shape and the peak width was taken to be 1.5°.

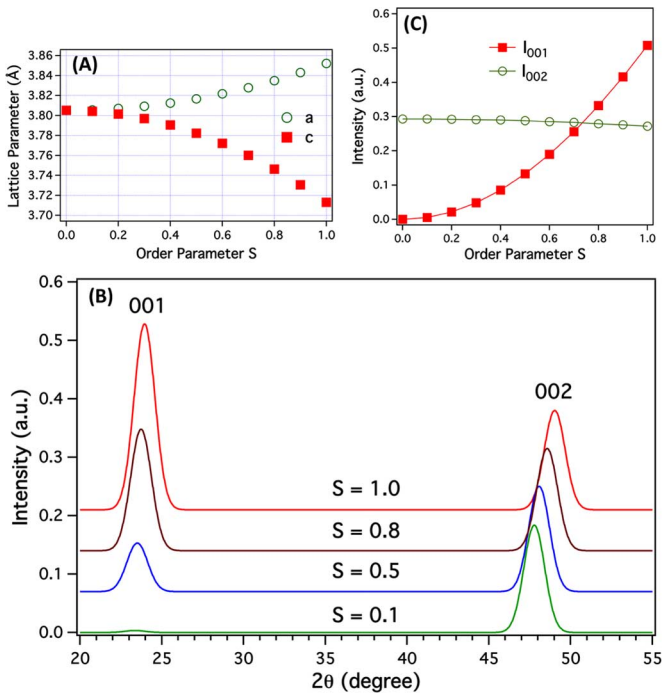

Fig. 1. (a) Lattice parameters, a and c, versus S. (b) Simulated XRD spectra of FePt film at various S. (c) Intensity of (001) and (002) peaks versus S.

Films were fabricated by RF sputtering using a Leybold Heraeus Z400 sputtering system at a base pressure $< 5 \times 10^{-7}$ Torr and an Ar sputtering pressure of 10 mTorr. $FePt - SiO_x$ media were sputter deposited as multilayers from FePt and SiO_x targets onto MgO/Ta/Si(001) substrate [5], [17]. MgO (002) textured layers were sputtered at room temperature and then heated to 370°C, 410°C, or 475°C prior to deposition

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of $FePt - SiO_x$. The substrate heater was held at the desired temperature throughout the deposition of $FePt - SiO_x$ films. The film crystal structure was characterized by XRD with $CuK\alpha$ radiation from a Philips X'Pert Pro diffractometer.

The diffraction profiles were fitted to the pseudo-Voigt function, which is better at describing the peak shape [18], [19]. The peak height, the peak width, the Lorentz-fraction parameter, and the cubic function baseline are variables for deconvolution fitting.

III. Results and Disscusion

Homogenous ordering refers to the uniformity in ordering among the FePt grains. Fig. 1(b) displays the simulated XRD spectra of (002)-textured FePt films with different order parameter values. In homogenous ordering, we observed that the relationship $d_{001} = 2d_{002}$ (or $\sin\theta_{002} = 2\sin\theta_{001}$) holds true at any order parameter. On the one hand, the intensity of the fundamental (002) peak stays relatively constant with variation in S, whereas that of the superlattice (001) peak increases quadratically with S as plotted in Fig. 1(c). The small decrease of (002) peak intensity at high angles is due to a reduction of atomic scattering factors of the Fe and Pt and Lorentz polarization factors [15]. On the other hand, both (001) and (002) peaks move to higher angles because of the compression in c-axis [Fig. 1(a)] as FePt crystals become more ordered. Take note that the position of the (002) peak moves by a larger relative amount than that of the (001) peak.

If S varies from grain to grain, which we call inhomogeneous ordering, the intensity of the (001) peak has its contribution mostly from the FePt grains with high S, whereas the intensity of the (002) peak will come equally from all grains regardless of their S. Therefore, in inhomogeneous ordering, we expect a shift of the (002) peak to smaller 2θ values (Δ_{002}) relative to the value calculated from the (001) peak position, i.e. from $\sin\theta_{002} = 2\sin\theta_{001}$. The Δ_{002} values can be used to quantify the distribution of the ordering among grains. Δ_{002} here is normalized and written as

$$\Delta_{002} = \frac{2\theta_{002} - 2\theta_{002}^{measured}}{2\theta_{002} - 2\theta_{FCC200}}, \tag{3}$$

where $2\theta_{002}$ and $2\theta_{002}^{measured}$ are the (001)-derived and actual 2θ values of $L1_0$ (002), and $2\theta_{FCC200}$ is the 2θ value of FCC (200). Fig. 2 shows the simulated XRD patterns of media having the same average S of 0.58 but a different grain ordering distribution. A narrow ordering distribution with a standard deviation $\sigma_S = 0.16$ results in Δ_{002} of 0.22 [Fig. 2(a)]. On the other hand, a wider distribution with $\sigma_S = 0.33$ gives rise to a larger Δ_{002} value of 0.56 [Fig. 2(b)].

The shift of the (002) peak to smaller 2θ values is, however, commonly believed to be due to the presence of the FePt FCC (200) peak (disordered grains) and/or in-plane variants of the FePt $L1_0$ (200) [20], [21]. Bipolar ordering, in which some grains are FCC disordered within the homogenously ordered grains, can be considered as a special case of inhomogeneous ordering. Fig. 3(a) and 3(b) display the XRD scans and cross section transmission electron microscope (TEM) image of $FePt - 30\%SiO_x$ magnetic film deposited at 410°C. The

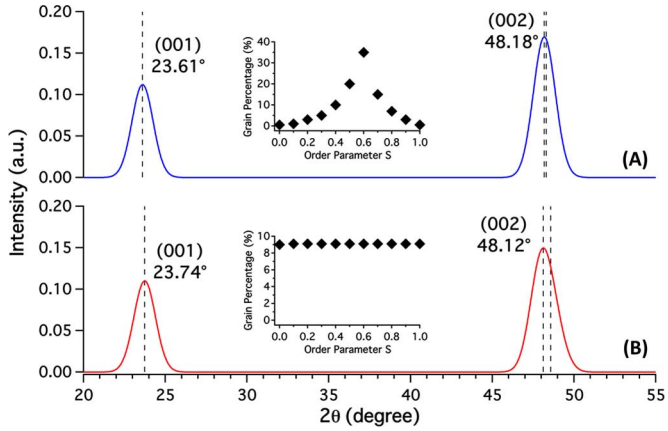

Fig. 2. Simulated XRD spectra of FePt film with (a) a narrow S distribution, and (b) a wide S distribution. Insets are the S distributions.

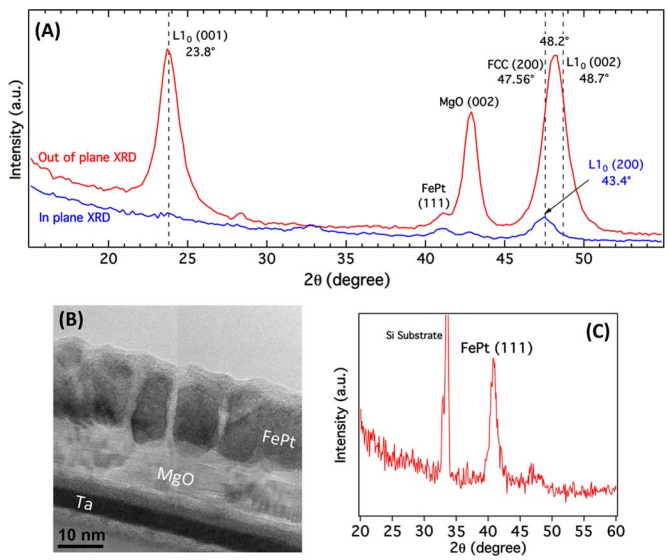

Fig. 3. (a) Out-of-plane and in-plane XRD scans of $FePt - 30\%SiO_x$ film. (b) Cross-section TEM image $FePt - 30\%SiO_x$ film. (c) XRD scan of the FePt film sputtered at room temperature.

columnar and well-isolated FePt grains are clearly seen. The film has the order parameter of 0.5, which was calculated from integral intensity ratio of FePt (001) and FePt (002) peaks with absorption and Lorentz correction factors included [15], [22]. In the out-of-plane pattern, FePt (002) has 2θ of 48.2°, which is in between FCC (200) and (001)-derived values. The 2θ of FCC (200) was obtained from the known (111) position of the FePt film sputtered at room temperature [Fig. 3(c)]. The 2θ of in-plane variants $L1_0$ (200), which is minimal in this sample as seen in the in-plane spectra, is difficult to determine. It can be approximated to a $L1_0$ (200) from the in-plane scan, which is 47.4°. The out-of-plane pattern was analyzed by deconvoluting the FePt (002) for three different cases. In analysis 1 [Fig. 4(a)], bipolar ordering, the (002) peak comprises FCC (200) at fixed position (47.56°) and $L1_0$ (002) calculated from $L1_0$ (001). It does not, however, show a good fit. In analysis 2 [Fig. 4(b)], addition of in-plane $L1_0$ (200) at 47.4° still does not give a good fit. In analysis 3 [Fig. 4(c)], by allowing the (002) peak to move to smaller 2θ values during deconvolution, we are able to achieve

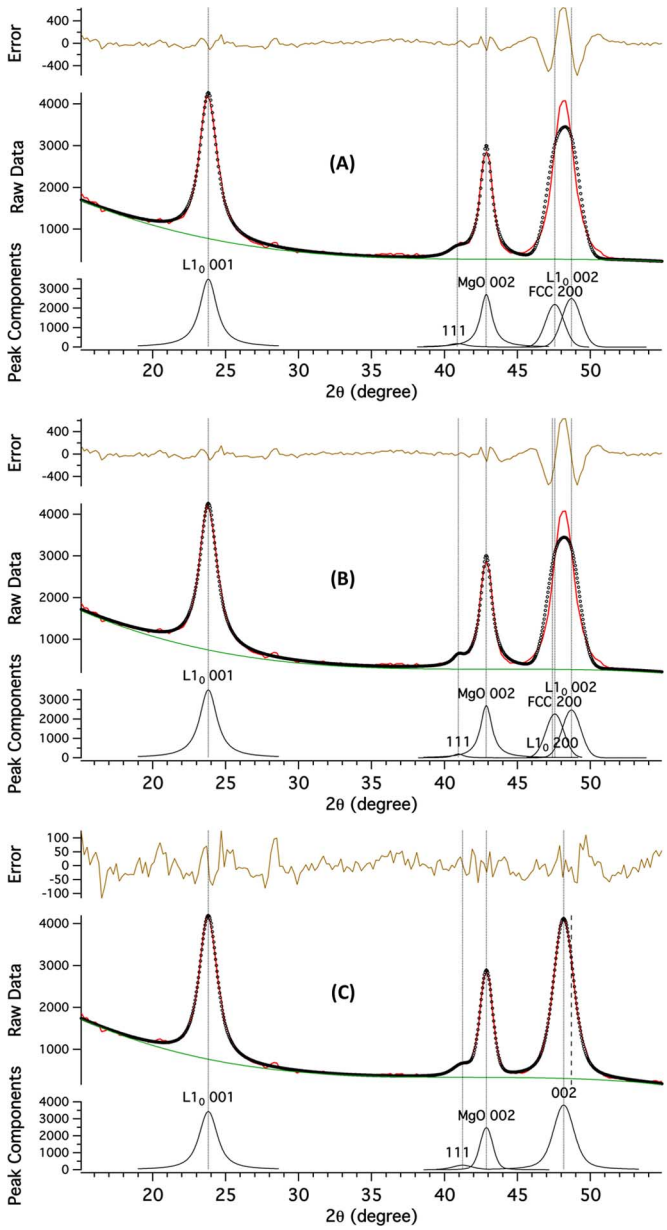

Fig. 4. XRD fitting results for (a) bipolar ordering, (b) bipolar ordering and in-plane variants, and (c) inhomogeneous ordering. The solid and circle curves are the measured and fitting spectra, respectively.

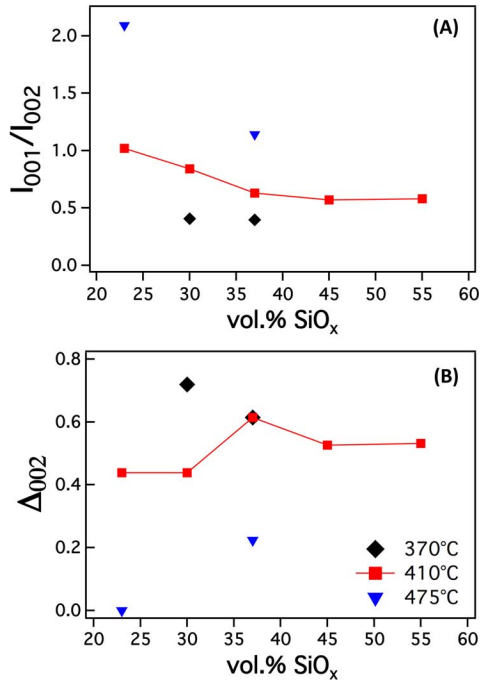

Fig. 5. (a) XRD integral intensity ratio I_{001}/I_{002} and (b) Δ_{002} of $FePt - SiO_x$ thin films with various oxide volume fractions at different temperature.

an excellent fit and hence demonstrate the existence of inhomogeneous ordering among FePt grains.

The A1 to $L1_0$ transition at equilibrium condition is of first-order [23], [24]. That means the transformation is discontinuous and proceeds via nucleation and growth of highly ordered $L1_0$ structure within disordered FCC matrix. This type of reaction has been observed experimentally in post-annealing of disordered FCC FePt films [25]. We expect the FePt grains in such magnetic films to exhibit the bipolar ordering character. When the system is, however, away from equilibrium with large supercoolings, the transition can occur homogenously and continuously [26], [27]. One example is the case of FePt films deposited with *in-situ* heating condition [28] which is how our samples were fabricated. Farrow *et al.* [29] speculated that the *in-situ* heating based deposition is controlled by surface diffusion which requires a much less activation energy than bulk diffusion in post-annealing counterpart. In continuous reaction, the films would start with small chemical order and progressively increase in time, thus, the grain-to-grain ordering variation is apparently inevitable. From the dependence of I_{001}/I_{002} ratio and Δ_{002} on the SiO_x segregant percentage at different heating temperature [Fig. 5], we can infer the trend that the samples with higher ordering have better uniformity of grain ordering. This experimental observation agrees with the theoretical analysis by Zhu *et al.* [8]. Nevertheless, the highly or completely ordered grains require high processing temperature, which negatively affects the media microstructure [17], [28], [30]. The partial order is probably more favorable and hence developing a proper fabrication method to control the ordering uniformity is essential.

Our present technique based on the left shift of the (002) peak is able to indicate the local ordering variation in the FePt granular media. However, the fluctuation from grain to grain in the film plane may not be the only mechanism of $L1_0$ ordering inhomogeneity. When the grain size is in the nano-scale range, the equilibrium properties at the grain boundary become important [31]. The variation in local composition of Fe and Pt can happen within the grains due to different surface segregation tendencies of the two elements [32]. In this case, the $L1_0$ ordering at the grain surfaces will be different from that inside the grains. In addition, the degree of ordering can be varied in the direction of film growth due to temperature fluctuation during sputter deposition. The temperature change can be significant especially if the heating and sputtering are done in separate chambers. Our future work will focus on improving this technique to address different types of local ordering variation in the film media.

IV. Conclusion

We have discussed the characteristics of XRD profiles for three grain ordering types that the $L1_0$ FePt film media may exhibit. If FePt grains are homogeneously ordered, there is a one-to-one relationship between (002) and (001) positions. If inhomogeneous ordering exists among the grains, (002) angle is smaller than the (001)-derived value. The angle shift can qualitatively describe the distribution of FePt grain ordering. Since we have shown that films deposited with *in-situ* heating are more prone to inhomogeneous grain ordering, further effort is needed for a more quantitative assessment. We believe that this technique will be of great aid in understanding the variation of atomic ordering among the grains of polycrystalline atomically ordered thin films.

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References

[1] D. Weller, A. Moser, L. Folks, M. E. Best, L. Wen, M. F. Toney, M. Schwickert, J. U. Thiele, and M. F. Doerner, "High K_u materials approach to 100 $\mathrm{Gbits/in^2}$," *IEEE Trans. Magn.*, vol. 36, pp. 10–15, 2000.

[2] J. S. Chen, B. C. Lim, J. F. Hu, B. Liu, G. M. Chow, and G. Ju, "Low temperature deposited $L1_0$ FePt-C (001) films with high coercivity and small grain size," *Appl. Phys. Lett.*, vol. 91, p. 132506, 2007.

[3] L. Zhang, Y. K. Takahashi, A. Perumal, and K. Hono, "$L1_0$-ordered high coercivity (FePt)Ag-C granular thin films for perpendicular recording," *J. Magn. Magn. Mater.*, vol. 322, pp. 2658–2664, 2010.

[4] C. P. Luo, S. H. Liou, and D. J. Sellmyer, "$FePt : SiO_2$ granular thin film for high density magnetic recording," *J. Appl. Phys.*, vol. 87, pp. 6941–6943, 2000.

[5] E. Yang and D. E. Laughlin, "$L1_0$ FePt-oxide columnar perpendicular media with high coercivity and small grain size," *J. Appl. Phys.*, vol. 104, p. 023904, 2008.

[6] S. Jeong, "Structure and magnetic properties of polycrytalline FePt and CoPt thin films for high density recording media," Ph.D. dissertation, Carnegie Mellon Univ., Pittsburgh, PA, 2002.

[7] G. Kai-Zhong and H. N. Bertram, "Transition jitter estimates in tilted and conventional perpendicular recording media at 1 $\mathrm{Tb/in^2}$," *IEEE Trans. Magn.*, vol. 39, pp. 704–709, 2003.

[8] J. G. Zhu, Y. G. Peng, and D. E. Laughlin, "Toward an understanding of grain-to-grain anisotropy field variation in thin film media," *IEEE Trans. Magn.*, vol. 41, pp. 543–548, 2005.

[9] K. Sato and Y. Hirotsu, "Long-range order parameter of oriented L10-FePt nanoparticles determined by electron diffraction," *Mater. Trans.*, vol. 44, pp. 1518–1522, 2003.

[10] T. Miyazaki, O. Kitakami, S. Okamoto, Y. Shimada, Z. Akase, Y. Murakami, D. Shindo, Y. K. Takahashi, and K. Hono, "Size effect on the ordering of $L1_0$ FePt nanoparticle," *Phys. Rev. B*, vol. 72, p. 144419, 2005.

[11] M. Tanase, J. G. Zhu, C. Liu, N. Shukla, T. J. Klemmer, D. Weller, and D. E. Laughlin, "Structure optimization of FePt nanoparticles of various sizes for magnetic data storage," *Metall. Mater. Trans. A*, vol. 38, pp. 798–810, 2007.

[12] M. Conley and D. Plalmer, Crystal Diffract for Windows. CrystalMaker Software Ltd., 1.1.2 ed., 2007.

[13] B. W. Roberts, "X-ray measurement of order in CuAu," *Acta Metall.*, vol. 2, pp. 597–603, 1954.

[14] J. A. Christodoulides, P. Farber, M. Dannl, H. Okumura, G. C. Hadjipanaysi, V. Skumryev, A. Simopoulos, and D. Weller, "Magnetic, structural and microstructural properties of FePt/M ($\mathrm{M} = \mathrm{C}$, BN) granular films," *IEEE Trans. Magn.*, vol. 37, pp. 1292–1294, 2001.

[15] B. E. Warren, *X-ray Diffraction*. Reading, MA: Addison-Wesley, 1969.

[16] PDF # 431359, JCPDS-International Centre for Diffraction Data vol. 2.02, 1999.

[17] E. Yang, H. Ho, D. E. Laughlin, and J.-G. Zhu, "Columnar grain growth of $\mathrm{FePt(L1_0)}$ thin films," *J. Appl. Phys.*, vol. 111, p. 07B720, 2012.

[18] R. A. Young and D. B. Wiles, "Profile shape functions in Rietveld refinements," *J. Appl. Crystall.*, vol. 15, p. 430, 1982.

[19] F. E. Spada, F. T. Parker, C. L. Platt, and J. K. Howard, "X-ray diffraction and Mossbauer studies of structural changes and $L1_0$ ordering kinetics during annealing of polycrystalline $\mathrm{Fe_{51}Pt_{49}}$ thin films," *J. Appl. Phys.*, vol. 94, pp. 5123–5134, 2003.

[20] J. S. Chen, Y. Xu, and J. P. Wang, "Effect of Pt buffer layer on structural and magnetic properties of FePt thin films," *J. Appl. Phys.*, vol. 93, pp. 1661–1665, 2003.

[21] J. S. Chen, B. C. Lim, Y. F. Ding, and G. M. Chow, "Low-temperature deposition of $L1_0$ FePt films for ultra-high density magnetic recording," *J. Magn. Magn. Mater.*, vol. 303, pp. 309–317, 2006.

[22] E. Yang, D. E. Laughlin, and J. G. Zhu, "Correction of order parameter calculations for FePt perpendicular thin films," *IEEE Trans. Magn.*, vol. 48, pp. 7–12, 2012.

[23] D. Fontaine, "Ordering instabilities and pretransitional effects," *Metall. Mater. Trans. A*, vol. 12, pp. 559–566, 1981.

[24] D. E. Laughlin, K. Srinivasan, M. Tanase, and L. Wang, "Crystallographic aspects of $L1_0$ magnetic materials," *Scripta Mater.*, vol. 53, pp. 383–388, 2005.

[25] R. A. Ristau, K. Barmak, L. H. Lewis, K. R. Coffey, and J. K. Howard, "On the relationship of high coercivity and $L1_0$ ordered phase in CoPt and FePt thin films," *J. Appl. Phys.*, vol. 86, pp. 4527–4533, 1999.

[26] A. Khachaturyan, T. Lindsey, and J. Morris, "Theoretical investigation of the precipitation of δ' in Al-Li," *Metall. Mater. Trans. A*, vol. 19, pp. 249–258, 1988.

[27] W. A. Soffa and D. E. Laughlin, "Decomposition and ordering processes involving thermodynamically first-order order $\rightarrow$ disorder transformations," *Acta Metall.*, vol. 37, pp. 3019–3028, 1989.

[28] M.-G. Kim and S.-C. Shin, "Temperature dependence of growth morphology of sputtered (FePt/Pt) films on MgO (100) substrate," *J. Appl. Phys.*, vol. 90, pp. 2211–2215, 2001.

[29] R. F. C. Farrow, D. Weller, R. F. Marks, M. F. Toney, S. Hom, G. R. Harp, and A. Cebollada, "Growth temperature dependence of long-range alloy order and magnetic properties of epitaxial $\mathrm{Fe_xPt_{1-x}}$ ($\mathrm{x \approx 0.5}$) films," *Appl. Phys. Lett.*, vol. 69, pp. 1166–1168, 1996.

[30] Y. Peng, J.-G. Zhu, and D. E. Laughlin, "$L1_0$ FePt-MgO perpendicular thin film deposited by alternating sputtering at elevated temperature," *J. Appl. Phys.*, vol. 99, p. 08F907, 2006.

[31] R. DeHoff, *Thermodynamics in Materials Science*, 2nd ed. Boca Raton: Taylor & Francis, 2006.

[32] M. Müller and K. Albe, "Lattice Monte Carlo simulation of FePt nanoparticles: Influence of size, composition, and surface segregation on order-disorder phenomena," *Phys. Rev. B*, vol. 72, p. 094203, 2005.