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Hoan Ho
Carnegie Mellon University

David E. Laughlin
Carnegie Mellon University, laughlin@cmu.edu

Jian-Gang Zhu
Carnegie Mellon University

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An X-Ray Diffraction-Based Method for Evaluating Inhomogeneous Ordering at the Grain Level of L1₀ – FePt Media

Hoan Ho, David E. Laughlin, and Jian-Gang Zhu

Data Storage Systems Center, Carnegie Mellon University, Pittsburgh, PA 15213

A technique based on x-ray diffraction (XRD) is introduced to understand the variation of the grain-to-grain L1₀ 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, L1₀ FePt, ordering variation, thin film media, x-ray diffraction.

I. INTRODUCTION

L1₀ 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 L1₀ superstructure, and varies as a quadratic function from 0 T for a disordered A1 phase to ~11.5 T for a fully ordered L1₀ phase [6]. Any variation in L1₀ 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 L1₀ film media.

II. EXPERIMENTAL METHODS

CrystalDiffract [12] was employed to generate simulated XRD patterns for L1₀ FePt crystals with various long-range order parameters, S. The lattice constants, namely a and c, and site occupancies of the Fe₅₀Pt₅₀ unit cell with respect to S are defined as follows [13]–[15]

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

$$S = P_{\alpha}^{Fe} - P_{\beta}^{Fe} = P_{\beta}^{Pt} - P_{\alpha}^{Pt}, \quad (2)$$

where a₀ and c₀ are the lattice constants of a fully ordered L1₀ FePt crystal (a₀ = 3.852 Å, c₀ = 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

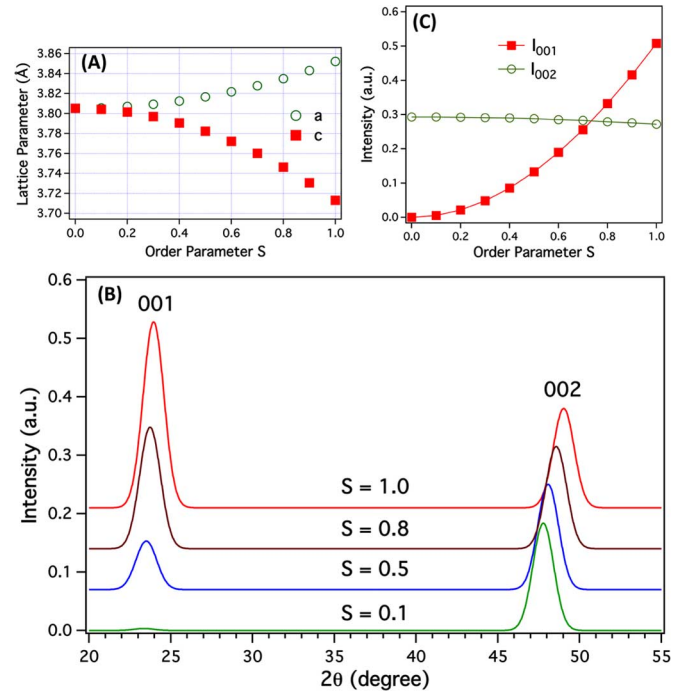


Fig. 1. (a) Lattice parameters, a and c, versus S. (b) Simulated XRD spectra of FePt film at various S. (c) Intensity of (001) and (002) peaks versus S.

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°.

Films were fabricated by RF sputtering using a Leybold Heraeus Z400 sputtering system at a base pressure < 5 × 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 α 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 DISCUSSION

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

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

where $2\theta_{002}$ and $2\theta_{002}^{measured}$ are the (001)-derived and actual 2θ values of L1₀ (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 L1₀ (200) [20], [21]. Bipolar ordering, in which some grains are FCC disordered within the homogeneously 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

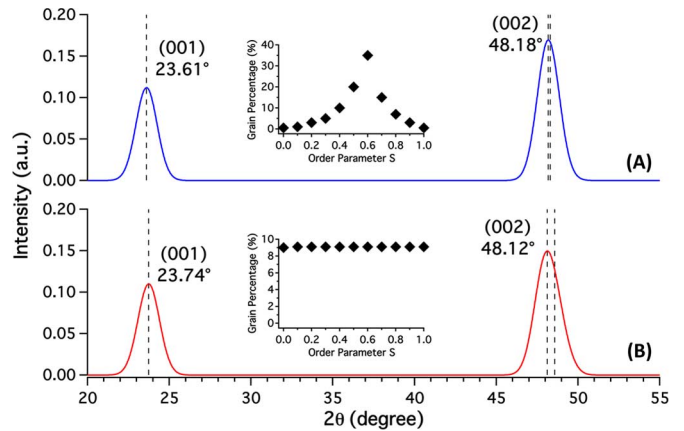


Fig. 2. Simulated XRD spectra of FePt film with (a) a narrow S distribution, and (b) a wide S distribution. Insets are the S distributions.

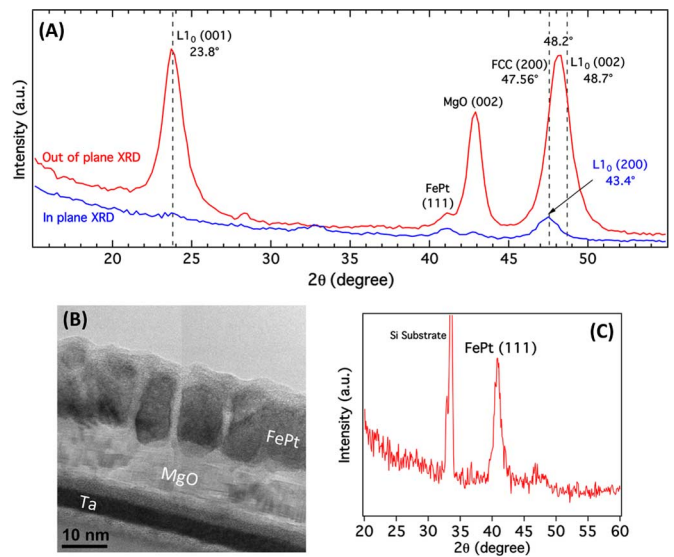


Fig. 3. (a) Out-of-plane and in-plane XRD scans of FePt – 30%SiO_x film. (b) Cross-section TEM image FePt – 30%SiO_x film. (c) XRD scan of the FePt film sputtered at room temperature.

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 L1₀ (200), which is minimal in this sample as seen in the in-plane spectra, is difficult to determine. It can be approximated to a L1₀ (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 L1₀ (002) calculated from L1₀ (001). It does not, however, show a good fit. In analysis 2 [Fig. 4(b)], addition of in-plane L1₀ (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

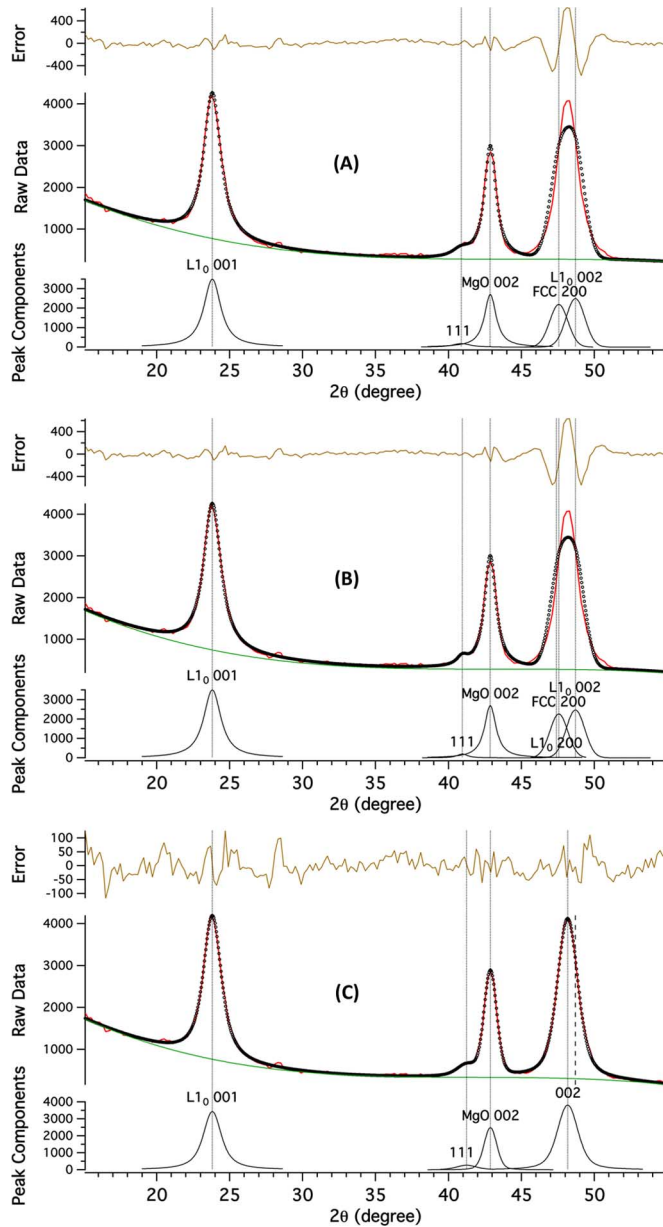


Fig. 4. XRD fitting results for (a) bipolar ordering, (b) bipolar ordering and in-plane variants, and (c) inhomogeneous ordering. The solid and circle curves are the measured and fitting spectra, respectively.

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

The A1 to L1₀ 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 L1₀ 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 homogeneously 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

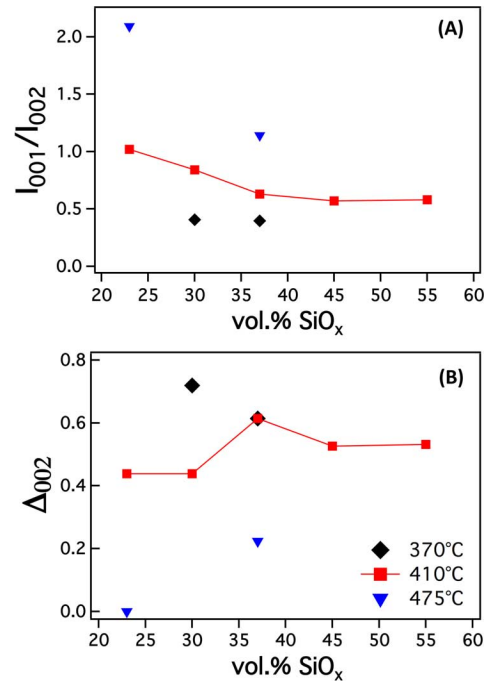


Fig. 5. (a) XRD integral intensity ratio I_{001}/I_{002} and (b) Δ_{002} of FePt – SiO_x thin films with various oxide volume fractions at different temperature.

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 L1₀ 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 L1₀ 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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