Correction of Order Parameter Calculations for FePt Perpendicular Thin Films

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The order parameter of FePt thin films plays an essential role in determining such diverse materials properties as magneto-crystalline anisotropy, magnetic coercivity and magnetic recording density. Typically, the order parameter for a bulk material is obtained by measuring the X-ray integrated intensity ratio of a super lattice peak to a fundamental peak and comparing this ratio to a theoretical value which has been calculated for a fully ordered sample. In this work, we present an analysis of the order parameter calculation in FePt $L1_0$ thin films taking into account the geometric features of the X-ray diffractometer, the crystallographic texture of FePt films, and the finite thickness of the films. The theoretical ratio of the (001) super lattice peak of FePt and the (002) fundamental peak of FePt is calculated as a function of the full width half maximum (FWHM) diffraction peaks from film and thickness for FePt thin films with perpendicular texture. A reliable order parameter calculation equation for Fe50Pt50 fiber textured perpendicular recording media is established.

Index Terms—**FePt, fiber texture, Lorentz factor, order parameter.**

I. INTRODUCTION

FePt ($L1_0$) is the most promising material to be used as media for achieving ultra-high recording density due to its large magneto-crystalline anisotropy energy (MAE) as well as its good environmental stability [1]–[8]. In order to achieve such type of recording media, obtaining FePt $L1_0$ thin films with high atomic order parameter is crucial [9]–[12]. The typical method of measuring an order parameter is to compare the ratio of a superlattice peak intensity to that of a fundamental peak intensity from measured X-ray diffraction peaks, and compare the ratio to a theoretically calculated ratio. This can be expressed as [13]–[18]

$$S = \left(\frac{I_s \times I_f^*}{I_f \times I_s^*}\right)^{\frac{1}{2}}. \tag{1}$$

Here, S is the atomic order parameter, whose value is between 0 (disordered) and 1 (fully ordered), the subscript S refers to the superlattice peaks, and the subscript f refers to the fundamental peaks. I_f^* and I_s^* are the theoretically calculated peak intensities for a fully ordered specimen, while I_f and I_s are the integrated peak intensities from the experimental results for a sample whose order parameter is to be measured. For this equation to hold, the superlattice planes must be parallel to the fundamental planes. In the case of FePt $L1_0$, the (001) superlattice and (002) fundamental reflections are chosen to calculate the order parameter of specimens. By measuring X-ray integrated intensity ratio of (001) and the (002) peak and comparing it to the theoretical value for a fully ordered sample, the order parameter of FePt samples can be obtained. This method is valid for thick films with no crystalline texture.

Calculations showed the relative peak intensities ratio of (001) and (002) peak for fully ordered polycrystalline Fe50Pt50 $L1_0$ films is ~2 [19]. This value has been widely used as the theoretical intensity ratio for fully order FePt in order parameter calculations for FePt thin films [20]–[22]. However, experimental results on FePt thin films with (001)/(002) intensity ratio much higher than 2 have been reported [23], [24]. Since the order parameter cannot be greater than 1, the theoretical value of the (001) to (002) intensity ratio needs to be reevaluated.

In this study, we analyze the effect of crystallographic texture and film thickness of FePt thin films on the theoretical (001) to (002) peak intensity ratio. The effect of texture is included by the full width half maximum FWHM of the diffraction peaks. Also, certain geometric features of the X-ray diffractometer need to be included in the calculation. The theoretical ratio of FePt(001) and FePt(002) as a function of texture and film thickness is obtained for fully ordered Fe50Pt50 thin films with perpendicular texture.

II. THEORY

The theoretical relative integrated intensity from an X-Ray diffraction experiment is usually expressed as [13]:

$$I^* = F^2 \times P \times A(\theta) \times e^{-2M} \times \frac{1+\cos^2 2\theta}{\sin 2\theta} \times \varphi \tag{2}$$

where F is the structure factor for the hkl planes, P is the multiplicity factor for the hkl planes, θ is the Bragg diffraction angle for the hkl planes, $A(\theta)$ is the absorption factor, e^{-2M} is the temperature factor where M equals to $(\sin\theta/\lambda)^2$. The expression $(1+\cos^2 2\theta)/2$ is the polarization factor due to unpolarized X-ray source. The expression $2\varphi/\sin 2\theta$ is the Lorentz factor, where $2/\sin 2\theta$ describes the peak height and peak broadening due to appreciable off-brag-angle diffraction, and φ is the powder ring distribution factor [25], which represents the geometrical fraction favorably oriented for diffraction and detection. For a randomly oriented polycrystalline sample

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TABLE I
RELATIVE INTENSITIES OF (001) AND (002) PEAK IN A RANDOMLY TEXTURED $\mathrm{L1_0}$ FePt WITH INFINITE THICKNESS

(h k l)	**(0 0 1)**	**(0 0 2)**
2θ	23.99	49.09
sin θ/λ	0.135	0.269
f_{Fe}	22.38	17.7
f_{Pt}	71.52	60.81
$\lvert F\rvert^2$	9659	24655
Temp. Factor	0.9643	0.8649
Absorption	1	1
Polarization	1.835	1.429
Lorentz	23.67	6.37
Multiplicity	2	2
Relative intensity	0.30	0.14

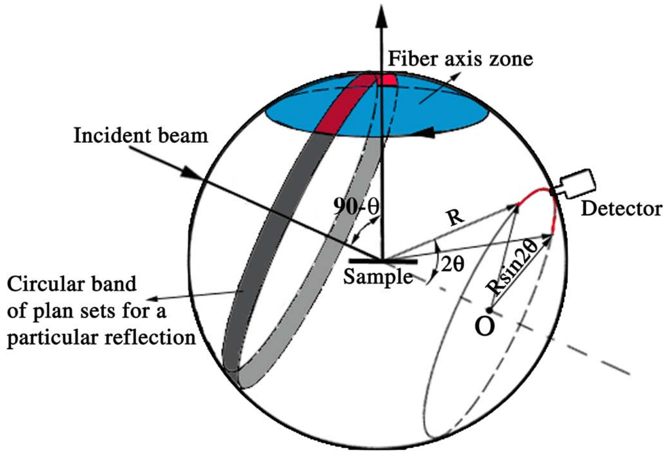

Fig. 1. The geometric relations of the distribution of plane normals, incident beam and reflected rays for fiber textured specimen. R is the goniometer radius.

(no texture), φ is proportional to $1/\sin\theta$. For a single crystal sample, the term is a constant.

For stoichiometric FePt with the $\mathrm{L1_0}$ structure, assuming the Fe atoms occupy the (0 0 0) and (1/2 1/2 0) sites, the Pt atoms occupy the (1/2 0 1/2) and (0 1/2 1/2) sites. The structure factor F for reflection can be calculated as

$$F = f_{\mathrm{Fe}}\left(1 + e^{i\pi(k+h)}\right) + f_{\mathrm{Pt}}\left(e^{i\pi(k+l)} + e^{i\pi(l+h)}\right) \quad (3)$$

where f_{Fe} and f_{Pt} are the atomic scattering factors of Fe and Pt. For non-stoichiometric FePt or FePt with ternary additions, the change of composition has to be considered in the calculation of structure factor and therefore the calculation of order parameter.

The relative peak intensities for (001) and (002) reflections in the fully ordered randomly textured polycrystalline $\mathrm{L1_0}$ Fe50Pt50 with infinite thickness are tabulated in Table I, which yields the relative intensity ratio of (001) to (002) peak of ~2.14.

A. *Correction for Lorentz Factor*

As mentioned before, the specimen considered in Table I was assumed to have a completely random texture of its grains with a uniformly distributed orientation of all its planes. However, in order to develop FePt as a perpendicular recording media, the thin film is engineered to have the c axes of its grains in a [001] fiber texture, that is, to have its (001) planes parallel to the plane of the film [26]–[28]. Thus, neither one of those two powder ring distribution factors (φ) are appropriate for a fiber textured polycrystalline sample. We must determine the form of the Lorentz factor for a sample with fiber texture. In Fig. 1, we consider for a particular reflection the crystals that are oriented at or near a particular Bragg angle. Diffraction will occur only from the crystals with their plane normals lying in the circular band shown in the figure. However, for a fiber textured specimen, the plane normals all lie in the dome shaped region (shown as a blue dome). Therefore, only the planes whose normals lie in the intersection of the circular band and the dome (shown as a red stripe in the figure) will diffract. The location of the diffracted beams are displayed as a red arc near the detector. The fraction of grains that are favorably oriented for a particular diffraction will be given by the ratio of the area of the red stripe to that of the dome. For $\theta < 90 - \mathrm{FWHM}/2$, this ratio is calculated as

$$\frac{Area(red\_stripe)}{Area(blue\_dome)} = \frac{\cos\theta}{\sin^2\left(\frac{\mathrm{FWHM}}{2}\right)} \times \arccos\left(1 - \frac{2\sin^2\left(\frac{\mathrm{FWHM}}{2}\right)}{\cos^2\theta}\right).$$

Calculation indicates that the change of θ and FWHM does not influence this ratio much. For FePt, $(001)(\theta = 11.99)$ to $(002)(\theta = 24.54)$ ratio of this part is 0.999 for $\mathrm{FWHM} = 1^\circ$, and 0.995 for $\mathrm{FWHM} = 45^\circ$. Therefore, it is safe to say that for diffraction from (001) and (002) of $\mathrm{L1_0}$ FePt films with good perpendicular texture (no in-plane variants), this ratio is θ independent. However, for cases with c axis variants in the sample [29], the plane normals are unevenly distributed over the whole sphere, this ratio is proportional to $\cos\theta$ since the ratio of the area of the circular band to that of the whole sphere is proportional to $\cos\theta$ [13].

Another geometrical factor that must be included is the fraction of the diffracted beams that can be detected by the detector. As shown in Fig. 1, for polycrystalline samples with randomly distributed plane normals, the detector size is very small compared to the length of the diffraction line. This value is inversely proportional to the length of the diffraction line, which is proportional to $1/\sin 2\theta$.

Fig. 2 shows the geometry necessary to determine the fraction of the diffracted rays that can be detected by the detector for fiber textured samples. With a fiber texture, there are more diffracted rays near the detector region. The angle of the diffracted arc intercepted by the detector is given by $\alpha = 2\arcsin(D/2R\sin 2\theta)(D \ll R)$, where D is the chord length of the intercepted diffraction arc, and R is the radius of the goniometer of the X-ray diffractometer.

The fiber axis distribution can be measured by the X-ray rocking curve, which is a direct measure of the distribution of plane normal orientations present at the irradiated area of the sample. The distribution function can be determined by curve fitting of the rocking curve as shown in Fig. 3.

Thus, the fraction of the diffracted beam intercepted by the detector is the integral of distribution function evaluated be-

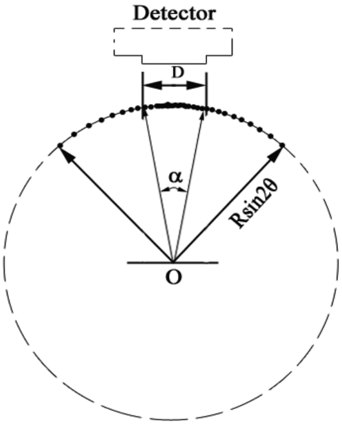

Fig. 2. Interception of arc of diffracted rays (Detection part of Fig. 1), R is the goniometer radius.

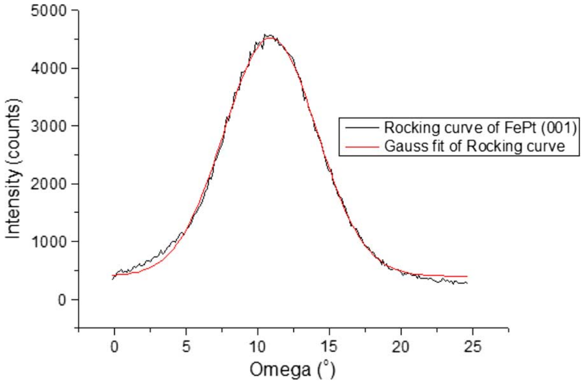

Fig. 3. Gaussian fit for a typical FePt(001) rocking curve with perpendicular texture. The fitting equation is $y = y_0 + (1/w) \times e^{-2(x/w)^2}$, where $w = (\mathrm{FWHM}/\sqrt{2\ln 2})$.

tween the limits 0 and α. For fiber textured samples with no in-plane variants, the fraction intercepted by the detector is

$$\propto \int_0^{2\arcsin\frac{D}{2R\sin 2\theta}} \frac{1}{w}\times\exp\left(-2\left(\frac{x}{w}\right)^2\right)dx,\ \text{where } w=\frac{\mathrm{FWHM}}{\sqrt{2\ln 2}}$$

where $\propto\ erf(2\sqrt{\ln 2}(\arcsin(D/R\sin 2\theta)/\mathrm{FWHM}))$. Therefore, the powder ring distribution factor φ for fiber textured samples with no in-plane variant is $\propto\ erf(2\sqrt{\ln 2}$ $(\arcsin(D/R\sin 2\theta)/\mathrm{FWHM}))$, and the Lorentz factor is

$$\begin{aligned}&\propto 1/\sin 2\theta \times \varphi\\ &\propto 1/\sin 2\theta \times erf\left(2\sqrt{\ln 2}\frac{\arcsin\frac{D}{R\sin 2\theta}}{\mathrm{FWHM}}\right).\end{aligned} \tag{4}$$

For fiber textured samples with in-plane variants, the Lorentz factor is proportional to

$$\begin{aligned}&\propto 1/\sin 2\theta \times \varphi\\ &\propto 1/\sin 2\theta \times \cos\theta \times erf\left(2\sqrt{\ln 2}\frac{\arcsin\frac{D}{R\sin 2\theta}}{\mathrm{FWHM}}\right).\end{aligned} \tag{5}$$

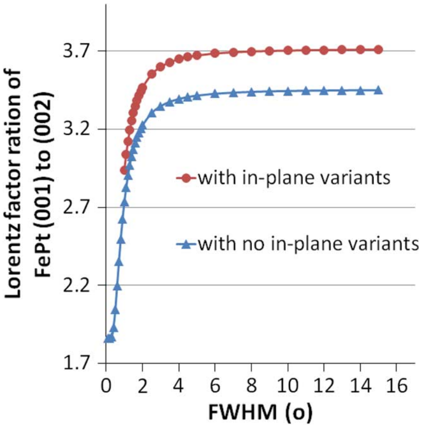

Fig. 4. Lorentz factor ratio of $\mathrm{L1_0}$ FePt (001) to (002) peak as a function of FWHM for fiber textured specimen.

As (4) and (5) shows, the Lorentz factor for fiber textured samples is a function of the FWHM as well as the geometric features of the X-ray diffractometer. This equation can be used for calculating the relative Lorentz factor of any fiber textured polycrystalline material including powder, thin films, and bulk materials.

It is important to point out that the chord length of the intercepted diffraction arc (D value) varies greatly with the geometry of the X-ray diffractometer. For example, in the Debye-Scherrer method, D is related to the Debye-Scherrer film width. For X-ray diffractometers, it is usually related to the angular divergence of the diffraction ray capturing device. For textured samples, the most commonly used Diffractometer is the X'pert Pro Diffractometer, X-Ray lens with parallel beam geometry. It has a parallel plate collimator placed before the optional soller slits and the detector. The parallel plate collimator has a divergence of 0.27°. The goniometer radius, R, is 320 mm. D in this case equals

$$D = R \times \tan(\mathrm{divergence}).$$

Insert the D and R values into (4), the Lorentz factor ratio of FePt(001) to FePt(002) as a function of FWHM can be plotted as shown in Fig. 4.

The Lorentz factor ratio of (001) to (002) for single crystal $\mathrm{L1_0}$ FePt is 1.86 and, for randomly oriented polycrystal FePt, it is 3.86. Even though the slope of these plots changes greatly with different "D" values, the perpendicular textured FePt always has the Lorentz ratio lie in between these two values. Furthermore, specimens with larger FWHM values always show higher Lorentz factor ratios of (001) to (002).

B. *Correction for Absorption Factor*

So far, we have considered thick specimens with preferred orientation. When the specimen is sufficiently thick, the effective volume irradiated by an incident beam is constant and independent of θ because of the balancing of two opposing effects, the irradiated area of cross section and penetration depth [13]. Therefore, it has been concluded that the absorption decreases the intensities of all diffracted beams by the same amount and

TABLE II

Relative Intensities of (001) and (002) Peak in a Fiber Textured L1_0 FePt With No In-Plane Variants, Where t (nm) is the Sample Thickness, D is the Chord Length of the Intercepted Diffraction Arc, and R is the Goniometer Radius. When Using These Equations, D and R Should Have the Same Unit; so do FWHM and arcsin

(h k l)	(0 0 1)	(0 0 2)
Absorption	$1-e^{-0.0032t}$	$1-e^{-0.0016t}$
Lorentz	$4.92\times erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{2.46D}{R}}{FWHM}\right)$	$2.65\times erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{1.32D}{R}}{FWHM}\right)$
Relative intensity	$0.062\left(1-e^{-0.0032t}\right)erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{2.46D}{R}}{FWHM}\right)$	$0.058\left(1-e^{-0.0016t}\right)erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{1.32D}{R}}{FWHM}\right)$

therefore does not enter into the calculation of the relative intensities, provided the specimen is effectively of infinite thickness.

However, from the definition of "infinite thickness" [13], [30], the specimen has to be at least 100 to 200 μm, which is a few orders of magnitude thicker than the thin films in modern magnetic recording media. Most of the thin films in current devices are less than 60 nm. Thus, the thickness has to be considered in the absorption factor calculation for our thin films.

The absorption factor can be calculated as

$$A = \frac{I_D}{I_0} \tag{6}$$

where I_D is diffracted beam outside the specimen, which is the integrated intensity and the I_0 is the incident beam. The absorption factor calculates the fraction of incident beam being effectively diffracted, not the fraction being absorbed. The diffracted beam intensity is proportional to the effective irradiated volume; it is also decreased by absorption by a factor of e^{-ul}, where l is the incident beam travel distance and u is the average mass absorption coefficient. Therefore, I_D can be calculated as

$$I_D = I_0 ab \int e^{-ul} dv \tag{7}$$

where a is the volume fraction of specimen having the correct orientation for the reflection of the incident beam, and b is the fraction of the incident energy being effectively diffracted [13]. For the particular specimen arrangement used in a diffractometer, the incident and diffracted beams have an equal angle θ. Therefore, I_D equals [31]:

$$I_D = \frac{I_0 ab}{\sin\theta} \int_{x=0}^{x=t} e^{-2ux/\sin\theta} dx, \quad I_D = \frac{I_0 ab}{\sin\theta}(1 - e^{-2ut/\sin\theta}).$$

Therefore, $A(\theta) = (I_D/I_0) = (ab/2u)(1 - e^{-2ut/\sin\theta})$,

$$A(\theta) \propto 1 - e^{-2ut/\sin\theta}. \tag{8}$$

As the equation shows, the absorption factor for thin films is θ dependent, and is a function of film thickness. This equation can be used for calculating relative absorption factor of any material.

C. Relative Intensity

We are now in a position to consider the relative intensity ratio of (001) to (002) peaks as a function of both the thickness and film texture. The corrected relative peak intensities for (001) and (002) reflections in the fully ordered fiber textured polycrystalline L1_0 FePt are tabulated in Table II. This shows that the ratio of the intensities of the 001 and 002 peaks depends on FWHM, D, R, and t. Thus, unless these parameters are accounted for, the calculation of the order parameter is not complete.

For fully ordered fiber texture FePt with in-plane variants, the Lorentz factor and relative intensity are slightly different, as shown in Table III.

Fig. 5 shows the theoretical intensity ratios of fully ordered and fiber textured FePt (001) peak to (002) peak as a function of film thickness and FWHM for X'pert Lens with parallel beam geometry. As is shown in the figure, the thickness of a thin film changes the theoretical intensity ratio dramatically; thinner films will exhibit a much higher intensity ratio of (001)/(002) than the thicker films. While with the same thickness, highly textured films show lower intensity ratios. For instance, a 10 nm thick fully ordered FePt film with 8° of FWHM will have an intensity ratio of (001)/(002) as high as 3.93, which explains the high ratio we obtained in previous experimental results [23]. On the other hand, a 100 nm thick fully ordered FePt film with 1° of FWHM, the intensity ratio of (001)/(002) is only 2.9. Note that a fiber textured polycrystal sample with 0.1° of FWHM has the same intensity ratio of (001) to (002) peak as a single crystal; while for a "fiber" textured sample with $\mathrm{FWHM} = 180°$, because of the uneven distribution of plane normals, the intensity ratio of (001) to (002) is 4.26 compared to 4.27 for a completely randomly oriented polycrystal sample.

D. Order Parameter

From the above discussion and calculations, we can see that for the same ratio of 001 to 002 intensities, (measured in the same diffractometer) samples may have drastically different

TABLE III
RELATIVE INTENSITIES OF (001) AND (002) PEAK IN A FIBER TEXTURED $L1_0$ FePt WITH IN-PLANE VARIANTS, WHERE t (NM) IS THE SAMPLE THICKNESS, D IS THE CHORD LENGTH OF THE INTERCEPTED DIFFRACTION ARC, AND R IS THE GONIOMETER RADIUS

(h k l)	**(0 0 1)**	**(0 0 2)**
Absorption	$1-e^{-0.0032t}$	$1-e^{-0.0016t}$
Lorentz	$4.81\times erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{2.46D}{R}}{FWHM}\right)$	$2.41\times erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{1.32D}{R}}{FWHM}\right)$
Relative intensity	$0.061\left(1-e^{-0.0032t}\right)erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{2.46D}{R}}{FWHM}\right)$	$0.053\left(1-e^{-0.0016t}\right)erf\left(2\sqrt{\ln 2}\,\dfrac{\arcsin\dfrac{1.32D}{R}}{FWHM}\right)$

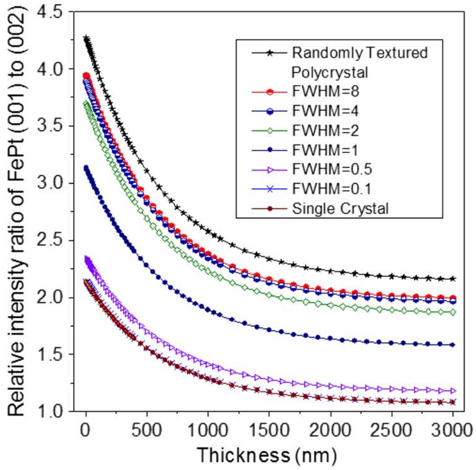

Fig. 5. The theoretical intensity ratio of fully ordered fiber textured (no in-plane variants) FePt (001) peak to (002) peak as a function of film thickness and FWHM.

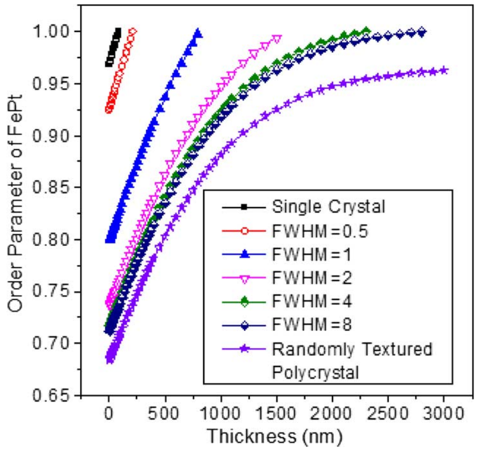

Fig. 6. With the measured FePt(001) to (002) peak intensity ratio as 2, the true order parameter of fiber textured FePt films as a function of film thickness and texture.

order parameters depending on the geometry of the X-ray diffractometer, values of FWHM, and thickness. Fig. 6 plots the actual order parameters of FePt $L1_0$ fiber textured samples with the same experimental (001) to (002) intensity ratio of 2 but different thicknesses and FWHM values. As is shown in the figure, with the same experimental (001) to (002) intensity ratio 2, for a 10 nm thick sample with FWHM value equals to 8 degrees, the order parameter is only 0.7; while if it is a single crystal, it has an order parameter of 0.97.

III. CONCLUSION

The theoretical XRD peak intensity ratio of FePt(001) to FePt(002) was calculated for FePt thin films with perpendicular texture. With geometry of the X-ray diffractometer, film texture, and film thickness taking into account, the Lorentz factors and absorption factors were recalculated. It was found that the Lorentz factor can be described by an equation that depends on Bragg angle, goniometry of the X-ray equipment, and the degree of preferred orientation. The powder ring distribution factor φ for fiber textured samples is recalculated as shown in (4) and (5). The absorption factor is a function of sample thickness when the sample is less than a few hundred micrometers thick. More reliable order parameters of FePt films were obtained by using this new theoretical intensity ratio.

The equations for calculating Lorentz factor and absorption factor not only applies to FePt, but also to any fiber textured polycrystalline materials including powder, thin films, and bulk materials.

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