Determination of the Direction of the c-Axis of $L1_0$ FePt Thin Films with the Mössbauer Spectroscopy

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The continued growth of storage capacity requires new innovations in recording media and in particular, in magnetic nanostructures. FePt thin films in the $L1_0$-phase are interesting candidates for high-density magnetic recording media due to their large magnetocrystalline anisotropy. In the present work, we investigated the magnetic and structural properties of FePt thin films directly grown on MgO(110) with molecular beam epitaxy. The purpose was to gain insight in the correlation between the magnetization process and the morphology of the FePt thin films. We introduce conversion electron Mössbauer spectroscopy to derive the direction of the easy magnetization axis with respect to the substrate. The results are compared to the characterization performed with high angle X-ray diffraction.

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1. Introduction

The crystal growth of metallic thin films is strongly influenced by the substrate. Not only the crystal structure can be manipulated by an appropriate choice of the substrate, but also the epitaxial relations with the substrate. The most common used technique to define the crystal orientation is high angle X-ray diffraction (XRD). However, X-ray diffraction is no longer accessible when dealing with thin films of a few atomic layers. In this paper we describe an alternative approach to determine the crystal orientation based on the Mössbauer spectroscopy. With the Mössbauer spectroscopy thin films on the subnanometer scale can be characterized. As an example we studied the crystal properties of an FePt thin film.

The growth of FePt thin films on MgO(100) results in the AuCu(I) crystal structure which has an extremely high magnetocrystalline anisotropy [1, 2]. In view of technological applications for magnetic recording media, the need exists to

control the direction of the easy magnetization axis. The easy magnetization axis of FePt on MgO(100) points out of the film plane along the c-axis of the tetragonal $L1_0$ structure [3]. The direction of the c-axis can be manipulated during the crystal growth by choosing a proper substrate. It has been reported that FePt deposited on a Pt or Au seed layer on MgO(110) also grows in the $L1_0$ structure but has its c-axis oriented in the film plane [4–8] while the c-axis is reported to be canted out of the film plane when deposited directly on MgO(110) [9].

We will demonstrate the advantages of the Mössbauer spectroscopy for deriving the crystal orientation of a FePt thin film directly grown on MgO(110). Via the Mössbauer spectroscpy we will show how to investigate domains, distributions and magnetism in thin films. In addition, the exact canting angle of the easy magnetization axis will be derived. High angle X-ray diffraction is used to verify the conclusions based on the Mössbauer measurements.

2. Structural characterization

An epitaxial ^{57}FePt(30 nm) thin film was grown with molecular beam epitaxy on a polished MgO(110) substrate held at 500°C. The pressure during the growth was below 1×10^{-10} Torr. The thickness, the deposition rate, and the relative atomic flux was controlled with calibrated quartz crystals. The deposition rates for ^{57}Fe and Pt are 0.041 Å/s and 0.053 Å/s, respectively. The sample was characterized with conversion electron Mössbauer spectroscopy and high angle X-ray diffraction.

2.1. Conversion electron Mössbauer spectroscopy

Conversion electron Mössbauer spectroscopy (CEMS) has, due to its high sensitivity to the atomic and electronic surroundings of the Mössbauer isotope ^{57}Fe, successfully been utilized in the study of ultrathin films [10, 11]. Here, we use the technique to investigate the direction of the c-axis of FePt $L1_0$ to gain more insight in the crystal growth of FePt $L1_0$ on MgO(110). For FePt the magnetic moments, and thus the magnetic hyperfine field $\boldsymbol{B}_{\mathrm{hf}}$, are parallel aligned with the c-axis of the tetragonal $L1_0$ crystal structure [11, 3]. The angle θ between the incident gamma-radiation and $\boldsymbol{B}_{\mathrm{hf}}$ influences the relative intensities of the lines of a CEMS spectrum. The ratios of the peak intensities are given by

$$3 : \frac{4\sin^2(\theta)}{1+\cos^2(\theta)} : 1 : 1 : \frac{4\sin^2(\theta)}{1+\cos^2(\theta)} : 3. \tag{1}$$

When a distribution on the hyperfine fields is present, the Mössbauer peaks at higher velocities become broader and the peak amplitudes become smaller compared to the inner Mössbauer lines. Nevertheless, the ratios between the spectral areas are remained such that Eq. (1) is also valid for a non-zero hyperfine field distribution. The angle θ in Eq. (1) is incorporated as a fit parameter of the analysis model.

CEMS measurements were performed at room temperature using a 50 mCi ^{57}Co in a Rh matrix source. The thin film was incorporated as an electrode of a
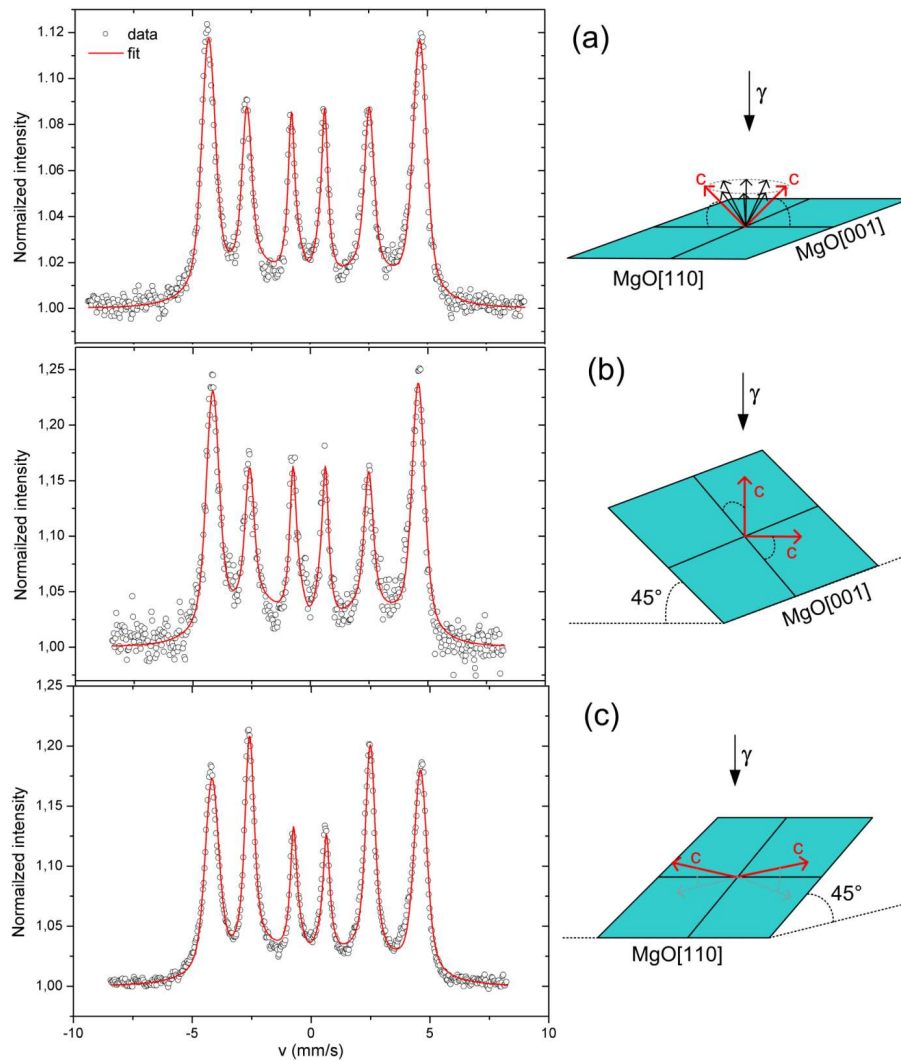
Fig. 1. Conversion electron Mössbauer spectra taken (a) with the source perpendicular to the sample film, (b) the MgO[110] direction canted 45° and (c) the MgO[001] direction canted 45° and an illustration of the different setups respectively.

proportional gas detector. The spectra in Fig. 1 could be fitted consistently with a Voight-based distribution model [12] of which the mean hyperfine field of 27.2 T matches well with the literature [11]. A Gaussian distribution width σ of 1.2 T is found. Spectrum 1a was obtained with the gamma radiation of the ^{57}Co source perpendicular to the sample plane as illustrated in the right column of Fig. 1a. From the analysis an angle $\theta = 44(1)°$ was extracted indicating a 44(1)° canted c-axis out of the film plane. However, due to the perpendicular configuration,

with CEMS no lateral preferential direction can be derived. Still, it is possible to search for the direction of the *c*-axis by mounting the Mössbauer source at an angle $> 0°$ with respect to the sample normal. If the *c*-axis is canted towards MgO [110], the CEMS spectrum obtained with the source positioned as illustrated in Fig. 1b would show two sites, namely one site with the magnetization perpendicular oriented to the incident gamma radiation and one site with the magnetization parallel oriented. Hence, the average angle between the gamma radiation of the ^{57}Co source and $\boldsymbol{B}_{\mathrm{hf}}$ is 45°. This means that the CEMS spectrum can also be analyzed with a single site of which $\boldsymbol{B}_{\mathrm{hf}}$ is positioned under 45° with respect to gamma radiation. On the other hand, if the *c*-axis is canted towards MgO[100], the analysis of the CEMS spectrum obtained with the photons positioned as in illustration 1c would give an angle $\theta = 45°$.

To reveal whether the *c*-axis is canted towards MgO[110] or MgO[100], the proposed asymmetric CEMS spectra are obtained. An angle of $43(2)°$ is calculated from the relative peak intensities of spectrum 1b which indicates that the *c*-axis is canted 45° towards MgO[110]. This interpretation is confirmed by the analysis of the third spectrum. The extracted angle of $61.0(4)°$ derived from the analysis of spectrum 1c matches well with the theoretical angle of 60° between the *c*-axis of the L1$_0$ structure and the incident gamma radiation of the ^{57}Co source.

In summary, from the CEMS spectra the direction and the canting angle of the *c*-axis could be derived, namely the *c*-axis is canted 45° in both direction from the sample normal towards the MgO[110] direction.

2.2. High angle X-ray diffraction

The results obtained with the Mössbauer spectroscopy are verified with a classical phase determination technique, namely high angle X-ray diffraction. XRD scans of the FePt thin film on MgO(110) are taken with an X-ray diffractometer combined with a double bound Ge(022) monochromator and a Cu X-ray source (K_{α}(Cu) $= 1.5406$ Å). The XRD-scan in Fig. 2a shows an epitaxial peak around $2\theta = 69°$. From this scan only, it is difficult to assign the diffraction peak to the FePt(220) or the FePt(022) diffraction as the difference between the *a*- and the *c*-lattice parameter is very small ($c/a = 0.98$). Performing an *a*-symmetric XRD--scan searching for the FePt(00*n*) diffractions will reveal the correct orientation. The *c*-axis was experimentally found after performing an *a*-symmetric XRD-scan with the film plane canted 45° such that the optics plane is parallel to the MgO[001] direction. In other words, the $\theta{-}2\theta$-scan was performed along the FePt(004) or the FePt(400) direction. A second scan was performed after rotating the sample 180° around the sample normal. The XRD-scans in Fig. 2b show both the FePt(004) diffraction peak as the FePt(400) diffraction peak indicating that the epitaxial FePt thin film is symmetrically twinned. From the XRD characterizations we can conclude that the *c*-axis is canted 45° from the MgO[110] direction to the sample normal. We obtained the same conclusion from the CEMS analysis.
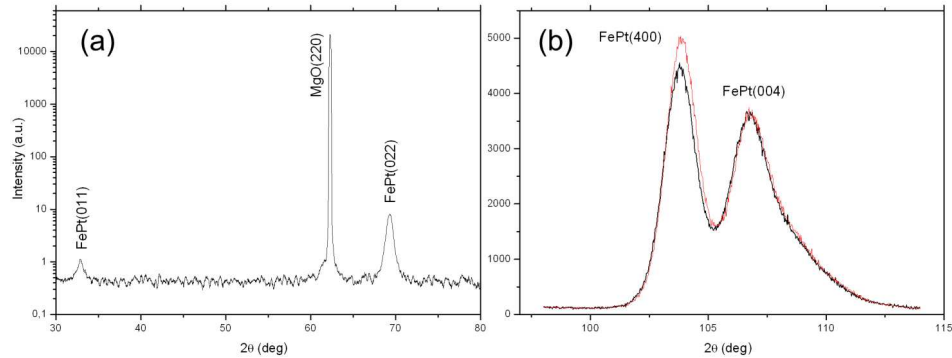
Fig. 2. (a) Symmetric high angle X-ray diffraction scan of FePt(30 nm) L1$_0$ on MgO(110) and (b) an *a*-symmetric XRD scan showing the FePt(400) and FePt(004) diffraction in black. The scan in gray is taken after rotating the sample 180° around the sample normal.

From the FePt(400) and the FePt(004) diffraction peaks the *a*- and the *c*-lattice parameter can be derived respectively: $a_{\mathrm{FePt}} = 3.914$ Å and $c_{\mathrm{FePt}} = 3.838$ Å. This leads to an in-plane lattice misfit with MgO ($a_{\mathrm{MgO}} = 4.200$ Å) of $\Delta_{[110]} = -7.7\%$ along MgO[110] and of $\Delta_{[001]} = -6.8\%$ along MgO[001]. Supposing FePt grows with its *c*-axis oriented in the film plane on MgO(110), the misfits would be given by $\Delta_{[110]} = -8.1\%$ and $\Delta_{[001]} = -6.8\%$. These larger misfit values explain the experimentally found crystal orientation of FePt thin films directly grown on MgO(110).

3. Conclusions

In conclusion, we were able to derive the easy magnetization axis of L1$_0$ FePt on MgO(110) with Mössbauer spectroscopy. An unique direction of the *c*-axis with respect to the substrate was obtained. The results are confirmed by symmetric and *a*-symmetric high angle X-ray diffractions scans and can be summarized with the following epitaxial relations:

FePt[100] ∥ MgO[001,]

FePt[0$\bar{1}$1] ∥ MgO[011],

FePt[001], MgO[001] = 45°.

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