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Modifying magnetic properties of ultra-thin magnetite films by growth on Fe pre-covered MgO(001)

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Iron oxide films were reactively grown on iron buffer films, which were deposited before on MgO(001) substrates to analyze the influence of the initial iron buffer layers on the magnetic properties of the magnetite films. X-ray photoelectron spectroscopy and low energy electron diffraction showed that magnetite films of high crystalline quality in the surface near region were formed by this two-step deposition procedure. The underlying iron film, however, was completely oxidized as proved by x-ray reflectometry and diffraction. The structural bulk quality of the iron oxide film is poor compared to magnetite films directly grown on MgO(001). Although the iron film was completely oxidized, we found drastically modified magnetic properties for these films using the magnetooptic Kerr effect. The magnetite films had strongly increased coercive fields, and their magnetic in-plane anisotropy is in-plane rotated by 45° compared to magnetite films formed directly by one step reactive growth on MgO(001). © 2015 AIP Publishing LLC.

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I. INTRODUCTION

The modification of the magnetic properties of thin ferromagnetic films is interesting for spintronic devices including magnetic tunnel junctions (MTJs).1 MTJs are basically stacks of two conducting ferromagnetic films, which are separated by an insulating non-magnetic film, the tunnel barrier. The tunnel probability through this barrier depends strongly on the alignment of the magnetization in the magnetic films. If the magnetization in these two films is parallel to each other, there is a high tunnel current for positive tunnel magnetoresistance (TMR),2 while there is a low tunneling current if the magnetization is antiparallel to each other. For negative TMR, it is vice versa.3 A high coercive field of the magnetic material promotes the stability of the alignment of the magnetization in these magnetic films. The coercive field, however, depends on the magnetic anisotropy of the film, which is determined by the crystal structure and film thickness. The strength of the TMR-effect is influenced by the magnetic field/tunnel barrier interface roughness.4,5 The higher the interface roughness, the weaker the TMR-effect.

Magnetite is a promising material for spintronic applications since it is half-metallic6 and has a high spin polarization at the Fermi level.7 Moreover, the Curie temperature of this ferrimagnetic oxide is quite high (858 K),8 and it has a magnetic saturation moment of 4 μB.9 MgO(001) is an ideal substrate to grow magnetite films with high crystalline quality. The lattice mismatch between the inverse spinel structure of magnetite (aFe₃O₄ = 0.8396 nm) is only 0.3% concerning the doubled MgO lattice constant (aMgO = 0.4212 nm, rock salt structure). Several preparation methods such as pulsed laser deposition,10 sputter deposition,11,12 and reactive molecular beam epitaxy (RMBE)13–18 have been used to grow magnetite on MgO(001).

The ideal substrate temperature during the deposition of magnetite thin films on MgO(001) is 250°C since higher growth temperatures induce interdiffusion and segregation of magnesium ions from the substrate to the surface of the magnetite film.19–21 However, lower temperatures than 250°C may lead to polycrystalline22 and poorly ordered magnetite films.23

During the preparation of MTJs, the material stacks are usually annealed to improve the crystalline quality of the barrier material and, therefore, increase the TMR. In case of MgO tunnel barriers adjacent to magnetite electrodes, the interdiffusion of the magnesium ions during annealing affects the magnitude and the sign of the TMR.24

The magnesium interdiffusion can be reduced, and higher annealing temperatures for magnetite are possible if iron buffer layers are deposited on the MgO substrate prior to the deposition of the magnetite films.25 Here, a 10 nm iron film functions as a sacrificing layer that is completely oxidized after the annealing process at higher temperatures, while the surface near region of the 10 nm overlaying magnetite film remains unaffected by the Mg interdiffusion. Another effect of these iron buffer layers is the reduced formation of anti phase boundaries (APBs),26 which are known to have significant influence on the magnetic properties of magnetite.27,28
Iron grows in the bcc structure with a lattice constant of 0.2866 nm so that the lattice mismatch concerning the surface unit cell of MgO ($a_{\text{MgO}} = 0.2978$ nm) is 3.75%. The lattice mismatch between magnetite and iron is 3.45%.

In a former study, we have investigated the magnetic in-plane anisotropy related to strain and thickness of magnetite films on MgO(001). Here, we investigate the impact of an initial iron buffer film on the magnetic properties of an overlying magnetite film. Since iron is a ferromagnet, it is reasonable that iron buffer layers will influence the magnetic properties of the overlying magnetite film. Therefore, in this study, we focus on the influence of the iron buffer layer on the in-plane magnetic anisotropy of magnetite films. The magnetite film thickness has been varied to study the thickness dependence. The magnetic anisotropy has been investigated by measurements via magneto-optic Kerr effect (MOKE). Furthermore, the stoichiometric and structural properties were analyzed by x-ray photoelectron spectroscopy (XPS), low energy electron diffraction (LEED), x-ray reflectometry (XRR), x-ray diffraction (XRD), and grazing incidence x-ray diffraction (GIXRD).

II. EXPERIMENTAL SETUP AND SAMPLE PREPARATION

The samples were prepared in an ultra high vacuum multi chamber system with a preparation chamber (base pressure of $10^{-8}$ mbar) and an analysis chamber (base pressure of $10^{-10}$ mbar). In the analysis chamber, it is possible to characterize film surfaces by LEED and XPS. The x-ray source of the XPS system is an Al K$_\alpha$ anode (1486.7 eV). The preparation chamber is equipped with an electron beam evaporator for iron, a heatable manipulator, and an oxygen source.

Ultra-thin Fe$_3$O$_4$ films were grown on iron buffer layers deposited on MgO(001) substrates before. Prior to film deposition, the MgO substrates were annealed at 400°C in a 10$^{-4}$ mbar oxygen atmosphere. Initially, Fe films were epitaxially grown at 250°C substrate temperature. Thereafter, Fe$_3$O$_4$ films of different thicknesses have been deposited by RMBE evaporating Fe in a 1 x $10^{-4}$ mbar oxygen atmosphere and at 250°C substrate temperature. During growth, the deposition rate has been controlled by a quartz microbalance, and after each film deposition, the stoichiometry and surface structure were checked by XPS and LEED, respectively.

XRR, XRD, and GIXRD measurements were carried out at the beamline P08 of HASYLAB (DESY, Hamburg) to determine the film thickness, interface properties, and the structure of the films. P08 is an undulator beamline with a preparation chamber (base pressure of 4 mbar oxygen atmosphere). Initially, Fe films were epitaxially grown at 250°C substrate temperature. Thereafter, Fe$_3$O$_4$ films of different thicknesses have been deposited by RMBE evaporating Fe in a 1 x $10^{-4}$ mbar oxygen atmosphere and at 250°C substrate temperature. During growth, the deposition rate has been controlled by a quartz microbalance, and after each film deposition, the stoichiometry and surface structure were checked by XPS and LEED, respectively.

A Kohzu 4S + 2D type diffractometer is installed at the endstation. The diffracted signal was measured by a Mythen strip detector.

Here, we anticipate that the two-step growth procedure leads to singular homogeneous magnetite films due to complete oxidation of the initial Fe films. This result is independent of the Fe buffer layer thickness. Therefore, all films are characterized by the final thickness of the oxide film in the following.

The magnetic properties of the films were analyzed by MOKE measurements in a setup reported earlier. The MOKE measurements were performed in the longitudinal setup geometry using a HeNe laser ($\lambda = 632.8$ nm) at an incidence angle of 45°. A photoelastic modulator with a modulation frequency of $f = 42$ kHz is used to modulate the polarization of the reflected beam. After the modulated polarization signal has been converted into an intensity signal by an analyzer (polarizer at 45°), the intensity is detected by a photo diode. The measured intensity of the 2f-signal (using the lock-in technique) is proportional to the Kerr rotation angle within the small angle approximation.

III. RESULTS

A. In situ surface characterization by XPS and LEED

Figure 1(a) depicts the distinct quadratic (1 x 1) structure of an initial clean MgO(001) surface, which originates from the surface unit cell of the rocksalt structure. The iron films have an ordinary (1 x 1) surface structure (cf. Fig. 1(b)). The (1 x 1) structure of iron is basically the same as the (1 x 1) structure of MgO since the surface unit cell of the body centered cubic iron unit cell adapts the surface unit cell of MgO. One of the iron films grown on MgO, however, exhibits a $(\sqrt{2} \times \sqrt{2})R45^\circ$ surface reconstruction shown Fig. 1(c). The $(\sqrt{2} \times \sqrt{2})R45^\circ$ iron reconstruction is an indicative of a step-terrace-based thin film that is pure and atomically flat. Therefore, both iron surfaces are crystalline and well-ordered and should be convenient buffer layers for magnetite growth.

![FIG. 1. LEED patterns](image)
The LEED measurements of iron oxide films grown by RMBE on the iron buffer films show always a \((\sqrt{2} \times \sqrt{2})R45^\circ\) superstructure, which is the well-known surface structure of well ordered magnetite \(^{33}\) (cf. Fig. 1(d)). Recently, a new model has been developed to explain the surface structure of magnetite based on subsurface cation vacancy stabilization of the magnetite(001) surface. \(^{34}\) The growth of \(\text{Fe}_3\text{O}_4\) and Fe films on MgO(001) substrates occurs in the so-called cube-on-cube epitaxy with \(\text{Fe}_3\text{O}_4(001); \langle 100 \rangle || \text{MgO}(001); \langle 100 \rangle\) and \(\text{Fe}(001); \langle 110 \rangle || \text{MgO}(001); \langle 100 \rangle\), respectively. Furthermore, the brilliant LEED patterns with sharp diffraction peaks demonstrate that the surface of the magnetite films has high quality.

The Fe 2p XPS measurements of the freshly prepared iron films (not shown) resulted in no trace of oxidation of the surface of the iron films. The Fe 2p\(_{3/2}\) and Fe 2p\(_{1/2}\) peaks have a binding energy of 706.8 eV and 719.7 eV, respectively, which is consistent with already reported binding energies of metallic iron. \(^{35,36}\) The XP spectra of all iron oxide films in Fig. 2 show the typical Fe 2p peak shape for stoichiometric magnetite. The peak position of the Fe 2p\(_{3/2}\) and the Fe 2p\(_{1/2}\) peaks is located at 710.6 eV and 723.6 eV, which is in well agreement with literature. \(^{37}\) In addition, the charge transfer satellites at 714.7 eV and at 718.8 eV, which are indicators for wustite and maghemite, respectively, are not visible separately since both satellites overlap forming a flat plateau between the Fe 2p doublet peaks. One exception is the sample with 20 nm RMBE grown iron oxide, which has a slightly higher amount of \(\text{Fe}^{2+}\) ions than the other films, and the peak is slightly shifted to smaller binding energies.

The Fe 3p peak region was analyzed to observe the Mg 2p peak (49 eV) that is next to the Fe 3p peak (55.5 eV). The measured Fe 3p spectra in Fig. 2 show no residual iron from the iron buffer films and no Mg 2p peak from the magnesium of the substrate. The Fe 3p peak is in any case an overlap of the Fe 3p originating from the \(\text{Fe}^{2+}\) ions and the Fe 3p originating from the \(\text{Fe}^{3+}\) ions. Hence, during the deposition of the iron oxide film, the surface of the iron film is completely oxidized and no segregation of \(\text{Mg}^{2+}\) ions can be observed.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig2.png}
\caption{XP spectra of the Fe 2p peak and the Fe 3p peak region.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig3.png}
\caption{CTR scans along (a) the (00L) rod and (b) the (01L) rod of iron oxide films grown by RMBE on previously on MgO deposited iron films and of iron oxide grown directly on MgO.}
\end{figure}

\section*{B. Film structure characterization by XRD}

XRD and GIXRD experiments were performed to analyze the atomic structure of the magnetite films formed by the two-step deposition procedure. The vertical direction is indexed with respect to the atomic layer distance, which is half size of the bulk unit cell due to the spacing between (001) crystal planes. During the diffraction experiments, the surface unit cell of MgO(001) is used for indexing of the 3D reciprocal space spanned by the MgO substrate. The surface unit cell is rotated by 45° in the (001) plane. As a consequence, e.g., the MgO(004)\(\beta\) rock salt bulk reflection is denoted by MgO(002)\(\delta\) using the surface indexing. Compared to MgO, magnetite has an almost doubled bulk lattice constant (cf. LEED patterns presented in Fig. 1). Thus, the magnetite (224)\(\delta\) inverse spinel bulk reflection is very close to (011)\(\delta\).

For different samples, the crystal truncation rod (CTR) scans along the (00L) direction and the (01L) direction are presented in Figs. 3(a) and 3(b), respectively. For comparison, a CTR scan of a 20 nm magnetite film grown directly on MgO without a previously grown iron film is also shown (one-step deposition procedure). On both CTRs, the latter shows clear diffraction peaks at magnetite Bragg positions and well developed Laue fringes. Therefore, the film has magnetite structure and has very homogeneous film thickness.

In contrast to this, the iron oxide films grown on Fe buffer layers by the two step deposition technique do not
show well developed magnetite Bragg peaks on the (00L) CTR. The Fe$_3$O$_4$(008)$_B$ Bragg peak is hardly seen for the 10 nm film and appears in the shoulder of the MgO(004)$_B$ Bragg peak for the 20 nm film. The picture is clearer for the (01L) CTR since the Fe$_3$O$_4$ (224)$_B$ Bragg peak is well separated from any MgO Bragg peak. Therefore, the Bragg peaks presented in Fig. 3(b) confirm the growth of magnetite films. However, the missing Laue fringes show that these films formed by the two-step deposition procedure are much rougher than magnetite films formed by the one-step growth procedure. So, the growth of magnetite on initially deposited iron films leads to films with inhomogeneous thickness and a bad crystalline quality although the surface structure of these films is well-ordered.

The (224)$_B$ Bragg reflection of magnetite originates exclusively from Fe$^{3+}$ ions on the tetrahedral sites of the inverse spinel structure.$^{17}$ Thus, since the (224)$_B$ Bragg reflection of magnetite can clearly be seen, this indicates an at least partially ordered tetrahedral sublattice of a crystalline magnetite film. However, compared to the Bragg peak for the magnetite film formed without any Fe buffer layer, the low intensity of the Fe$_3$O$_4$ (224)$_B$ Bragg peaks can be attributed to some higher degree of crystalline disorder of these two-step formed magnetite films.

In addition, we searched for the Fe(002)$_B$ Bragg peak and for the Fe(101)$_B$ Bragg peak but could not find any intensity. Therefore, the crystalline Fe of the buffer layer is completely oxidized. Furthermore, our XRR measurements (not shown), which are not only sensitive to crystalline material, can be fitted by one-layer models also indicating the complete consumption of the initial Fe film. In addition, the XRR analysis shows that the surface roughness of the magnetite films is 0.65 (±0.15) nm and the Fe$_3$O$_4$/MgO interface roughness is 0.7 (±0.3) nm. The surface and interface roughnesses are independent of the magnetite film thickness.

Thus, both results indicate that these films formed by the two-step deposition procedure have homogeneous stoichiometry and consist of magnetite while the buffer layer is completely consumed during the second step of preparation.

### C. Magnetic properties by MOKE

The MOKE measurements were performed at room temperature and reveal that all films have in-plane magnetization due to shape anisotropy. The Kerr rotation as a function of the external magnetic field is shown in Fig. 4. The Kerr angle $\kappa$ is a measure of the magnetization orientation. The magnetic easy axes are determined from the maximum Kerr rotation. Both 20 nm magnetite films feature a distinct fourfold magnetic anisotropy. The magnetic easy axes of the magnetite film grown directly on MgO point into Fe$_3$O$_4$ [110] directions as determined from the maximum coercive fields (cf. Fig. 5 (black)). This result is in agreement with the well-known [111] magnetic easy axes of bulk magnetite$^{39,40}$ if projected on the planar magnetite film due to the shape anisotropy.

We have measured magnetization curves for different azimuthal sample directions $\chi$ between MgO[100] and the applied magnetic field to analyze the magnetic structure in more detail. As can be seen in Fig. 5, the coercive field depends on the sample rotation. Both 20 nm magnetite films feature a distinct fourfold magnetic anisotropy. The magnetic easy axes of the magnetite film grown directly on MgO point into Fe$_3$O$_4$ [110] directions as determined from the maximum coercive fields (cf. Fig. 5 (black)). This result is in agreement with the well-known [111] magnetic easy axes of bulk magnetite$^{39,40}$ if projected on the planar magnetite film due to the shape anisotropy.

The coercive field plots of magnetite grown on Fe buffer layers shown in Fig. 5 (red) exhibit an unexpected behavior compared to the magnetite film without a Fe buffer layer. First, the magnetic easy axes are rotated by 45° so that the maximum of the coercive field point into Fe$_3$O$_4$ [100] directions. There is no dependence on the magnetite film thickness for this effect. The iron buffer layer can also not be the direct reason for this anisotropy rotation since both XRR and
XRD experiments have confirmed that there is no residual crystalline iron, which could influence the magnetic anisotropy. Second, the coercive fields of the films grown with the Fe buffer layer are drastically enhanced for all in-plane directions of the applied external field.

Fig. 6 presents the azimuthal dependence of the coercive field for all films studied here. The easy directions of all films are aligned to $[100]$ as shown by the maxima of the coercive field. The coercive fields of the 10 nm magnetite film is smaller than the coercive fields of the thicker films. Above 20 nm thickness, these fields do not differ very much, although the magnetic in-plane anisotropy seems to be diminished with increasing film thickness. This is studied in more detail in the following.

The coercive field in magnetic easy direction as a function of film thickness is depicted in Fig. 7. First, it becomes larger with increasing film thickness. However, the coercive field is slightly smaller for the sample with 87 nm compared to the sample with 28 nm film thickness. This may be due to a decreasing shape anisotropy.

The angular dependence of the coercive fields of all films displays additional maxima at exact magnetic hard directions (cf. Fig. 6). These maxima point to magnetic multidomain states in magnetic remanence and incoherent magnetization rotation (some magnetic domains rotate clockwise, some counterclockwise) between magnetic saturation and magnetic remanence. This effect can be described by magnetic frustration of the magnetic moments when the next magnetic easy axis has to be chosen during the reversal process.

Thus, we compared the coercive field of the easy direction with the coercive fields for $15^\circ$ off magnetic hard direction to characterize the in-plane magnetic anisotropy of the magnetite films studied here.

In order to estimate the anisotropy constant $K$ for a fourfold in-plane crystalline anisotropy without any additional anisotropies of varied symmetries, one typically uses

$$H_c = H_k = \frac{2K}{M_S}$$

(deduced from the Stoner-Wohlfarth model). Here, $H_k$ is the anisotropy field and can be determined from the saturation field in magnetic hard axes. The anisotropy constant $K$ can only be obtained in the ratio to $M_S$, since the absolute value of the saturation magnetization $M_S$ is not accessible in MOKE measurements. Unfortunately, the incoherent rotations of magnetic moments in the magnetic hard directions of our samples do not allow to relate the saturation field to $H_k$ properly within the Stoner-Wohlfarth model. Furthermore, the small Kerr rotation and the low signal-to-noise ratio do not provide reliable saturation field values. However, the coercive fields $H_c$ of the detected magnetization curves give a clear identification of the anisotropy, which therefore can be used to quantify the anisotropy by the ratio of coercive field values of magnetization curves for magnetic easy axes and for $15^\circ$ off magnetic hard directions.

The ratio is decreasing with increasing magnetite film thickness (cf. Fig. 8). This means that the strength of the fourfold in-plane magnetic anisotropy is decreasing with increasing film thickness.
increasing film thickness. This suggests that the magnetic fourfold anisotropy is influenced by the Fe$_3$O$_4$/MgO interface and the Fe$_3$O$_4$ surface. The ratio can be fitted with the expression

\[
\frac{H_{c_{\text{easy axis}}}}{H_{c_{\text{hard axis}}}} \propto K_{d}^{V} = K^{V} + \frac{2}{l} K^{S}.
\]  

(2)

Here, $K^{V}$ denotes the thickness independent bulk contribution, while the second term in Eq. (2) describes the contribution $K^{S}$ of the interface, which decreases with increasing film thickness.$^{44}$

The factor two is due to the consideration of both Fe$_3$O$_4$ surface and Fe$_3$O$_4$/MgO interface. The result of this fit is shown in Fig. 8, where the ratio of the applied coercive field in magnetic easy and 15° off magnetic hard direction is plotted against the reciprocal film thickness. The suitable fit confirms our assumption that the fourfold anisotropy is induced by the magnetite surface and the Fe$_3$O$_4$/MgO interface, whose influence weakens with increasing film thickness.

IV. DISCUSSION

XPS and LEED experiments of all films have revealed that there is no influence of the initial iron buffer layers on the near surface stoichiometry and the surface structure, respectively. The Fe 2p peaks show no characteristic satellites, indicating other iron oxide species nor metallic Fe and LEED exhibit the typical ($\sqrt{2} \times \sqrt{2}$)R45° surface superstructure of magnetite. One exception is the Fe 2p peak of the 28 nm film, which features a slightly smaller binding energy and a small charge transfer satellite indicating a higher amount of Fe$^{2+}$ ions. However, the LEED pattern is still the ($\sqrt{2} \times \sqrt{2}$)R45° surface superstructure observed only for magnetite with well-ordered surfaces. Thus, the surface characterization of the magnetite films grown on iron buffer layers shows no difference to magnetite films directly grown on MgO by RMBE.

However, XRD and GIXRD measurements have revealed that the structural quality of the bulk of these magnetite films grown on iron buffer layers is worse compared to magnetite films deposited directly on MgO. Although the presence of the Fe$_3$O$_4$(224)$_B$ indicates the growth of crystalline magnetite, the weak and broad Fe$_3$O$_4$(008)$_B$ Bragg reflections and the overall missing Laue fringes point out that the magnetite films on iron are not well ordered. Moreover, the crystallinity of the magnetite films does not increase with increasing magnetite film thickness but seems to be an inherent feature of the two-step preparation procedure. Thus, the initial iron buffer layers impair the structural properties of the magnetite films, although the iron buffer layer was completely oxidized during the reactive molecular beam epitaxy of magnetite as proven by XPS and XRD measurements. Furthermore, we like to emphasize that the effect of the two-step procedure on the surface roughness is less dramatic. Compared to films formed by one-step procedure, the surface roughness is only slightly increased. This finding is supported by the high brilliance of the LEED pattern.

This result is quite surprising on first sight since all materials Fe, Fe$_3$O$_4$, and MgO are lattice matched. Therefore, the bad crystalline quality of the magnetite film has to be attributed to the limited atomic transport during the oxidation of the Fe buffer layer when the Fe atoms have to take other sites. Obviously, the low crystalline quality of the rearranged oxidized buffer layer is continued in the magnetite film grown on top of the buffer layer.

The missing Mg 2p signal in the XP spectra of the Fe 3p region has shown that the iron buffer films have suppressed the Mg interdiffusion and that iron buffer films also functioned as a sacrificing layer since we can not detect any residual metallic iron or Mg with XPS and XRD. These findings confirm the results reported in literature$^{25}$ mentioned in the Introduction.

The MOKE measurements reveal that all samples have a fourfold magnetic in-plane anisotropy. However, the magnetite films grown on an iron buffer layer exhibit a magnetic in-plane anisotropy, which is rotated by 45° (cf. Fig. 5). Thus, these magnetite films have the magnetic easy axes along the in-plane (100) directions, instead along the (110) directions, which are typical for magnetite films.$^{39,40}$ This anisotropy rotation is independent of the film thickness (cf. Fig. 6). The rotation of the in-plane magnetic easy axes of magnetite films was observed before on magnetite grown directly on SrTiO$_3$ by infrared pulsed laser deposition.$^{45}$ In that study, the authors debate beside the higher strain on SrTiO$_3$ the presence of APBs as the reason for this surprising result. In our study, the rotated in-plane anisotropy might also be attributed to the presence of APBs. The low crystalline quality of these films as determined by XRD strengthens this assumption.

As mentioned in the Introduction, it has been reported that iron buffer films reduce the formation of APBs.$^{26}$ However, as we do not find any residual iron buffer layer in these samples, the formation of APBs could still be an explanation for this unexpected behavior. Further experiments have to be performed to shed more light on this special behavior of the magnetic anisotropy.

An evidence for the absence of metallic iron in the 10 nm to 28 nm films is also the small Kerr rotation in saturation. For pure iron films, the Kerr rotation is normally ten
times larger than for magnetite. Thus, residual iron should amplify the Kerr rotation. While the Kerr rotation in saturation of magnetite with and without iron buffer layer is almost equal, the applied coercive field is clearly larger for magnetite with iron buffer layer (cf. Fig. 5).

The large coercive field can also be attributed to the high defect density in the magnetite films. Therefore, this effect also points to a large amount of APBs. It has been reported that the coercive field usually decreases with increasing film thickness. This effect has been attributed to the reduced defect density for thicker films. For the films studied here, the coercive field for the easy axes increases until the film thickness is approximately 30 nm. Therefore, we conclude that the defect density increases initially for these films. An increasing coercive field with increasing film thickness has been observed in one of our prior studies too. For the thickest film, however, the coercive field decreases pointing to a less disordered film as usually expected.

The ratio of the applied coercive field in magnetic easy and 15° off magnetic hard direction is decreasing with increasing magnetite film thickness (cf. Fig. 8). This indicates that the fourfold shape anisotropy is influenced by the Fe₃O₄/MgO interface and the Fe₃O₄ surface, whose influence becomes negligible with increasing film thickness. A similar observation has been made before at Co films with a film thickness dependence. However, the strength of the magnetic anisotropy decreases with increasing film thickness. XPS and XRD measurements have indicated that the reduced defect density for thicker films. For the films investigated, the surface sensitive measurements like XPS and LEED indicate that iron buffer layers do not influence the structural and stoichiometric properties of magnetite. However, the characterization of the structural and magnetic properties of the whole film by XRD and MOKE reveals that the crystalline quality is poor and the magnetic in-plane anisotropy is rotated by 45°. Both crystalline quality and the rotation of the magnetic in-plane anisotropy show no film thickness dependence. However, the strength of the magnetic anisotropy decreases with increasing film thickness. XPS and XRD measurements have indicated that the iron buffer layer is completely oxidized during the second growth stage of the magnetite. The small Kerr rotation in the MOKE experiments of the samples with film thickness up to 28 nm confirms this result since a remaining iron film would cause a higher Kerr rotation.

Taken as a whole, the prepared magnetite films on initial grown iron buffer layers are convenient as magnetic layer in a MTJ due to the relatively small surface roughness and the higher coercive field compared to magnetite grown directly on MgO.

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