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## ABSTRACT

We have studied the structural and magnetic properties of enhanced-permeability-dielectric FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayer thin films deposited on 8"-Si wafers in an industrial magnetron sputtering system. The EPD-multilayers consist of 25 periods of alternating nanometer-thick FeCo-layers deposited by DC sputtering from a Fe<sub>60</sub>Co<sub>40</sub> target and Al<sub>2</sub>O<sub>3</sub>-interlayers deposited by RF sputtering from an Al<sub>2</sub>O<sub>3</sub> target. We tuned the magnetic properties of these thin films by varying the thickness of FeCo-layers from 1.1nm to 2.1nm, while the thickness of Al<sub>2</sub>O<sub>3</sub>-interlayers remained unchanged (3.5nm). The formation of layers of disconnected FeCo-nanoparticles separated by an Al<sub>2</sub>O<sub>3</sub>-matrix was revealed by grazing incidence small angle X-ray-scattering. Further insight into the microstructure of these layers was obtained from X-ray-reflectivity, highly asymmetric-X-ray-diffraction and non-coplanar grazing-incidence-diffraction. The Fe/Co ratio in the FeCo-layers obtained from X-ray-fluorescence measurements was  $(59\pm1)/(41\pm1)$ , which is in very good agreement with the nominal value in the Fe<sub>60</sub>Co<sub>40</sub> target. Using the standing wave technique we found that most of the Fe and Co atoms were located inside the polycrystalline grains, except for a small fraction that diffused into the Al<sub>2</sub>O<sub>3</sub>-matrix, and that the thinner the FeCo-layers thickness the higher the fraction of diffused atoms with respect to those in the grains. Zero-field-cooled, field-cooled, and hysteresis (*B-H*) and (*M-H*) measurements showed that the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers with FeCo-layers thinner than 1.7–1.8 nm exhibit superparamagnetic behavior (no coercivity and remanence) at room temperature with peak relative low-field permeability up to 887. By exceeding this critical thickness, the neighboring FeCo-aggregates started to coalesce, and this led to the ferromagnetic behavior revealed by a finite coercivity and remanence in the hysteresis loops.

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## I. INTRODUCTION

Nanoscale magnetic materials have received wide-spread interest in recent years due to their great potential for the development of new generation devices combining the advantages of semiconductor electronics with the spin-selective transport, e.g. magneto-resistive sensors, ultrahigh density memory cells and high-frequency programmable logic systems.<sup>1–3</sup> In particular, discontinuous ferromagnetic-insulator multilayers consisting of separated layers of closely spaced ferromagnetic nanoparticles embedded in an insulating matrix have received special attention because the magnetic and transport properties of these structures can be accurately

tuned within a wide range by simply changing the thickness of the ferromagnetic layer.<sup>4–12</sup>

Enhanced-permeability-dielectrics (EPD) are a particular class of discontinuous ferromagnetic-insulator multilayers<sup>13</sup> where the magnetic nanoparticles are smaller than the superparamagnetic (SP) limit. These materials have been introduced to reduce the switching field in arrays of single-layer magnetoresistive-random-accessmemory (MRAM) bits<sup>13,14</sup> and the nulling clock field in nanomagnetic logic (NML) systems<sup>15</sup> by increasing the magnetic flux density in the EPD material surrounding these devices.

In this work, we present a study of the structural and magnetic properties of nanoscale FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayer thin films deposited on 8"-Si wafers in an industrial, high-throughput magnetron sputtering system. We discuss the interdependence between structural and magnetic properties in these materials for FeCo-layer thicknesses in the ~1–2 nm range, where these layers are either discontinuous and/or near the continuous-discontinuous transition regime. Our objective is to fabricate EPD thin films with high magnetic permeability ( $\mu_r \gg 1$ ), no hysteresis, and no remanence, while still retaining their insulating properties.

#### **II. EXPERIMENTAL**

We deposited FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers on 8"-Si wafers in a high-throughput Evatec LLS-EVO-II batch sputter system (supplementary material).<sup>16-21</sup> The EPD-multilayers consisted of 25-periods of alternating FeCo-layers with nominal thickness in the 0.5-2nm range deposited by DC sputtering from a Fe<sub>60</sub>Co<sub>40</sub> target, and 3.5nm-thick (nominal) Al<sub>2</sub>O<sub>3</sub>-interlayers deposited by RF sputtering from an Al<sub>2</sub>O<sub>3</sub> target. Since our objective was to produce EPD layers with the highest relative permeability, we selected the Fe60Co40 magnetic alloy for this work because this material has the largest saturation magnetization of any material (~2.45 T).<sup>22</sup> Structural properties (e.g. layer thickness, vertical and lateral grain size, separation between the grains) of the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers were investigated by means of grazing-incidence-small-angle-X-ray-scattering (GISAXS), X-rayreflectivity (XRR), highly-asymmetric-X-ray diffraction (AXRD), non-coplanar grazing-incidence-diffraction (GID) and X-rayfluorescence (XRF) measurements (supplementary material). The discontinuity of the FeCo-layers was also qualitatively studied by room-temperature magnetic-force-microscopy (MFM) (supplementary material).

The evolution of magnetic properties with the FeCo-layer thickness was first investigated by means of *B-H* hysteresis loop measurements (averaged over the entire 8" wafers) in fields up to 0.1T using a Shb Instruments MESA200 measurement system. Magnetization measurements were then performed with a Quantum Design physical properties measurements (PPMS) setup in fields up to 7T, using a vibrating-sample-magnetometer (VSM) option. Temperature scans of magnetization were performed for both zero-field-cooling (ZFC) and field-cooling (FC) directions. Thus, ZFC measurements were performed by cooling the sample from room temperature to 10K in zero applied magnetic field, and then magnetization was recorded while heating the sample under a 1500e applied magnetic field. Moreover, magnetic field dependent magnetization measurements were performed at room temperature and at 10K.

#### **III. RESULTS AND DISCUSSION**

Owing to the much higher surface energy of FeCo compared to that of Al<sub>2</sub>O<sub>3</sub>,<sup>12</sup> the FeCo-layers exhibit a granular structure consisting of isotropically distributed nanoclusters as revealed by MFM and GISAXS measurements (see Fig. 1 and supplementary material). It was found that the approximate mean distance D between the nanocluster centers increases with the FeCo-layer thickness (Table I), and this can be attributed to the increase of nanoclusters with the FeCo-layer thickness. The individual layer thicknesses of the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers with nominal FeCo-layer thicknesses of 0.5-2nm (samples S1-S4) obtained from XRR measurements (Fig. 2a, b) are summarized in Table I. Thus, the FeCo-layer thicknesses obtained from XRR are in general larger than the nominal ones. To determine the average transversal and lateral grain sizes in the FeCo-layers we performed detector scans in both AXRD and GID geometries (Fig. 2c, d). Figure 2c shows the AXRD patterns (incidence angle  $\theta_i = 6^\circ$ ), where the peak at  $2\theta = 44.7^\circ$  corresponds to (110) crystallographic planes of the bcc-FeCo. For sample S1 the FeCo(110) peak was undetectable, which suggests that the FeCo-layers in this sample are amorphous.

We determined the grain size *d* along an inclined direction with respect to surface normal by  $(\theta_i - 6^\circ)$  using the Scherrer equation  $d=0.95\lambda/(FWHM \cdot \cos\theta)$ , where  $\lambda=0.154$ nm is the wavelength of X-rays,  $\theta=22.35^\circ$ ), and FWHM is the full-width-at-half-maximum in radians, corrected by the resolution function (Table I).<sup>23</sup> Since  $t_{XRR} \cdot d_{AXRD}$ , the FeCo-nanoclusters are limited by the layer thickness in the growth direction. However, in the GID measurements the FeCo(110) diffraction peaks are much sharper, and therefore the grains are larger (Table I). Moreover, it is also observed that the thicker the FeCo-layers the larger the grains, which is consistent with the GISAXS measurements. We have also performed AXRD measurements using a 2D-detector that allows recording the diffraction



**FIG. 1.** Growth model of the discontinuous  $FeCo/Al_2O_3$  multilayers. For thicknesses below a certain limit ( $t_0$ ) the FeCo layers are discontinued, and above this critical thickness the FeCo particles gradually merge forming quasi-continuous layers;  $d_1$  and  $d_2$  are the average lateral dimensions of the magnetic particles for FeCo layer thickness  $t_1$  and  $t_2$ , respectively, and *D* is the average nearest-neighbor distances between the FeCo nanoclusters.

**TABLE I.** Structural analysis of  $25 \times (FeCo/Al_2O_3)$ -multilayers by various X-ray scattering techniques, where  $t_{nom}$  and  $t_{XRR}$  are the nominal and experimental layer thicknesses obtained from XRR,  $d_{AXRD}$  and  $d_{GID}$  are the grain sizes obtained from AXRD and GID measurements, respectively, and  $D_{GISAXS}$  is the mean distance between the FeCo-nanoclusters. One should note that  $d_{AXRD}$  is determined along a direction inclined with the surface normal by an angle of 12.35°, whereas  $d_{GID}$  is determined in the sample plane.

	t <sub>nom.</sub> [nm]		t <sub>XRR</sub> [nm]				
Sample	FeCo	$Al_2O_3$	FeCo	Al <sub>2</sub> O <sub>3</sub>	d <sub>AXRD</sub> [nm]	$d_{GID} [nm]$	D <sub>GISAXS</sub> [nm]
S1	0.5	3.5	$1.15 \pm 0.10$	$3.32\pm0.10$	_	_	3.4
S2	1.0	3.5	$1.64\pm0.10$	$3.37\pm0.10$	$1.30\pm0.10$	$8.0 \pm 1.0$	5.5
S3	1.5	3.5	$1.85\pm0.10$	$3.79\pm0.10$	$1.90\pm0.08$	$10.5 \pm 1.0$	7.4
S4	2.0	3.5	$2.10\pm0.10$	$3.85\pm0.10$	$2.30\pm0.06$	$12.0\pm1.0$	9.6

signal out of the scattering plane ( $\pm 15^{\circ}$ ), and as such is also sensitive to texture. Thus, we found out that the FeCo(110) peak at  $2\theta$ =44.7° consists of a Debye ring shape (supplementary material), and therefore the FeCo-layers have no preferential orientation in the detecting range.

To get insight into chemical composition of FeCo-layers, we performed incidence angle scans  $(2\theta_{inpl}=44.7^{\circ})$  for GID and XRF (Fig. 2e, f), which are depth-sensitive to crystalline material content and chemical composition, respectively.<sup>24,25</sup> We observe that both GID and XRF profiles exhibit S-shaped kinks for incidence angles corresponding to the positions of superlattice peaks in the XRR patterns, which are due to the standing waves formed inside

the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayer by the incident and reflected waves. The fitting parameters of the GID profiles were the thicknesses of the FeCo-layers, whereas the XRF profiles were modelled with the Fe and Co concentrations (black lines in Fig. 2e, f). Since the amplitude of the S-shaped kinks in the GID and XRF profiles is different, the crystallinity can vary within the multilayer; in other words, not all Fe and Co atoms that contributed to the XRF profiles are located inside the polycrystalline grains, but they can also diffuse into the Al<sub>2</sub>O<sub>3</sub>-matrix. Moreover, the FeCo layers may also contain a certain fraction of Al<sub>2</sub>O<sub>3</sub>. For samples S1–S4 in Table I we found that 15%, 12%, 10% and 8% of the Fe and Co atoms have diffused into the Al<sub>2</sub>O<sub>3</sub>-matrix with respect to the FeCo-layers.



**FIG. 2.** a) Reflectivity RSM of 25-period FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayer with 2nm-thick (nominal) FeCo-layers (sample S4). The peaks along  $Q_z$ -direction for  $Q_y=0$  (depicted by black arrows) arise from the constructive interference of X-rays scattered at various buried interfaces. The white arrows indicate the diffused scattering due to the granular FeCo layers. b) Specular XRR scans (shifted vertically for clarity) along  $Q_z$ -direction ( $Q_y=0$ ) for samples S1–S4 in Table I. Black line is fit of the XRR pattern for sample S2. c) Detector 20-scans (shifted vertically for clarity) in the AXRD scattering geometry for samples S1–S4 (incidence angle  $\theta_i=6^\circ$ ); the black line is a Gaussian fit of the FeCo(110) diffraction peak. d) Detector  $2\theta_{inpl}$ -scans (shifted vertically for clarity) in the GID geometry for samples S1–S4 (detector angle  $\theta_i=1^\circ$ ); the black line is a Gaussian fit of the FeCo(110) diffraction peak. e) Incidence angle  $\theta_i$  scans (shifted vertically for clarity) of Fe and Co XRF signals. The black lines in panels (e) and (f) represent fits using the standing wave technique.

Thus, the total intermixing decreases when the FeCo layer thickness increases. Furthermore, by comparing the intensity of the XRF profiles for Fe and Co, after correcting for the different scattering radii of the Fe and Co atoms, we can deduce that the Fe/Co ratio in the FeCo-layers for samples S1 to S4 is  $(59\pm1)/(41\pm1)$ , which is agrees very well with the nominal value of the Fe<sub>60</sub>Co<sub>40</sub> target.

Figure 3 shows temperature dependent magnetization measurements under weak magnetic field for ZFC and FC directions for the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers with FeCo-layer thicknesses ranging from 1.33nm to 2.03nm (measured by XRR). The multilayer with 1.33nm thick FeCo-layers exhibits a clear paramagnetic response (blue curve in Fig. 3a). Increasing the FeCo-layer thickness to 1.42nm leads to formation of magnetic nanostructures, which appear as a thermally relaxed peak on the ZFC curve at 26K (Fig. 3b). Furthermore, the multilayer with 1.56nm thick FeColayers also exhibits a thermally relaxed behavior on the ZFC curve at a slightly elevated temperature, 31K (Fig. 3b). We attribute the peaks in the ZFC curves of EPD samples with 1.42nm and 1.56nm thick FeCo-layers to the SP behavior of the discontinuous FeCo-layers.<sup>13</sup> However, the multilayer with 1.69nm thick FeCo-layers exhibits a broader thermal relaxation compared to the thinner films (Fig. 3b), and this could be assigned to a broader size distribution of the FeConanostructures leading to the existence of both ferromagnetic and SP phases at the same time.

Thermally relaxed SP behavior disappears by further increasing the FeCo-layer thickness (Fig. 3a, b). One can estimate the average magnetic particle volumes (V) from the observed blocking temperatures  $(T_b)$  by using the formula  $T_b \approx K_u V/30k_B$ , where  $K_u$  is the anisotropy energy of the bulk material, and  $k_B$  is the Boltzmann constant.<sup>26</sup> Using an anisotropy energy of  $0.1 MJ/m^3$  for  $Fe_{60}Co_{40}$ alloy,<sup>27</sup> the above formula yields average magnetic particle volumes of 25.7nm<sup>3</sup> and 128nm<sup>3</sup> (particle diameters of 3.66nm and 6.25nm if one assumes they are spherical) for the multilayers with nominal FeCo-layer thicknesses of 1.42nm and 1.56nm, respectively. However, since the particles are limited in the growth direction by the layer thickness, they could be more realistically approximated by thin disks with thicknesses of 1.42nm and 1.56nm, and the diameters of 4.8nm and 10.2nm, respectively. These values are consistent with the GID measurements discussed before, where sample S2 with 1.64nm thick FeCo-layers have grains with an average diameter of 8nm. Magnetization of the SP particles above blocking temperature can be expressed by  $M=M_0L(x)$ , where  $M_0$  is the saturation magnetization and L(x) represents the Langevin function with  $x=\mu B/k_B T.^{28}$  By fitting the experimental magnetization data with the Langevin equation one can estimate the average magnetic particle volume. Thus, for multilayers with FeCo-layer thicknesses of 1.42nm and 1.56nm one obtains average particle volumes of 25nm<sup>3</sup> and 37nm<sup>3</sup>, respectively. While the first value matches that obtained from  $T_b$ , the second one does not. This discrepancy could be attributed to the nature of the Langevin function, which needs further correction for different particle shapes with broader size distribution.<sup>29</sup>

Room-temperature magnetic-field-dependent magnetization measurements of the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers with FeCo-layer thicknesses (measured by XRR) ranging from 1.33nm to 2.03nm are presented in Fig. 4a. Saturation magnetization scales with the FeCo-layer thickness, and eventually for the thickest layers (1.96nm, 2.03nm) reaches the magnetization values expected for thick (>50nm) FeCo-films.<sup>30,31</sup> Similar to the magnetic-fielddependent magnetization measurements, magnetic flux densities (B, not shown here) also scale with the FeCo-layer thickness. Due to their higher sensitivity at low fields, we used the *B*-*H* hysteresis loop measurements to determine the coercive field  $(H_c)$  as a function of layer thickness (Fig. 4d, e). We observe that the multilayers with FeCo-layers below the thickness of 1.74nm exhibit SP behavior (no coercivity and remanence) at room temperature. By exceeding this critical thickness, the multilayers start to gradually exhibit ferromagnetic properties revealed by a finite coercivity and remanence in the hysteresis loops. We also used the B-H hysteresis loops to estimate the magnetic permeability as shown in Fig. 4c, d. The obtained peak magnetic permeability for the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers versus the FeCo-layer thickness at the field  $H^* \sim 0.3$  Oe is shown in Fig. 4f, where the sample with 1.33nm-thick FeCo-layers was excluded because it only has a paramagnetic response. Thus, we observe that relative permeability exhibits an increase from 7 to 20 when the thickness of the FeCo-layers increases from 1.42nm to 1.56nm, but then it drops to 5.6 when the thickness is increased to 1.69nm, and this could be due to the formation of larger structures having a ferromagnetic response, as explained above. By further increasing the FeCo-layer thickness, the relative permeability increases rapidly and at 2.03nm it exceeds 10000. These results show that the magnetic properties of the FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers are extremely sensitive to the FeCo-layer thickness. Compared to similar EPD thin films based



**FIG. 3.** Temperature-dependent magnetization curves, e.g. zero-field cooled/ field cooled (ZFC-FC) measured under a 150 Oe magnetic field for 25-period FeCo/Al<sub>2</sub>O<sub>3</sub>-multilayers with the FeCo layer thickness ( $d_{FeCo}$ ) measured by XRR ranging from 1.33nm to 2.03nm. To better observe the behavior of magnetization at low temperatures for the discontinuous multilayers, the ZFC-FC magnetization curves for samples with 1.42nm, 1.56nm, 1.69nm and 1.74nm FeCo-layers are depicted separately, together with the corresponding blocking temperatures.



on FeCo and Al<sub>2</sub>O<sub>3</sub> reported in literature, our FeCo/Al<sub>2</sub>O<sub>3</sub> multilayers exhibit substantially higher relative permeabilities for similar FeCo-layer thicknesses.<sup>13,15</sup>



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## **IV. CONCLUSIONS**

In summary, we deposited nanostructured  $FeCo/Al_2O_3$ multilayers in an industrial, high-throughput Evatec LLS EVO II magnetron sputtering system. By performing a thorough X-ray scattering analysis we revealed the granular structure of the FeCo-layers, which evolves from isolated nanoparticles towards continuous layers with increasing layer thickness. Magnetic measurements showed that the multilayers with FeCo-layers below the thickness of 1.74nm exhibited superparamagnetic behavior (no coercivity and remanence) at room temperature, and by exceeding this critical thickness, the neighboring FeCo-islands started to coalesce, and this led to the ferromagnetic behavior revealed by a finite coercivity and remanence in the hysteresis loops.

#### SUPPLEMENTARY MATERIAL

See supplementary material for details on the LLS EVO II sputter tool, and X-ray scattering and MFM experiments.

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