Tailoring the soft magnetic properties of sputtered multilayers by microstructure engineering for high frequency applications

Claudiu V. Falub,1 Hartmut Rohrmann,1 Martin Bless,1 Mojmír Meduňa,2,3 Miguel Marioni,4 Daniel Schneider,1 Jan H. Richter,1 and Marco Padrun1

1Evatec AG, Hauptstrasse 1a, CH-9477 Trübbach, Switzerland
2Department of Condensed Matter Physics, Masaryk University, Kotlářská 2, CZ-61137 Brno, Czech Republic
3CEITEC, Masaryk University, Kamenice 5, CZ-60177 Brno, Czech Republic
4Nanoscale Materials Science, Empa, Überlandstrasse 129, CH-8600 Dübendorf, Switzerland

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II. EXPERIMENTAL

Soft magnetic Ni78.5Fe21.5, Co91.5Ta4.5Zr4 and Fe52Co28B20 thin films laminated with SiO2, Al2O3, AlN, and Ta2O5 dielectric interlayers were deposited on 8” Si wafers using DC, pulsed DC and RF cathodes in the industrial, high-throughput Evatec LLS-EVO-II magnetron sputtering system. A typical multilayer consists of a bilayer stack up to 50 periods, with alternating (50-100) nm thick magnetic layers and (2-20) nm thick dielectric interlayers. We introduced the in-plane magnetic anisotropy in these films during sputtering by a combination of a linear magnetic field, seed layer texturing by means of linear collimators, and the oblique incidence inherent to the geometry of the sputter system. Depending on the magnetic material, the anisotropy field for these films was tuned in the range of (7-120) Oe by choosing the appropriate interlayer thickness, the aspect ratios of the linear collimators in front of the targets, and the sputter process parameters (e.g. pressure, power, DC pulse frequency), while the coercivity was kept low, (0.05-0.9) Oe. The alignment of the easy axis (EA) on the 8” wafers was typically between ±1.5° and ±4°. We discuss the interdependence of structure and magnetic properties in these films, as revealed by atomic force microscopy (AFM), X-ray reflectivity (XRR) with reciprocal space mapping (RSM) and magneto-optical Kerr effect (MOKE) measurements. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). [http://dx.doi.org/10.1063/1.4973945]

I. INTRODUCTION

The continuous trend towards miniaturization with increased functionality for internet-based mobile systems increasingly requires that various high-performance devices (e.g. III-V electronic/optoelectronic devices, low-power sensors, RF filters, micro- and nanoscale transducers, inductors and transformers, MEMS actuators, etc.) be combined with silicon complementary metal-oxide-semiconductor (Si-CMOS) technology in a monolithic integrated circuit (IC) fabrication process.1–3 As far as the CMOS-integrated magnetic and electro-magnetic devices are concerned, soft magnetic layers (typically several microns thick) with tunable uniaxial anisotropy are key for applications requiring operation from high to ultra-high frequencies (from a few MHz to several GHz) and low power consumption.4,5 These films should have high saturation magnetization (Ms) and electrical resistivity (ρ), and a well-defined anisotropy with a specific in-plane EA and anisotropy constant (Ku).6 The AC permeability (μ) perpendicular to EA remains largely constant up to the ferromagnetic resonance (FMR) frequency of the material (fFMR ~ Hk/4πMr, where Hk is the anisotropy field).7 Thus, by tuning Ms and Hk with appropriate material choices one can adapt to different frequency requirements (e.g. GHz applications typically require films with Hk>20 Oe). Moreover, to avoid hysteresis...
loss (i.e. coercivity $H_c \leq 1$ Oe) these films are laminated with intermediate non-magnetic interlayers, which not only stop the crystallite growth, but also reduce the Eddy current loss.

In the case of magnetron sputtering, the in-plane uniaxial magnetic anisotropy is strongly affected by the film microstructure/stress that can be controlled by process parameters (e.g. temperature, pressure, power, etc.). It is well known that the amount of disorder at the surfaces and interfaces, which is also influenced by process parameters, can greatly disturb the soft magnetic properties of thin films, such as coercivity, magnetic domain structure, magnetization reversal and magnetic anisotropy. Therefore, when tailoring the soft magnetic multilayers for high-frequency operation it is important to predict and control the microstructure and surface/interface roughness.

In this paper, we present a systematic study on the interdependence of structural and magnetic properties of Ni$_{78.5}$Fe$_{21.5}$, Co$_{91.5}$Ta$_{4.5}$Zr$_4$ and Fe$_{52}$Co$_{28}$B$_{20}$ soft magnetic thin films laminated with SiO$_2$, Al$_2$O$_3$, AlN, and Ta$_2$O$_5$ dielectric interlayers, sputtered in an industrial, high-throughput magnetron sputtering system. In view of their large saturation magnetizations, e.g. $\sim (0.8-1)$ T for Ni$_{78.5}$Fe$_{21.5}$, $\sim (1.5-1.7)$ T for Co$_{91.5}$Ta$_{4.5}$Zr$_4$ and $\sim (1.8-2)$ T for Fe$_{52}$Co$_{28}$B$_{20}$, these soft magnetic materials currently receive much attention for their potential for high frequency applications.

II. EXPERIMENTAL

We prepare multilayers based on the Ni$_{78.5}$Fe$_{21.5}$, Co$_{91.5}$Ta$_{4.5}$Zr$_4$ and Fe$_{52}$Co$_{28}$B$_{20}$ soft magnetic alloys with different interlayer materials, interlayer thickness, total film thickness, process parameters and angular distribution of the sputtered material. More specifically, we deposit these films on 8” bare Si wafers and Si/200nm-thermal-SiO$_2$ wafers in the high-throughput Evatec LLS-EVO-II sputter system. The system is equipped with five cathodes placed in a vacuum chamber (base pressure <10$^{-8}$ mbar), a moveable shutter and a rotating substrate cage housing that allows nine 8” wafers to be processed in the same batch (see Fig. S1 in the supplementary material for the schematics of this system). The magnetic sublayers, which represent the greater fraction of the soft magnetic multilayer stack, are deposited by DC and pulsed DC (duty cycle 40% unless otherwise specified) sputtering at $\sim (0.5-2.0) \times 10^{-3}$ mbar from long-life (nominal 250 kWh) targets, which allow $\sim 1000$ 8” wafers to be sputtered with 1 µm thick soft magnetic material. The non-magnetic (AlN, Ta$_2$O$_5$, Al$_2$O$_3$, SiO$_2$) interlayers are deposited by RF or reactive DC sputtering from monoblock Al, Ta, Al$_2$O$_3$ and SiO$_2$ targets at $\sim 5 \times 10^{-3}$ mbar.

To induce in-plane magnetic anisotropy one option is to resort to modifications to sputter geometry allowed by the LLS-EVO-II system. By reducing the speed with which the Si wafers mounted on the rotating cage move into a position facing the sputter cathodes, we can increase the fraction of time during which there is an oblique incidence of the sputtered material, which leads to a horizontal linear order in the growing films. By using horizontal or vertical collimators (HC, VC) with different aspect ratios a:b (collimator plate height: collimator plate separation), this effect can be enhanced or reduced, giving additional freedom to engineer the magnetic anisotropy. The other option for inducing uniaxial anisotropy is by depositing the films in the presence of a linear magnetic field parallel to the wafer plane, which is designed such that the magnetic field of the magnetron located behind the opposite target is not perturbed.

Magnetic properties of the sputtered multilayers, e.g. $H_c$, $H_k$ and the angular dispersion of EA ($D$), are mapped on the 8” wafers by means of MOKE. Surface morphology and root-mean-square roughness ($R_q$) are measured by AFM using Park-Systems-NX20 and Bruker-Dimension-Icon3 microscopes. The degree of crystallinity is probed by grazing incidence X-ray diffraction (GIXRD) using a Bruker-D8-Discover diffractometer. Layer thicknesses and surface/interface roughness are determined by XRR with specular scans and RSMs that are collected at various azimuths between 0° (∥EA) to 90°(⊥EA) using a Rigaku-SmartLab-3kW diffractometer.

III. RESULTS AND DISCUSSION

A. Structural properties

Figure 1 shows the surface morphology probed by AFM of various soft magnetic multilayers sputtered in the LLS-EVO-II system. The DC-sputtered NiFe multilayers with Ta$_2$O$_5$ and AlN targets at $\sim (0.8-1)$ T for Ni$_{78.5}$Fe$_{21.5}$, $\sim (1.5-1.7)$ T for Co$_{91.5}$Ta$_{4.5}$Zr$_4$ and $\sim (1.8-2)$ T for Fe$_{52}$Co$_{28}$B$_{20}$, these soft magnetic materials currently receive much attention for their potential for high frequency applications.
interlayers exhibit a random, granular microstructure with no visible order (Fig. 1(a)). GIXRD measurements reveal the crystal structure of these films as face-centered-cubic (fcc) with almost no preferential texture. The $R_q$ of the 1 $\mu$m thick NiFe multilayers with 2 nm, 4 nm and 8 nm thick Ta$_2$O$_5$ interlayers are 1.89 nm, 1.92 nm and 2.01 nm, respectively. Moreover, the NiFe multilayers with AlN interlayers exhibit similar surface morphology and roughness. In contrast, the multilayers based on CoTaZr and FeCoB alloys exhibit surface morphologies consisting of ripples elongated perpendicular to the incident flux direction (Fig. 1(b–e)). These surface ripples are a consequence of the self-steering effect during deposition, and they usually develop for oblique incidence deposition at small incidence angles.$^{26,27}$ No crystallographic phases are present in the GIXRD patterns, which indicates that these multilayers are amorphous. We find that the total thickness of the film not only affects the roughness but also surface morphology (Fig. 1(b,d)) where by increasing the thickness of the CoTaZr/Al$_2$O$_3$ multilayers from 1 $\mu$m to 4 $\mu$m the elongated surface ripples become more pronounced. One-dimensional (1D) AFM scans reveal the periodicity of the ripples as $\sim$22.5 nm.

Surface morphology of the multilayer films can be controlled by using collimators for both the magnetic and non-magnetic interlayers. Thus, by sputtering the magnetic layers with a 2:1 HC (sample S1 in Table I), the surface ripples become more evident (Fig. 1(e)); periodicity of the ripples determined from 1D-AFM scans is 35 nm. In this case, the horizontal oblique incidence effect caused by the substrate rotation is enhanced, whereas the vertical oblique incidence contribution from the elongated target is suppressed. The difference between the ripple periodicity for the CoTaZr/Al$_2$O$_3$

![FIG. 1. 1 $\mu$m x 1 $\mu$m AFM scans of soft magnetic multilayers sputtered with different process parameters (values in the lower right corners represent the corresponding $H_k$ values): (a) 12×(80nm-NiFe/4nm-Ta$_2$O$_5$) [$R_q$=1.89 nm]; (b) 12×(80nm-CoTaZr/4nm-Al$_2$O$_3$) [$R_q$=0.30 nm]; (c) 10×(100nm-FeCoB/4nm-AlN) [$R_q$=0.46 nm]; (d) 48×(80nm-CoTaZr/4nm-Al$_2$O$_3$) [$R_q$=0.69 nm]. 1 $\mu$m x 1 $\mu$m AFM scans of 10×(100nm-FeCoB/4nm-AlN) multilayers (see Table I for process conditions): (e) S1 [$R_q$=0.97 nm]; (f) S2 [$R_q$=1.02 nm]; (g) S3 [$R_q$=1.15 nm]; (h) S4 [$R_q$=0.84 nm]; (i) S5 [$R_q$=0.93 nm].]
and FeCoB/AlN multilayers could be due to the different surface (Al₂O₃ vs. AlN buffer layer) and energy of the incoming particles (DC vs. pulsed DC)²⁸,²⁹ When in addition to the 2:1 HC for the FeCoB layers we use a 1:1 VC for the AlN interlayer (sample S2 in Table I), the surface morphology no longer exhibits elongated ripples (Fig. 1(f)) even though surface roughness remains unchanged (∼1 nm).

Additionally, we observe that the process pressure and the type of sputter process (e.g. DC, pulsed DC) has a large impact on the roughness and surface morphology (Fig. 1(g–i) and Table I), and we expect this to also greatly affect the magnetic properties. We should only point out that when a very low process pressure of 5X10⁻⁴ mbar is used for the FeCoB layers, the surface roughness of the 1 μm thick FeCoB/AlN multilayer is reduced by a factor of 7.7 (i.e. from 1.15 nm to 0.15 nm), and the surface morphology no longer exhibits ripples.

To obtain insight about the roughness of the buried interfaces, we perform XRR measurements. Figure 2(a) compares the specular XRR scans of 80nm-CoTaZr/4nm-Al₂O₃ multilayers with 6, 12 and 48 periods, corresponding to total thicknesses of ∼0.5 μm, 1 μm and 4 μm, respectively. For comparison, the specular scan of 80nm-NiFe/4nm-Ta₂O₅ multilayer with 12 periods (total thickness ∼1 μm) is also shown. We determine the individual layer thickness and the interface roughness by fitting the specular scans with a model that assumes the layer roughness to increase from the lowest interface to the top surface; an example of such a fit is presented in Fig. 2a for the CoTaZr/Al₂O₃ multilayer with 6 periods. Consequently, for the 6-period CoTaZr/Al₂O₃ multilayer XRR provides layer thicknesses/roughnesses of 81.1 nm/0.4 nm (CoTaZr) and 4.5 nm/0.2 nm (Al₂O₃), consistent...
with AFM ($R_q=0.27$ nm). Moreover, we find the individual layer roughness to increase from ~0.2 nm at the lowest interface to ~0.4 nm, ~0.6 nm and ~1 nm at the top surface for the 6, 12 and 48-period structures, respectively. For comparison, the interface roughness of the NiFe/Ta$_2$O$_5$ multilayer with 4 nm thick Ta$_2$O$_5$ interlayers is much higher, e.g. ~1.8 nm, in very good agreement with AFM (1.89 nm). In the case of FeCoB-based multilayers with different laminating materials, by fitting the specular XRR scans in Fig. 2(b) we find the Al$_2$O$_3$ interlayers to provide the smoothest FeCoB interfaces (<0.3 nm) where the roughness of the dielectric interlayers is ~0.4 nm, which is also in good agreement with AFM ($R_q=0.38$ nm). Furthermore, we also investigate the effect of interlayer thickness on surface and interface roughness. Thus, for FeCoB/AIN multilayers XRR shows that the interface roughness of AIN interlayers increases from ~0.4 nm to ~0.6 nm when their thickness increases from 4 nm to 20 nm; interface roughness of the top FeCoB layers is ~0.6 nm. Similar results are obtained for NiFe/Ta$_2$O$_5$ multilayers with Ta$_2$O$_5$ interlayers.

The XRR technique is used to also probe the diffuse scattering (i.e. off-specular region in the reciprocal space), which can give unique information about the correlation of interfacial roughness between successive layers. Figure 2(c,d) shows RSMs of 48×(80nm-CoTaZr/4nm-Al$_2$O$_3$) multilayer for two azimuths, e.g. perpendicular and parallel to the surface ripples, respectively. Consequently, the anisotropy of the multilayer stack is confirmed by the more intense diffused scattering for the measurement with the X-ray beam path along the surface ripples, e.g. for $Q_z=0.2238$ Å$^{-1}$ the intensity of the diffused scattering increases by a factor ~2.

### B. Magnetic properties

Figure 3(a,b) shows the MOKE hysteresis loops for ~1 μm thick 12×(80nm-NiFe/4nm-AlN), 12×(80nm-CoTaZr/4nm-AlN) and 10×(100nm-FeCoB/4nm-AlN) multilayers for EA and HA orientations; the corresponding $H_c$ and $H_k$ values are 0.08 Oe/0.12 Oe/0.20 Oe and 8.5 Oe/17.1 Oe/25.5 Oe, respectively. Moreover, we see that $D$ rapidly deteriorates, while $H_c/H_k$ increases/decreases when the interlayer thickness increases. This is consistent with the results discussed in the previous section, where roughness is larger for the multilayers with thicker interlayers. Furthermore, increasing

![MOKE hysteresis loops](image-url)

**FIG. 3.** MOKE hysteresis loops of 12×(80nm-NiFe/4nm-AlN), 12×(80nm-CoTaZr/4nm-AlN), 10×(100nm-FeCoB/4nm-AlN) for: (a) EA; (b) HA. (c), (d) and (e) $D$, $H_c$ and $H_k$ of 12×(80nm-NiFe/Ta$_2$O$_5$), 12×(80nm-CoTaZr/Al$_2$O$_3$) and 10×(100nm-FeCoB/AlN) as a function of interlayer thickness. (f) wafer distributions for 48×(80nm-CoTaZr/Al$_2$O$_3$): (h) $D$; (i) $H_c$; (j) $H_k$. MOKE hysteresis loops of 10×(100nm-FeCoB/4nm-AlN) multilayers in Table I for: (f) EA, (g) HA.
TABLE I. Magnetic properties ($D$, $H_c$, $H_k$) and $R_q$ of 10×(100nm-FeCoB/4nm-AlN) multilayers sputtered on 8" Si/SiO$_2$ wafers with different process conditions.

| Sample | Process conditions | $R_q$ [nm] | $D$ [°] | $H_c$ [Oe] | $H_k$ [Oe] |
|--------|--------------------|------------|---------|------------|------------|
| S1     | FeCoB: 2:1 HC      | 0.97       | 3.1     | 0.97±0.14  | 41.6±2.1   |
| S2     | FeCoB: 2:1 HC, AlN: 1:1 VC | 1.02 | 1.9     | 0.37±0.11  | 118±2      |
| S3     | FeCoB: 3.5×10$^{-3}$ mbar | 1.15 | 4.2     | 0.85±0.09  | 83.8±2.6   |
| S4     | FeCoB: non-pulsed DC | 0.84       | 2.6     | 0.18±0.03  | 65.1±1.3   |
| S5     | FeCoB: pulsed DC (10% duty cycle) | 0.93 | 1.6     | 0.24±0.06  | 53.9±1.2   |
| S6     | FeCoB: 5×10$^{-4}$ mbar | 0.15       | 2.8     | 0.34±0.04  | 28.9±0.3   |

the total thickness of the multilayer stack only slightly deteriorates the soft magnetic properties, as shown in Fig. 3(h–j) for 48×(80nm-CoTaZr/Al$_2$O$_3$). Thus, in spite of the complex, 4 μm thick structure, consisting of almost 100 interfaces, we obtain superior soft magnetic properties with excellent uniformity across the 8" wafer, e.g. $D<±2°$, $H_c<0.2$ Oe and $H_k<17.1$ Oe.

Finally, we want to emphasize the key role of process conditions in controlling the soft magnetic properties of sputtered multilayers. Figure 3(f,g) compares the hysteresis loops for EA and HA of FeCoB/AlN multilayers sputtered with different process conditions (see also Table I). Magnetic anisotropy in these structures is induced by the aligning magnetic field and the oblique incidence inherent to the LLS-EVO-II system, the latter being adjusted by means of linear collimators. The soft magnetic properties of these structures are summarized in Table I. Thus, by choosing the appropriate process parameters one can tune $H_k$ from ~29 Oe to ~120 Oe, while $H_c<0.9$ Oe and $D$ goes between ±1.5° and ±4°.

IV. CONCLUSIONS

In summary, we performed a systematic study of the structural and magnetic properties of high quality, soft magnetic multilayers based on the Ni$_{78.5}$Fe$_{21.5}$, Co$_{91.5}$Ta$_{4.5}$Zr$_{4}$ and Fe$_{52}$Co$_{28}$B$_{20$ alloys, deposited with the industrial, high-throughput Evatec LL-EVO-II magnetron sputtering system. We showed how soft magnetic properties of these multilayered structures can be adjusted by means of microstructure engineering. We provide an industrial sputtering solution for high-quality soft magnetic multilayers with tunable magnetic properties for applications requiring operation from high to ultra-high frequencies.

SUPPLEMENTARY MATERIAL

See supplementary material for schematics of the LLS-EVO-II tool.

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