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We report the effect of annealing temperature on the dynamic and static magnetic properties of MgO/CoFeB/Ta/Ru multilayers. Angular resolved ferromagnetic resonance measurement results show that the as-deposited film exhibits in-plane magnetic anisotropy, whereas in the annealed films the magnetic easy-axis is almost along the direction perpendicular to the plane of the layers. The extracted interfacial anisotropy energy, $K_i$, is maximized at an annealing temperature 225 °C, in agreement with the vibrating sample magnetometry results. Although the magnetization is not fully out-of-plane, controlling the degree of the magnetization obliqueness may be advantageous for specific applications such as spin-transfer oscillators. © 2016 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/10.1063/1.4954809]

I. INTRODUCTION

MgO-based magnetic tunnel junctions (MTJs) are currently the structures of choice for magnetic random access memories (MRAMs), as they exhibit extremely large tunnel magnetoresistance (TMR) values due to highly effective spin-dependent tunneling.1,2 Initial studies focused on devices where both free and reference layers have remnant states in the plane of layers.3-5 For the devices with in-plane configuration, an applied external magnetic field was essential for stabilizing the microwave emission. Devices with in-plane reference layers and out-of-plane magnetized free layers are also useful in the framework of spin-transfer-oscillators (STOs).6,7 Recently, it has been shown that obliquely magnetized free layers may offer an easier path towards STO operation without the need for external magnetic field.8 On the other hand, it has been reported that devices with perpendicular magnetic anisotropy (PMA) offer a better trade-off between reducing the writing power and maintaining thermal stability sufficient for data retention in spin-transfer torque RAM.9,10

MgO/CoFeB/Ta trilayers are known to exhibit particularly high PMA.11 To achieve this, as well as high TMR ratios in MTJs, the multilayers are typically annealed post-deposition in order to obtain highly oriented (001) MgO. Upon annealing, the easy-axis changes from in-plane to out-of-plane for layers with thicknesses in a certain range. The PMA is believed to originate at the interfaces, either from hybridization of Fe and O orbitals12 or intermixing of CoFeB with adjacent capping/seed layers.13 It was shown that both annealing temperature and CoFeB thickness have a significant impact on the formation of the PMA in that system.13 Also, development of the PMA

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can yield an oblique magnetization and the degree of this obliqueness can be used to tune the performance of the STOs.\textsuperscript{14,15}

In this study, MgO/CoFeB/Ta trilayers are annealed at different temperatures. Formation of PMA was investigated by means of dynamic as well as static magnetization measurements. We find that, the perpendicular component of the magnetization can be tuned by changing the annealing conditions to obtain neither strong in-plane nor strong out-of-plane anisotropy.

II. EXPERIMENTAL

For this study, MgO (2nm) / Co\textsubscript{40}Fe\textsubscript{40}B\textsubscript{20} (1nm) / Ta (5nm) / Ru (5nm) multilayers were grown by RF and DC sputtering using a TIMARIS deposition tool (Singulus Technologies AG) on Si/SiO\textsubscript{2} wafers. 1 \times 1 \text{cm}\textsuperscript{2} samples were annealed in an Ar environment for 45 minutes at temperatures between 150\textdegree{} and 300\textdegree{}C. Room temperature angular dependent ferromagnetic resonance (FMR) measurements were performed using a cylindrical microwave cavity with a resonance frequency of 9.67 GHz. During the measurements, the angle $\theta$ between the surface normal and the applied magnetic field was swept from $-180$\textdegree{} to $+180$\textdegree{} in steps of 2\textdegree{}. Here, $\theta = 0$\textdegree{} corresponds to the magnetic field aligned along the normal, while 90\textdegree{} corresponds to the magnetic field in the plane of the sample film. The magnetic field was swept from 0 to 0.8 T. The derivative of the microwave absorption signal $\chi''$ was measured for the complex rf-susceptibility $\chi(\omega)$ as a function of applied dc field.\textsuperscript{16} Before and after measuring each sample, a spectrum of the empty cavity was taken as a reference. For samples where several anisotropies can co-exist, angular resolved FMR is an ideal investigation technique allowing for a precise determination of the easy axes at work in the system. Using FMR, we are able to separate the contribution of the interfacial magnetic anisotropy generated at the interface of MgO and CoFeB as a function of annealing temperature. Complimentary static magnetization measurements were performed using a SQUID-VSM (Quantum Design, MPMS 3) for both in- and out-of-plane measurement geometries at room temperature.

III. RESULTS AND DISCUSSION

Representative FMR spectra for the as-deposited film and the film annealed at 150\textdegree{}C are shown in Fig. 1. The upper and lower panels correspond to field applied in-plane and out-of-plane directions, respectively. Upon annealing, the CoFeB film starts to develop PMA, causing a shift in the resonance field. The set of resonance fields, as extracted from out-of-plane angular FMR measurements, are shown in Fig. 1(b) and 1(c). The spectra corresponding to the as-deposited film exhibit two resonance peaks, which are found after fitting the FMR data. Those peaks are found to have similar intensities with in-plane-like (black data points) and isotropic-like behavior (red data points)(Fig. 1(b)). We associate this double resonance behavior with the co-existence of domains/grains with in-plane and tilted magnetic anisotropies, respectively. For the sample annealed at 150\textdegree{}C (green curve), the resonance field in the out-of-plane direction is lower than in the in-plane direction, which is a clear manifestation of PMA in the structure. At 225\textdegree{}C, the resonance field in the out-of-plane direction shows its lowest value (light-blue curve), which is about 0.07 T. When increasing the annealing temperature further, the resonance field in the out-of-plane direction returns towards the isotropic value observed in the as-deposited state. This indicates the loss of PMA, which can be attributed to structural deformations (e.g., interdiffusion between the layers) and hence the change of the magnetic properties of the CoFeB layer.\textsuperscript{17}

The competing magnetic anisotropies in the system reduce the out-of-plane demagnetizing field from $\mu_0M_s$ to the effective magnetization ($M_{\text{eff}}$). For the magnetic films with negligible uniaxial in-plane magnetic anisotropy, if the resonance frequency is known, $M_{\text{eff}}$ can be calculated from the out-of-plane resonance field values using the Kittel formula (Eq. (1)).\textsuperscript{19}

$$\mu_0M_{\text{eff}} = \frac{\omega}{\gamma} - \mu_0H_{\text{Res.\perp}}$$  \hspace{1cm} (1)
FIG. 1. (a) The derivative of the microwave absorption signal $\chi''$ of the complex rf-susceptibility $\chi(\omega)$ as a function of applied dc magnetic field. The black and the red data represent the as-deposited and 150$^\circ$C annealed samples, respectively. FMR spectra measured in the direction parallel (upper) and perpendicular (lower) to the film plane. Resonance fields versus different angles close to the direction parallel to the plane of the films (b) and perpendicular to the plane (c). (d) Effective magnetization as a function of annealing temperature as calculated from the data shown in panel (c) using equation (1).

where $\mu_0$ is the permeability of free space, $\omega$ is the angular frequency of the resonance, and $\gamma = \frac{2\mu_B}{g\hbar}$ is the gyromagnetic ratio (28.02 GHz/T). The as-deposited sample exhibits a negative effective magnetization ($-2.68$ mT), indicating that the magnetization lies in the plane of the sample. Upon annealing, $M_{\text{eff}}$ turns positive, due to the development of PMA in the structure. The effective magnetization increases with annealing temperature reaching a maximum of 0.28 T at 225$^\circ$C. Higher annealing temperatures yield lower $M_{\text{eff}}$ values (see Fig. 1(d)).

VSM measurements were performed for a set of as-deposited and annealed samples with the same size, for the in-plane (see Fig. 2) and out-of-plane geometry. For a proper comparison, the diamagnetic contribution of the substrate was subtracted for each sample. We define the saturation

FIG. 2. (a) Room temperature in-plane magnetization curves for as-deposited samples and those annealed at different temperatures. The magnetization values shown were calculated without considering the presence of a magnetic dead layer. The inset shows a zoomed view of both out-of-plane and in-plane field dependent magnetization curves of the sample annealed at 225$^\circ$C. (b) Room temperature in-plane and out-of-plane saturation fields versus annealing temperature. The maximum separation is observed around 225$^\circ$C.
field as the extrapolation of the low field slope of the magnetization curve to the saturation magnetization value. The change in saturation field as a function of annealing temperature is shown in Fig. 2 for each sample. In agreement with the FMR data, the VSM measurements yield higher saturation fields for the in-plane measurements compared to out-of-plane measurements, confirming that the easy axis of the films lie along the film normal after they are annealed. The VSM data also confirms the existence of PMA in the annealed films and that the PMA reaches a maximum at 225 °C, which is in agreement with previously reported values for the similar systems.13 The increase up to 225 °C in $M_s$ is related to the diffusion of boron out of CoFeB towards either the MgO or Ta interface, whereas the decrease is related to the interface deformations.21 An additional effect which may lead to a drastic change in saturation magnetization of the CoFeB film is the formation of a magnetic dead layer (MDL). An MDL is not expected to develop at the MgO/CoFeB interface, the second interface (in our case Ta/CoFeB) has a dramatic impact on PMA, as well. Annealing of Ta/CoFeB/MgO structures at intermediate temperatures (between 200°C and 300°C) improves the crystalline quality, which boosts the PMA, as we show here. However, further increase in the annealing temperature leads intermixing of Ta into the CoFeB, which degrades the PMA significantly. The further improvement of the anisotropy can be further done by simply employing a buffer layer, which will help to achieve similar anisotropy constants at higher annealing temperatures.28

Due to lack of 100% remanence in our VSM curves, we can conclude that the magnetization at zero field points along a direction which is neither in-plane nor out-of-plane, i.e. the magnetization

TABLE I. ($M_s^*$) Saturation magnetization values as calculated without considering the presence of magnetic dead layer (MDL). (MDL***) MDL thicknesses according to Jang et al.18 for the as-deposited sample and the sample annealed at 150°C, for the rest of the samples it is extrapolated from their data. ($M_s^{***}$) Saturation magnetization as calculated taking into account the presence of the MDL. Measured saturation magnetization ($M_s$), effective magnetization ($M_{eff}$), out-of-plane demagnetizing field ($\mu_0 M_s$) and interface magnetic anisotropy ($K_i$) values for each annealing temperature.

| Annealing Temperature [°C] | $M_s^*$ [10^6 A/m] | MDL [nm] | $M_s^{***}$ [10^6 A/m] | $\mu_0 M_s$ [T] | $\mu_0 M_{eff}$ [T] | $K_i$ [10^3 J/m^2] |
|----------------------------|--------------------|----------|------------------------|----------------|-------------------|-----------------|
| as-deposited                | 0.456              | 0.36     | 0.712                  | 0.894          | -0.00268          | 0.203           |
| 150                        | 0.480              | 0.48     | 0.923                  | 1.160          | 0.124             | 0.308           |
| 200                        | 0.496              | 0.49     | 0.972                  | 1.222          | 0.238             | 0.362           |
| 225                        | 0.690              | 0.49     | 1.352                  | 1.700          | 0.280             | 0.683           |
| 250                        | 0.580              | 0.49     | 1.137                  | 1.430          | 0.188             | 0.469           |
| 300                        | 0.506              | 0.49     | 0.992                  | 1.246          | 0.196             | 0.365           |

In order to qualitatively examine the changes in the anisotropy induced by annealing, we separate the different contributions to $\mu_0 M_{eff}$ in Eq. (1) as:

$$\mu_0 M_{eff} = \mu_0 M_s \frac{2K_b}{M_s} \frac{2K_i}{M_s} \frac{1}{t},$$

where $M_s$ is the saturation magnetization after MDL correction (column $M_s^{***}$ in Table I) $K_b$ and $K_i$ are the bulk magnetocrystalline and interfacial magnetic anisotropy energies, respectively, and $t$ is the thickness of the magnetic layer.

In the MgO/CoFeB system, the bulk magnetic crystalline anisotropy $K_b$ has been shown to be negligible.22 We can therefore reduce Eq. (2) further to only include the last term which relates to $K_i$. We can then estimate the evolution of $K_i$ as a function of annealing temperature (see Fig. 3), without the need for typical ferromagnetic layer thickness dependence measurements.23 The maximum value of $K_i$ is reached at 225°C. This is also the same temperature where the saturation magnetization peaks (shown in the inset). Even after annealing at 225°C, $K_i$ is considerably lower than the maximum value predicted for Fe/MgO (3.6 mJ/m²).24 While the PMA occurs mostly due to the CoFeB/MgO interface, the second interface (in our case Ta/CoFeB) has a dramatic impact on PMA, as well. Annealing of Ta/CoFeB/MgO structures at intermediate temperatures (between 200°C and 300°C) improves the crystalline quality, which boosts the PMA, as we show here. However, further increase in the annealing temperature leads intermixing of Ta into the CoFeB, which degrades the PMA significantly. The further improvement of the anisotropy can be further done by simply employing a buffer layer, which will help to achieve similar anisotropy constants at higher annealing temperatures.28

Due to lack of 100% remanence in our VSM curves, we can conclude that the magnetization at zero field points along a direction which is neither in-plane nor out-of-plane, i.e. the magnetization...
FIG. 3. The dependence of interface anisotropy with respect to the annealing temperature including the presence of the MDL. A sharp peak is observed for the $225^\circ C$ annealed sample. The change in saturation magnetization with respect to the annealing temperature is shown in the inset.

points at an oblique angle. The lack of fully out-of-plane magnetic anisotropy can be attributed either to the thickness of the CoFeB layer or to the thickness of MgO. The CoFeB thickness for this study was comparable to those reported by Skowkonski et.al.$^{14}$ where tilted magnetic anisotropy of CoFeB layers were used to stabilize spin-torque oscillations. On the other hand, the PMA strictly depends on the interface between CoFeB and MgO. The MgO thickness has recently shown to be an important parameter to optimize the PMA.$^{29}$

IV. CONCLUSION

In summary, we have used angular resolved ferromagnetic resonance to optimize the annealing temperature of a simple MgO / CoFeB / Ta / Ru multilayer. Our main result is that both in-plane and out-of-plane anisotropies are present in the as-deposited films and that post-annealing systematically shifts the magnetic easy axis from the in-plane to the out-of-plane direction. According to our measurements, the interface anisotropy progressively increases with annealing temperature to a maximum of $K_i = 0.683 \text{ mJ/m}^2$ at $225^\circ C$. Our optimized annealing temperature corresponds to an oblique magnetization angle, neither fully in-plane nor fully out-of-plane. Such layers could be useful for the development of zero-field spin-torque oscillator devices where oblique angle free-layers show more preferable response than those with very strong PMA.

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