

Manifold role of a Ta adhesion layer in pressure-tuned Co/Pt multilayers

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In this study, we systematically compare the impact of a 1.5-nm-thin Ta adhesion layer on the structural and magnetic behavior of Co/Pt multilayers (MLs) sputter-deposited at a low and a high Ar gas pressure. Our focus is a comprehensive structural analysis, centered on real-space cross-sectional scanning transmission electron microscopy (STEM) images, which are then correlated with complementary, reciprocal-space measurements from x-ray reflectometry (XRR) and x-ray diffraction (XRD). Drawing on this comparison and supplemented by atomic force microscopy (AFM), we develop a simplified model to fit the XRR data. Fits and data agree excellently, while the extracted density profiles match well with the STEM data. The structural features are then correlated with the magnetic ones, as determined by magnetic reversal loops and magnetic force microscopy (MFM) images. We find considerably better texture with the adhesion layer, as well as vastly different structural features at the higher deposition pressure between the different underlayers. At the lower pressure, the influence of the underlayer is much less pronounced. Magnetically, the Ta layer results in a marked improvement in the effective perpendicular magnetic anisotropy (PMA), increasing it up to an impressive 340% at the higher pressure, while remaining comparable at the lower one. We attribute the changes in structure and magnetism to two different factors: (1) The adhesion layer stops the implantation of Pt atoms into the substrate, improving layer smoothness and texture. (2) The momentum transfer to the film is reduced at higher sputter pressures, which reduces intermixing between Co and Pt, but increases overall roughness. Combining both, the roughness remains small enough to enable well-defined grains with sharper Co/Pt interfaces and lower intragrain curvature, resulting in the increased PMA. Our study provides a practical approach for controlling anisotropy in sputter-deposited ML systems containing heavy atom species, utilizing different combinations of adhesion layer and sputter pressure.

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I. INTRODUCTION

In the field of perpendicular magnetic anisotropy (PMA) material research, Co/Pt multilayers (MLs) excel with their high PMA values (10^5 – 10^6 J/m³) [1–6]. The ease of fabrication through industry-standard methods like sputter deposition [7] in a non-epitaxial process allows the use of non-crystalline substrates like oxidized silicon wafers or glass [2,7]. The magnetic properties of Co/Pt MLs are easily tunable by, for example, adjusting the individual layer thicknesses [6,8–10], the number of repeats [11,12], the temperature during and after deposition [1,6,8,13], different underlayers [13–16], and the type and pressure of the sputter gas [9,17–22].

The deposition pressure in particular can be used to tune the microstructure of the systems [18–24], since it changes the

energy and momentum of the energetic particles arriving at the substrate [23–25]. These include so-called neutrals, which are neutralized sputter gas ions reflected from the target and sputtered atoms ejected from it. Depending on the atomic mass of the target and sputter gas, the initial energy per particle is in the range of a few eV to 10s of eV for the target atoms and 10s to 100s of eV for the neutrals [17,23,26,27]. The outgoing particles scatter with the sputter gas, changing their energy and direction, which is governed by their initial energy, scattering cross section, and mass ratio to the sputter gas [23,28]. As the sputter pressure increases, so does the number of collisions of the energetic species with the sputter gas, thus reducing the mean-free-path of the particles and the distance needed to thermalize them [28]. Depending on the deposition conditions, structural changes of the sputtered films occur. They can then be attributed to a *normalized momentum*, introduced by Windischmann [23,29], which depends on the number (flux) and momentum of particles arriving on the film surface per deposited target atom.

Depending on the microstructure, a wide range of magnetic reversal mechanisms can be observed and purposely dialed in. For room-temperature depositions, a lower sputter deposition pressure results in films that are dominated by

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lateral exchange with full film reversal, which is useful, e.g., in magnetic tunnel junctions [16] and necessary for highly periodic stripe or bubble domain formation [10,12,19,30,31], as well as unhindered collective reversal in synthetic antiferromagnets [12,32,33]. At higher pressures, the films become more granular with laterally reduced exchange coupling at the grain boundaries, resulting in domain pinning and more grain-wise reversal of the magnetization [19,20,34], which has been investigated for perpendicular magnetic recording media applications [9,21,35,36].

Furthermore, Co/Pt MLs are often grown on an underlayer, templating a face-centered cubic FCC(111) texture in the ML, which is reportedly beneficial for the PMA of the system [3,14,37]. Pt is a common seed material, but other seeds like Cu and Ru are also in use for FCC and hexagonal close-packed (HCP) texturing [6,12,16,18,38]. However, the adhesion of these metal films to the usually oxidized substrate (e.g., SiO₂) might be insufficient [39], which can be mitigated by the use of an additional adhesion layer between the substrate and seed layer. Ta, Ti, and Cr are commonly used adhesion layer materials [39]. For Co/Pt MLs in particular, Ta is often used [6,10,11,15,16,31,33,40–42]. The impact of this adhesion layer on the magnetic and structural properties is, however, often neglected, and systems with and without adhesion layers are used interchangeably, especially when comparing older publications (without Ta) with newer ones (with Ta).

As our study shows, this is not always a valid assumption. Using two pressures—3 mTorr and 10 mTorr, representing a low, respectively high sputter gas pressure—we will highlight the impact of the presence or absence of a 1.5-nm-thin Ta layer below the Pt seed on the structure and magnetism of a Co/Pt ML system. We find a very different substrate-film interaction, depending on the underlayer, related to the normalized momentum caused by the deposition conditions. Based on the resulting initial film morphology, the film structure evolves very differently. Particularly at higher pressures, we observe a massive increase in anisotropy with a Ta adhesion layer, which we attribute to lower intermixing and flatter interfaces caused by a reduced normalized momentum, combined with low cumulative roughness as a result of a compact and dense seed growth.

Our paper is structured in the following way. In Sec. II, we detail our material system and describe our measurement procedures. Section III contains our structural and magnetic measurements, including a discussion and interpretation of the results. We then combine structure and magnetism in Sec. IV with the deposition conditions and underlayer choice to discuss the origin of the observed trends. We focus on the impact of the kinetics during the sputter process and the consequences on the PMA as a performance metric, in particular. Finally, Sec. V concludes and summarizes our study.

The results section is split into four parts. We begin Sec. III A with the characterization of the local real-space structure of the films obtained by cross-sectional scanning transmission electron microscopy (STEM) and surface-sensitive atomic force microscopy (AFM), which is complemented in Sec. III B with the discussion of the layer structure characterized using reciprocal-space x-ray

reflectometry (XRR) measurements. In this section, we will focus on developing a simplified model to fit the XRR data based on the real-space STEM and AFM images, which might aid in future analysis of similar systems. We conclude the structure characterization in Sec. III C with an evaluation of the crystal structure as determined by x-ray diffraction (XRD) and rocking curve measurements to complement the local evaluation from the STEM images. We then relate in Sec. III D the observed structural changes to the magnetic properties determined from out-of-plane (OOP) and in-plane (IP) magnetic hysteresis loops measured with superconducting quantum interference device vibrating sample magnetometry (SQUID-VSM). These findings are complemented by magnetic force microscopy (MFM) measurements of remanent magnetic states after OOP and IP demagnetization.

II. MATERIALS AND METHODS

The deposition of all samples was performed in an AJA International, Inc. ATC 2200-V sputter-up system with 2-inch targets. The Co and Pt guns are angled in a confocal configuration, whereas the Ta gun is placed in the center of the chamber and operated in a parallel configuration, with a target-sample distance of approximately 20 cm. During deposition, the sample plate was rotated at a constant speed of 75 rotations per minute. The base pressure was below 5×10^{-8} Torr, and depositions were carried out at room temperature. The Ar sputter gas with a purity of 99.9999% (6N) was injected at a constant rate of 25 standard cubic centimeters per second.

The samples were fabricated with direct current (DC) magnetron sputtering on thermally oxidized Si(100) wafer pieces of 14 mm $\times$ 14 mm in size with an oxide thickness of approximately 100 nm. A second batch of nominally identical samples was fabricated later on a natively oxidized SiO₂ wafer for TEM analysis, where the oxide thickness was around 2 nm. A 20-nm Pt seed was used to induce an FCC(111) texture in the ML. The ML system consists of 50 repetitions of Co 0.4 nm and Pt 0.7 nm to be comparable with earlier studies [19,20]. The large number of repetitions enables better analysis of cumulative growth effects [43] and also reduces the impact of growth-induced variations at the seed and cap-layer interface on the overall magnetic response of the sample [16,44]. The samples were capped with 2.3-nm Pt to prevent oxidation.

Ta, Pt, and Co were sputtered with a constant DC power of 50 W, 60 W, and 80 W, respectively. This resulted in deposition rates of approximately 0.4 Å/s for Ta, 0.9 Å/s for Pt at both pressures, 0.4 Å/s for Co at 3 mTorr and 0.2 Å/s for Co at 10 mTorr. We note that increasing the pressure from 3 mTorr to 10 mTorr reduced the sputter voltages by about 12% for Co and 4% for Pt, which is a known factor that contributes to microstructural changes [25]. Seed, ML, and cap were all deposited at the same Ar pressure, 3 mTorr representing low and 10 mTorr representing high sputter-deposition pressures. The Ta layer was consistently deposited at 3 mTorr, since we want to study the effect of the presence or absence of the Ta layer, and not a change of the Ta layer itself. In the following, the underlayer is either adhesion/seed or seed only for the samples

with and without Ta, respectively. This results in the following nominal sample stack: Si/SiO₂(100 or 2 nm)/Ta(0 nm or 1.5 nm)/Pt(20 nm)/[Co(0.4 nm)/Pt(0.7 nm)]₅₀/Pt(2.3 nm).

Bright-field (BF) and high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) imaging at various magnifications were performed with a Talos F200X (FEI) transmission electron microscope operated at an accelerating voltage of 200 kV. Prior to STEM analysis, each specimen mounted in a double-tilt low-background holder was placed for 8 s into a Model 1020 Plasma Cleaner (Fischione) to remove potential contamination. Cross-sectional TEM lamellae preparation was performed by *in situ* lift-out using a Helios 5 CX focused ion beam (FIB) device (Thermo Fisher). To protect the sample cross-section, a carbon cap layer was deposited beginning with electron-beam-assisted and then followed by Ga-FIB-assisted precursor decomposition. Subsequently, the TEM lamella was prepared using a 30-keV Ga-FIB with adapted currents. Its transfer to a three-post copper lift-out grid (Omniprobe) was performed with an EasyLift EX nanomanipulator (Thermo Fisher). To minimize sidewall damage, Ga ions with only 5-keV energy were used for final thinning of the TEM lamella to electron transparency. While the BF-STEM images show a combination of mass-thickness and diffraction contrast, the HAADF-STEM micrographs of the crystalline samples predominantly display atomic-number contrast with residual diffraction contrast.

All x-ray measurements were carried out using a Rigaku SmartLab diffractometer with a 9-kW rotating anode and Cu K_α radiation in a parallel beam configuration on the full-size samples. XRD and rocking scans were conducted using a two-bounce Ge(220) monochromator, selecting the Cu K_{α1} radiation and eliminating the impact of other wavelengths. XRR was measured with a Cu K_β filter to reduce the impact of the Cu K_β line while maintaining a much higher intensity than the monochromator configuration.

XRR curves were fitted using GenX 3 [45] version 3.6.27. The “spec_inhom” model was employed utilizing the Parratt algorithm [46]. Sample inhomogeneities were tested but found not to be necessary for modeling purposes. A Gaussian beam footprint correction was applied to fit the total reflection of the curves. The instrument resolution was fitted using the “fast conv” resolution type. The model will be explained later in the text. 30 fits of the same model and sample with different starting conditions were applied to ensure consistent scattering length density (SLD) profiles.

AFM and MFM were carried out on the full-size samples with a Bruker Dimension Icon AFM, using PPP-LM-MFMR magnetic tips from NANOSensors for the MFM images and a super-sharp TESP-SS tip from Bruker for the AFM images. MFM was performed in dual-pass lift mode. The MFM lift height was 30 nm for IP and 15 nm for OOP demagnetized samples. OOP and IP demagnetizations were performed with an electromagnet built into a Microsense EZ7 vector-VSM. The corresponding routine reduced the field from 1.8 T to 2 mT, alternating between positive and negative maximum fields. Per cycle, the maximum field amplitude was reduced to 96% of the previous maximum field.

Magnetic measurements were performed with a Quantum Design MPMS3 SQUID-VSM. Pieces were cut from the cen-

ter of each sample to approximately 4×4 mm² in order to fit within the SQUID-VSM sample chamber. For the OOP measurements, the cut sample pieces were placed in a plastic straw, clamped between two other plastic inserts to hold the sample in place. For the IP measurements, the samples were glued to a quartz holder using GE varnish. A linear background was subtracted from the magnetization curves to obtain a constant saturation. Effective anisotropy values were calculated using the area between the normalized OOP and IP curves, multiplied by the saturation magnetization. To account for the hysteresis, the absolute areas towards the *x* axis of forward and reverse curves were averaged and then divided by 2 to obtain the area of just one quadrant of the loop [47,48]. The saturation moment was obtained from saturation of the reversal loops, corrected by a sample-shape-dependent factor [49], then averaged between the IP and OOP values. The magnetic volume for the magnetization was determined by the sample area times the magnetic thickness. The magnetic thickness was assumed to be the number of ML repetitions times the bilayer thickness, including the Co and the polarized Pt of the ML [40,41,50,51].

III. RESULTS

A. Local real space imaging: Cross-section (TEM) and surface (AFM)

We start the structural characterization by analyzing the real-space structure using STEM images of the samples deposited on native SiO₂. Our analysis focuses on 100×100 nm² wide bright-field (BF-)STEM images in Fig. 1 and 780-nm-wide high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images in Fig. S1 within the Supplemental Material (SM) [52]. The insets in Fig. 1 are fast Fourier transforms (FFT) of the STEM images on the same intensity scale, limited to a range of ±5 nm⁻¹, representing a local perspective on the crystal structure. The major features in the FFT are labeled exemplarily in Fig. 1(c).

The film interface to the substrate is very different depending on the underlayer. With Ta, the transition from crystalline Si to amorphous SiO₂/Ta and crystalline Pt is smooth and continuous. In contrast, the samples without Ta show a darker layer between the Si and the SiO₂, which is thicker and more pronounced at 3 mTorr. The contrast suggests that this layer might be a platinum silicide, which was formed by contact with the Si wafer after Pt penetrated the native oxide layer. At 3 mTorr, the SiO₂ interface is smeared out on the upper and lower sides and even shows pinhole-like spots, under which the layer is thicker. This is not as pronounced at 10 mTorr, leading to a similar rough, but sharper interface. It is very likely that the formation of this layer considerably disturbs the crystal structure of the seed layer, giving rise to a much lower texture for the films without Ta.

The presence or absence of the implantation layer without and with Ta can be related to the adhesion of Pt and Ta to Si and SiO₂. Ta is known to bind strongly to SiO₂ [53], while Pt does not [39]. Additionally, Pt forms silicides with bare Si, but not with SiO₂ [39,54]. Despite the similar mass of Ta and Pt, the strong bonding of Ta to SiO₂ strongly suppresses

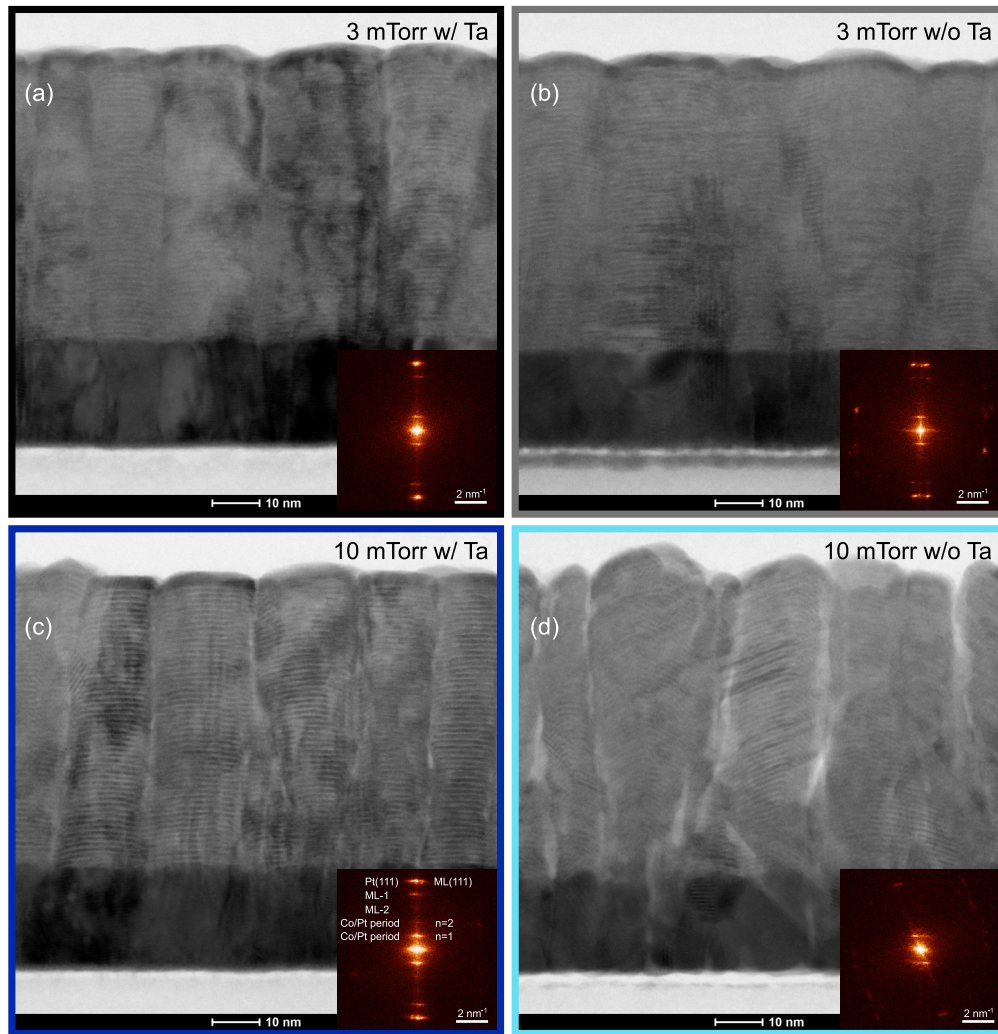


FIG. 1. Cross-sectional BF-STEM images of the samples. The insets are the corresponding FFT with a range of $\pm 5 \text{ nm}^{-1}$. (a) and (b) show the 3-mTorr samples and (c) and (d) the 10-mTorr samples, with and without Ta, respectively.

penetration through the SiO_2 layer caused by energetic bombardment. Instead, the formation of a chemical bond is facilitated. The thin Ta layer then blocks the following Pt from penetrating the SiO_2 . Without Ta, the Pt is pushed into and through the SiO_2 , as it does not bind with it. Instead, it reacts with the bare Si below to form a silicide.

The formation of PtSi during the sputter process was reported previously [54,55]. Shi *et al.* [54] in particular investigated the formation of platinum silicide for Pt films deposited at 200°C on etched Si substrates. They also presented STEM images, which are similar to our sample as shown in Fig. 1(b). They also found a thin film between Pt with darker and PtSi with lighter contrast, which contains oxygen according to x-ray photoelectron spectroscopy (XPS). They propose that this layer is formed after the deposition through diffusion of oxygen along the grain boundaries towards the Pt/PtSi interface. In our case, it is most likely the native SiO_2 layer, as its thickness is consistent between all samples, independent of the microstructure.

The Pt seed layers for the samples with Ta have a similar structure for both pressures: dense with a pronounced grain structure and overlaid contrast from structural defects. The

wide-range HAADF-STEM images in Fig. S1 within the SM [52] confirm this over a larger region, but further indicate a slightly higher amount of voids at the seed/ML interface at 10 mTorr. Without Ta, the seed at 3 mTorr is similar but with a less pronounced structural contrast due to the film's lower texture. At 10 mTorr, however, the seed is considerably rougher, showing voids especially at the interfaces, some of them penetrating through the full thickness.

As expected, the structure templated by the seed propagates into the ML, forming columnar grains. The grain boundaries get more pronounced with increasing thickness. Even a separation of the grains occurs at 10 mTorr, which is well visible in Figs. S1(c) and S1(d) within the SM [52]. Taking these HAADF-STEM images and the corresponding BF-STEM micrographs as a basis, the lateral grain size is around 19 nm at 3 mTorr and around 15 nm at 10 mTorr, independent of the underlayer. There are, however, many small grains as thin as 5-nm in size. The grains are also increasingly irregular at 10 mTorr, showing not only columnar, but also V-shaped and inverted V-shaped behavior, resulting in a broader grain-size and shape distribution and a lower vertical coherence of the ML structure.

However, this behavior is much more pronounced in the absence of Ta.

The layering of Co and Pt in the ML is vastly different between the samples. At 3 mTorr, the contrast between the layers is low and they show a curved, dome-like shape. At 10 mTorr with Ta, the contrast is considerably better and the grains are less curved at the interfaces. Conversely, at 10 mTorr without Ta, the grains exhibit a considerable amount of disorder, including large voids and irregular grains. Furthermore, the layering is only partially present, with the normal of the Co/Pt interfaces tilted in a wide angular range, which can also be seen from the FFT inset in Fig. 1(d).

The capping layer on top of the ML is well visible for all samples. Its surface follows the shape of the grains and enables their differentiation, even when no distinct grain boundaries are visible. The cap forms a compact and closed layer, which should serve its purpose in preventing oxidation of the Co layers. However, the 10-mTorr sample without Ta has pronounced voids between some grains, which could act as a pathway for oxygen. This could be one reason for the reduced saturation magnetization in this sample, as we will describe later in Sec. III D. Note that some grains overlap through the thickness of the cross-section, which is particularly well visible at the cap, where it causes different shadings of gray.

The local crystal structure of the samples is highlighted by the FFT insets. The features are assigned in Fig. 1(c). Visible are the FCC(111) peaks of Pt(111) from the seed layer and the ML laminate [ML(111)]. The difference between Pt(111) and ML(111) cannot be fully resolved in this FFT, but is well discernible in the XRD scans, which we will discuss in detail in Sec. III C. The ML(111) Bragg peak corresponds to an effective lattice spacing between Pt and Co caused by the dense laminate of both materials within the ML stack. Neither element can reach its bulk lattice constant because of the low individual layer-thickness, leading to the observed laminate peak. We can also identify two satellite peaks (ML-1 and ML-2), corresponding to the Co/Pt bilayer periodicity of the crystal structure, as well as XRR superstructure peaks (Co/Pt period $n = 1, 2$), corresponding to the periodicity of the chemical contrast between the layers.

With Ta, both samples show a pronounced FCC(111) fiber texture, with a majority of the lattice planes pointing in the OOP direction. The intensity profile along the vertical axis is consistent with the XRD scans presented later. XRD satellite and XRR superstructure peaks can be identified in the first and second orders. Second-order peaks are only visible for the 10 mTorr sample with Ta because of the low intensity of the peaks. With Ta, the angular distribution of the FCC(111) texture is slightly wider at 10 mTorr than at 3 mTorr. Upon closer inspection of the samples with Ta in Figs. 1(a) and 1(c), it is apparent that the crystal structure and chemical contrast do not follow each other. The lattice planes within one grain are parallel and point in a singular direction, while the chemical contrast of Co and Pt follows a certain curvature, consistent with previous studies [32,56]. The fiber texture is less pronounced for the samples without Ta. At 3 mTorr, pronounced FCC(111) spots at different angles are visible, which form almost a ring at 10 mTorr—a sign of a polycrystalline structure.

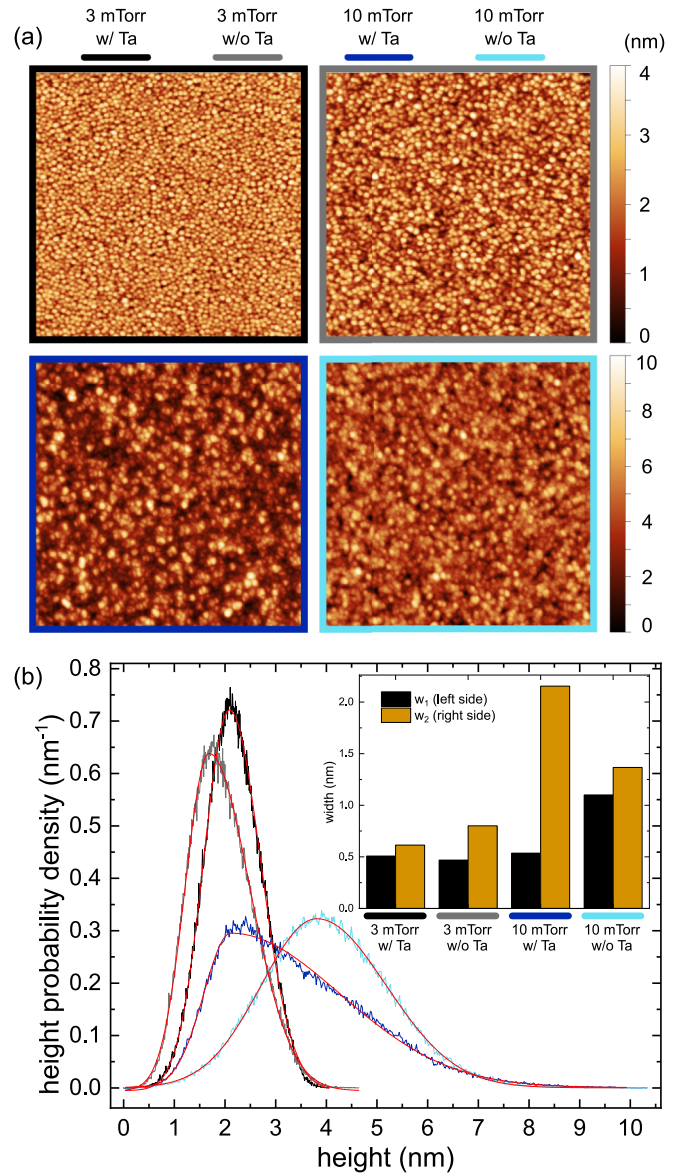


FIG. 2. AFM images with a scan area of $(1.0 \times 1.0) \mu\text{m}^2$ are presented in (a). The color scales are the same for each pressure. Height probability density functions are shown in (b), with the red curves indicating a split-Gaussian fit. The widths of the left- and right-hand sides are shown in the insets. The split-Gaussian fit is given by $y = y_0 + H \cdot \exp(-\frac{(x-x_c)^2}{2(w_{1,2})^2})$, with the y -axis shift y_0 , the peak height H , the center of the Gaussian x_c , and the left and right side widths of the Gaussian $w_{(1,2)}$ [w_1 for $(x < x_c)$ and w_2 for $(x \geq x_c)$].

We now switch focus to the surface of these samples as measured by AFM. Figure 2(a) displays $(1.0 \times 1.0) \mu\text{m}^2$ AFM scans of the samples on native SiO_2 . See also the larger $(5 \times 5) \mu\text{m}^2$ images in Fig. S2 within the SM [52], which were measured over the same area as the IP demagnetized MFM images shown later in Sec. III D. To better understand the roughness of the film, the height distribution of the AFM images is plotted in Fig. 2(b), with split Gaussian fits and the widths of the left- and right-hand sides in the insets. The latter was done to account for asymmetries in the height distribution.

All samples exhibit a pronounced fine-granular structure, with a clear difference in the surface structure between 3 mTorr and 10 mTorr. At 3 mTorr, the film is smoother, with a more even distribution of grains for the sample with Ta. At 10 mTorr, the roughness is higher and a pronounced asymmetry to larger grains is visible, especially with Ta. Adding Ta reduces the RMS roughness from 0.62 nm to 0.54 nm at 3 mTorr and slightly increases it from 1.25 nm to 1.47 nm at 10 mTorr. The larger (5×5) μm^2 images (see Fig. S2 within the SM [52]) confirm these trends, despite the lower resolution of the MFM tip. Since the granular structure is well visible for all samples, we can perform a grain size analysis. The results agree well with the lateral grain size evaluation based on the STEM images, showing no significant difference in size between the underlayer, but a small reduction from 18 nm to 15 nm when increasing the pressure. The size of the grains is evenly distributed at 3 mTorr, but there are more small grains than large ones at 10 mTorr.

While all height distributions in Fig. 2(b) have an asymmetry to larger heights, particularly the 10-mTorr sample with Ta is very asymmetric, with a pronounced tail towards larger heights while having a narrow distribution at smaller heights. At 10 mTorr without Ta, the distribution is again more even, but overall broader. The strong asymmetry in the height distribution can be explained by several protruding grains over a more level background. In the case of the 10 mTorr sample with Ta, a mixture of relatively flat sections and large, protruding grains can be used to describe the surface, which is also in agreement with the wide-range STEM images in Fig. S1(c) within the SM [52]. This also means that at 10 mTorr, the RMS value is not the best way to describe the roughness at the surface of these samples. Lastly, we note that the AFM image (and therefore height distribution) is influenced by the finite image resolution (512×512 pixels), as well as the dimensions and opening angle at the tip apex, leading to a broadening of too narrow or steep parts of the topography.

B. Global reciprocal-space sample cross-section

Complementing the local real-space structure analysis, we used XRR presented in Fig. 3(a) to get a depth profile for the samples in Fig. 3(b). Here, we focus especially on getting reliable information about the densities of the layers and the transitions between them by developing a model to fit the XRR data. Since XRR averages over a wide area of the sample, its results better represent the overall structure than local measurements. Furthermore, it is much easier and faster to prepare and measure the samples for XRR than for STEM.

XRR scans for the samples with and without Ta adhesion at 3 mTorr and 10 mTorr are depicted in Fig. 3(a). The superstructure/ML peak originating from the periodicity of the ML is visible at around 8° . Narrow oscillations correspond to the total film thickness and the wider oscillations (e.g., at angles below the superstructure peak) are caused by the 20-nm Pt seed layer thickness. The angle of total reflection is near 1° , followed by a strong reduction in intensity caused by the surface roughness of the samples and the decrease with $1/q^4$ following from Fresnel equations, where $q \propto \sin(\theta)$ is the scattering wave number.

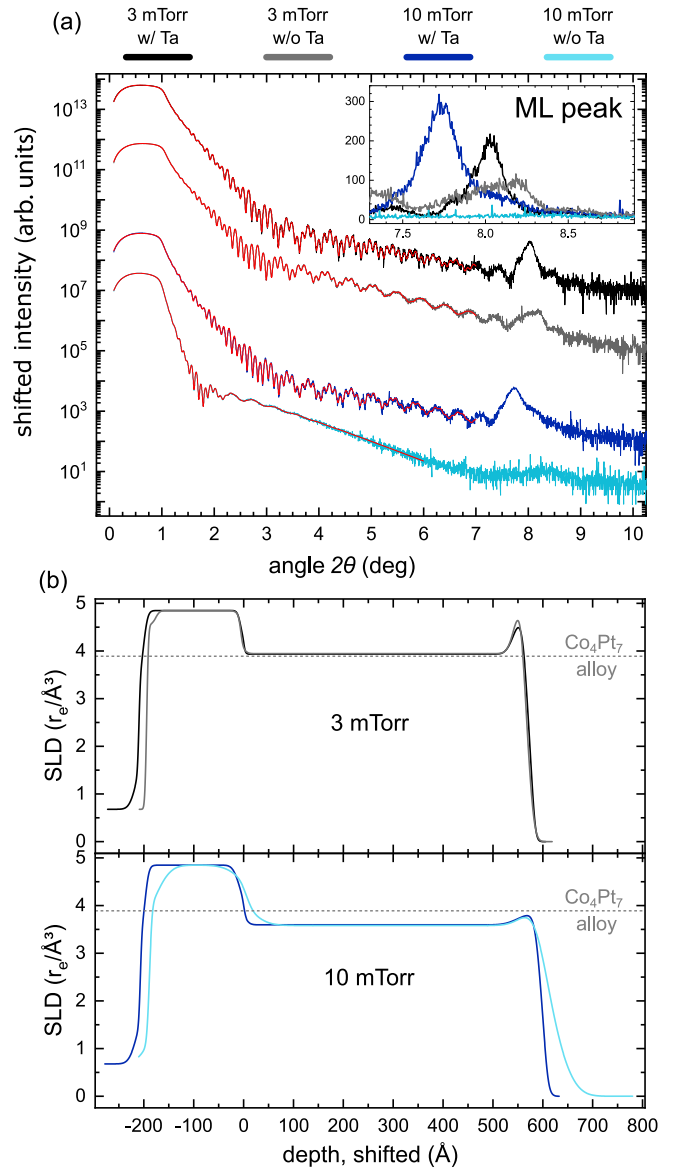


FIG. 3. In (a), XRR scans for the same samples are displayed on a semi-log scale alongside the best fits (red), with intensities shifted for better visibility. The inset highlights the superstructure peaks on a linear scale. Scattering length density (SLD) profiles derived from the fits are depicted in (b), where the dashed line represents the SLD of a Co_4Pt_7 alloy.

The best fits of the XRR scans in Fig. 3(a) are highlighted in red, overlaying the measurements. The corresponding scattering length density (SLD) profiles are presented in Fig. 3(b). The SLDs of 3 mTorr and 10 mTorr samples are split between top and bottom for better clarity, with the depth scale shifted such that 0 corresponds to the seed/ML interface. The SLD is proportional to the electron density and represents a laterally averaged depth profile of the sample. Since phase information is lost during measurement, the SLD profile cannot be recovered directly from the scans but must be modeled. From this modeled SLD, an XRR curve is simulated and fitted to the measured signal by refining the model [57]. However, simulating all samples with a comparable model is not

straightforward, given the very different structures resulting from the underlayer and sputter pressure. For example, Pierce *et al.* [20] introduced a complex model for comparable Co/Pt ML systems, in which the ML is split into five stacks, with CoPt alloy layers separating Co and Pt. This model fits the measurements reasonably well, but there were still major deviations visible, which impact the reliability of the density values obtained by the fits. We therefore chose a different approach to model our samples, reducing the number of parameters and achieving an excellent agreement between fit and data with the help of the STEM images in Fig. 1 and Fig. S1 within the SM [52]. Our simplified model is based on the following two approximations:

Firstly, the periodicity of the ML for the given system is only visible at and around the superstructure peak. This can be used to simplify the model. The 50 repetitions of the Co/Pt ML are approximated using a Co_4Pt_7 alloy with a 4:7 Co:Pt mass ratio, proportionally to the individual layer thicknesses in the ML. This alloy matches the average density of the ML but excludes the depth-dependent density oscillations caused by the layering. To exclude the ML, we limit the fitting range to a maximum of 7° for all but the 10 mTorr sample without Ta, which is only fitted up to 6° because of the very broad ML peak. The transition of seed/ML and ML/cap is also much easier to model with a CoPt alloy since the roughness of these interfaces does not overlap with the very thin, alternating Co and Pt layers. We also attempted to incorporate the layered structure of the ML, similar to Pierce *et al.* [20], but only got a poor agreement between fit and measurement, especially around the superstructure peak. This is expected from the complex granular structure of the ML with its three-dimensionally curved and tilted Co/Pt interfaces, as seen in the cross-sectional STEM images in Fig. 1.

Secondly, to better approximate the real density transition, additional transition layers (TLs) are added at the interface of substrate/underlayer, seed/ML and cap/air, respectively. The Névot–Croce factor is often used to describe roughness between two layers, for which the transition takes the form of an error function [58]. However, the real transition can follow a different shape (or function), for instance, because of a three-dimensional microstructure. The TLs better approximate the interface, but introduce further ambiguity in the fitting parameters. We identify two main contributions to such ambiguity: (1) The transition has another effective shape, similar to the asymmetric AFM height distributions in Fig. 2(b). RMS, and therefore the fitted roughness, is no longer a good quantity to describe the interface. (2) Different combinations of roughness, thickness and density can produce similar SLD transitions, adding further ambiguity to the parameter. Furthermore, thickness values may also reflect contributions from adjacent layers, making the precise determination of thicknesses challenging. Note that these ambiguities apply only to the parameters, not the SLD itself, as the fitting process uses the XRR curve simulated directly from the SLD. Alternatively, a graded interface approach [58] could be used. There, a transition is approximated by slicing it into many small layers, whose densities can follow any arbitrary function. However, this requires knowledge of the precise shape of the transition and an appropriate definition of roughness and thickness. It also increases the computational

complexity considerably, because of the increased number of interfaces. Both approaches should result in comparable SLD profiles. We chose the TL method because of its simpler implementation and faster evaluation speed. We will limit our analysis to the direct comparison of the SLDs, to avoid the ambiguity of the fitting parameters.

Especially for the cap/air transition, we can motivate the TL with the help of the STEM and AFM images. The derivative of the SLD is an approximation of the height distribution of the AFM scans displayed in Fig. 2(b). In the ideal case, an SLD transition shaped like an error function would result in a corresponding Gaussian height distribution in AFM. As discussed above, especially at 10 mTorr, the distribution is distinctly asymmetric, yielding a deviation from the idealized transition. Additionally, laterally averaging over the dome-shaped grains, which are seen in the STEM images, changes the transition as well, even for the 3 mTorr samples with their symmetric height distributions. A qualitatively similar microstructure can also be expected from the seed/ML interface, although roughness effects at the surface are more pronounced because of cumulative roughness effects and grain boundary development. The substrate/underlayer interface can also necessitate the transition layer to represent the implantation layers in Fig. 1. Other possible TLs were tested, but did not result in a significant improvement of the fits. Simpler models without the transition layers failed to fit all features in the XRR scans properly, leading to inconsistent SLD profiles between different fits of the same sample and model.

All these considerations result in the following two models for the respective underlayers:

- (i) without Ta: substrate(SiO_2)/**substrate-seed TL**/Pt_{seed}/Pt-ML TL/CoPt alloy/Pt_{cap}/cap-air TL,
- (ii) with Ta: substrate(SiO_2)/**Ta₂O₅/Ta**/Pt_{seed}/Pt-ML TL/CoPt alloy/Pt_{cap}/cap-air TL.

SiO_2 was chosen as substrate, since the 100-nm thermally oxidized SiO_2 film on top of the Si substrate is effectively not visible in the scans because of its thickness. Ta_2O_5 with a mass density of 8.2 g/cm^3 was chosen as a transition layer to the substrate for the samples with Ta. Previous studies suggest the formation of TaO_x or TaSi_x layers with different stoichiometries at the interface between SiO_2 and Ta for sputter-deposited films [53,59,60]. Because of the small thickness of these films and their low impact on the XRR curve, a single layer of Ta_2O_5 with a mass density of 8.2 g/cm^3 was determined to best fit the substrate/Ta TL. For the samples without a Ta adhesion layer, an implantation layer with a lower density than bulk Pt is added as the substrate/seed TL to account for the Pt implantation in the SiO_2 . The densities of SiO_2 , Ta_2O_5 , Ta, and Pt_{seed} were fixed to their bulk values. All other densities were fitted, as well as the thickness and roughness for each layer. Using these models, we can reliably and reproducibly fit even small details of the XRR curves (except for the superstructure peak), even with the significant simplification of modeling the ML as a CoPt alloy.

The SLD profiles obtained from the fit are presented in Fig. 3(b), with the depth profile aligned to the Pt seed surface for easier comparison. A dashed line indicates the SLD value of a Co_4Pt_7 alloy. The 1.5-nm Ta layer appears as a small shoulder between the substrate and the Pt seed, creating an offset between the profiles of the samples with and without Ta.

In the samples without Ta, the implantation layer is evident by the distinctly lower density at the bottom as compared to the rest of the Pt seed.

At 3 mTorr, the SLDs of the two samples almost coincide, showing nearly identical thicknesses with only minor differences at the transition between the layers. There is, however, a small reduction in roughness visible with Ta when observing the dampening of the narrow total thickness oscillations in the XRR curves [61]. While with Ta, this oscillation is visible up to 7° , it is already damped out at around 5° without Ta. Additionally, the AFM measurements confirm a slightly reduced roughness with Ta. This difference is small enough to be barely visible in the SLDs. The ML densities for both samples are identical and very close to the expected value of the Co_4Pt_7 alloy, supporting our simplified model.

At 10 mTorr, the difference between the samples becomes more pronounced. The seed/ML and cap/air interfaces are considerably smoother with Ta, which is also apparent qualitatively from the XRR curve and in agreement with the TEM images in Fig. S1 within the SM [52]. Because of the non-Gaussian-shaped height distribution for the sample with Ta, the AFM RMS values don't follow the trend, overestimating the roughness of this sample. It seems that in this case, the XRR is less sensitive to the higher grains protruding from the much smoother rest of the film. In general, a disagreement between XRR and AFM roughness values is known from literature and can be attributed to the large difference in investigated area and the non-Gaussian height distribution [62].

The ML densities are again comparable for both 10 mTorr samples, but now lie well below the Co_4Pt_7 density. Similarly, the density for the cap layer is much lower and the ML/cap transition is broader than for the 3 mTorr samples. This lower density can be attributed to more pronounced grain boundaries and a higher amount of voids in between the grains, as is known from the literature [20,21,43] and shown by the STEM images in Fig. S1 within the SM [52]. The cap is broad with a much lower density compared to the 3 mTorr samples. The grain boundaries and voids in the ML, its higher roughness, the different height of the grains and the thinness of the cap layer all contribute to the fitted SLD profile at the surface of the samples.

The densities of the Pt seed layer were chosen to be constant at the Pt bulk value in the models, given the small amount of voids in the seed layers for all samples, especially relative to the ML, as is apparent from the wide-range STEM images in Fig. S1 within the SM [52]. Given the good agreement of fits and data, this seems to be a good approximation of the real structure.

Additionally, we can directly compare the real cross-section profile with the SLDs. Using the wide-range HAADF-STEM images of Fig. S1 within the SM [52], we extract laterally averaged profiles and normalize the intensities between bulk Si and the maximum of the Pt seed. Fig. S3(a) within the SM [52] displays the resulting STEM profiles and Fig. S3(b) presents the SLDs on the same depth scale. A linear background, resulting from a slight increase of the TEM lamella thickness towards the Si substrate, is superimposed on the intensity of the sample. Independent of that, we see the same pressure dependence of the ML density relative to the seed. The seed/ML and cap/air interface also follow similar

roughness trends, although the cap layer is not as pronounced for the STEM profiles. The differences in thickness between 3 and 10 mTorr are also visible, as is the implantation layer for both STEM and SLD. However, their appearance is qualitatively different, since the XRR and STEM were performed on samples of thermal and native oxide, respectively. All in all, the STEM profiles confirm the validity of our XRR model.

We now change the focus from the densities to the superstructure peak analysis. Various structural properties impact its height and width. The superstructure peak intensity decreases with fewer repetitions, loss of OOP periodicity, or misalignment of interfaces relative to the surface normal [11]. Reduced density contrast resulting from intermixing or roughness also lowers intensity by lowering layer separation [1,8]. It is important to note that specular XRR cannot distinguish between intermixing and roughness [63] and that the superposition of the superstructure peak with the thickness oscillation distorts the observed peak shape.

For better comparability, the inset in Fig. 3(a) depicts a zoomed-in version of the superstructure peaks on a linear scale. At the same pressure, the samples with Ta exhibit a more pronounced superstructure peak than their counterparts without Ta. Increasing the sputter pressure increases the intensity with Ta and decreases it without Ta. Based on the structures observed in STEM and consistent with the XRD data in the next section, we can attribute the increased intensity with Ta to three factors: (1) enhanced crystal structure, (2) better layer separation and (3) reduced curvature, whereas (1) dominates at 3 mTorr, and (2) and (3) dominate at 10 mTorr. Without Ta, the pressure increase introduces more disorder in the ML, leading to a wide angular spread of the Co/Pt interface normal and a loss of coherence of the OOP periodicity.

Lastly, the precise position of the superstructure peak strongly depends on the period of the ML; in this case, the thickness of one Co/Pt repetition. Using the Bragg equation

$$n \cdot \lambda = 2 \cdot d_{\text{ML}} \cdot \sin(2\theta/2), \quad (1)$$

the bilayer thickness d_{ML} can be determined for the first order ($n = 1$) using the 2θ position of the superstructure peaks with the x-ray wavelength (Cu K_α) of $\lambda = 1.54 \text{ \AA}$. The measured bilayer thicknesses average to $d = 1.10 \text{ nm}$, with a spread over all samples of $\pm 0.04 \text{ nm}$, matching the nominal thickness. The effect of refraction is negligible at the used angles.

C. Crystallographic characterization

We conclude our structure analysis by evaluating XRD and rocking scans of the samples, giving us a global picture of the sample structure, complementing the local STEM analysis in Sec. III A. Figure 4(a) presents the XRD scans of the samples with and without the Ta adhesion layer, with an intensity offset applied for clearer visualization. Visible are the peaks of Pt(111) from the seed layer at 39.5° and the ML laminate peak in FCC(111) texture [ML(111)] at around 41° . Because of the dense laminate of Co and Pt within the ML stack, an HCP Co(002) or FCC Co(111) peak is not present in the XRD profile. We can also identify two satellite peaks corresponding to the periodicity of the Co/Pt period at around 33° (ML-1) and around 25° (ML-2). For the 10 mTorr sample without Ta, a broad FCC(200) peak emerges at around 47° . The Pt(200)

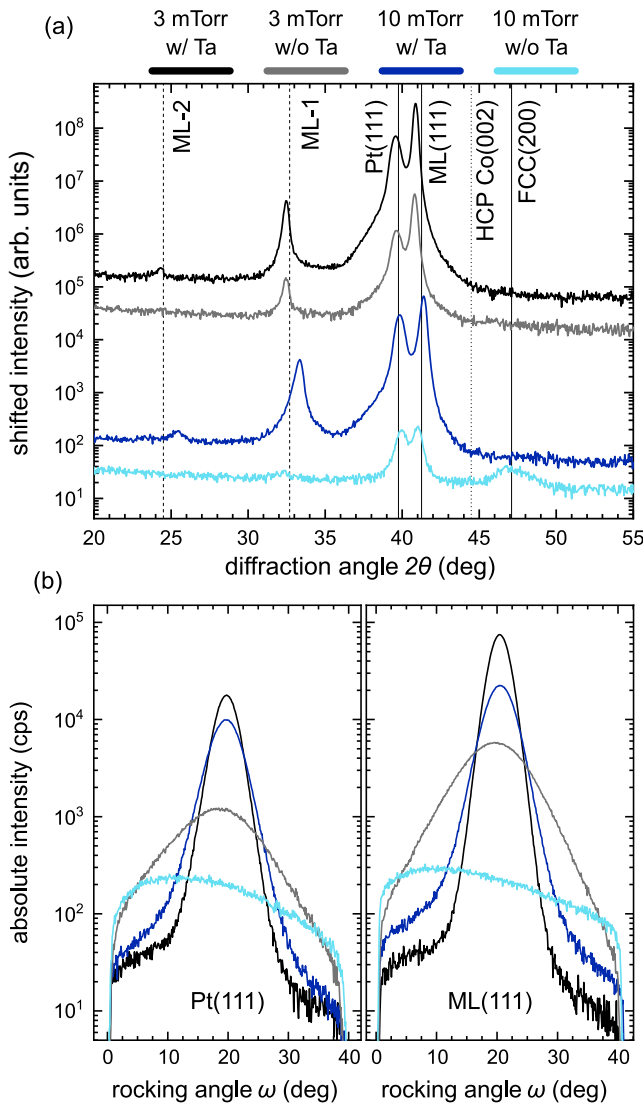


FIG. 4. XRD measurements of the samples with and without Ta adhesion layers sputtered at 3 mTorr and 10 mTorr are shown in (a). XRD intensities are shifted vertically for better clarity. The rocking curves with absolute intensities of the Pt(111) and ML(111) peaks are displayed in (b). In (a), the position of HCP Co(002) is indicated by the dotted line. FCC Co(111) would be at almost the same position. The position of ML(111) corresponds to the calculated bulk value of a Co_4Pt_7 alloy.

and ML(200) peaks cannot be fully distinguished because of the low degree of texture. The presence of an FCC(200) peak indicates polycrystalline growth in the OOP direction, consistent with the FFT in Fig. 1(d), showing the poor texture of this sample. In contrast, the other samples have a pronounced fiber texture with only FCC(111) peaks, although at 3 mTorr without Ta, the FFT in Fig. 1(b) displays some off-OOP-axis crystal components. These seem to be only a minor component, though, since there are no additional FCC peaks visible in the XRD pattern.

The pressure dependence of the crystal structure can be seen by the shift of the Pt(111) and ML(111) peaks. They shift to higher angles—and therefore lower out-of-plane lattice spacings—with increasing pressure, independent of the

Ta adhesion layer. This can be attributed to different amounts of stress in the film [20,23]. At 3 mTorr, the Pt(111) and ML(111) peak positions are similar for both underlayers, suggesting comparable lattice parameters and stress in the systems. For the 10-mTorr samples, a significant shift to higher angles occurs for the Pt(111) peak. The ML(111) is close to the 3-mTorr case without Ta, but shifts to considerably higher angles with Ta.

Assuming a biaxial IP stress and a positive Poisson ratio [64], the type of stress can be qualitatively estimated by comparing the 2θ positions with their respective bulk values, 39.76° for Pt(111) and 41.26° for ML(111) as shown in Fig. 4. The “bulk” Co/Pt ML is approximated by a Co_4Pt_7 alloy [8] with a linear mixing between the lattice parameters of FCC Pt(111) (2.265 Å, ICSD-52250) and FCC Co(111) (2.046 Å, ICSD-622435), resulting in a ML(111) lattice parameter of 2.185 Å ($2\theta = 41.26^\circ$). In this case, a lower-than-bulk angle is the result of a larger-than-bulk OOP lattice parameter, showing a tensile OOP strain and indicating IP compressive stress. Two major components are impacting the stress in the system. Firstly, the intrinsic stress caused by the deposition conditions, which causes a transition from compressive to tensile IP stress with increasing sputter pressure, shifting the XRD peaks to larger angles [23,65]. Secondly, the epitaxial strain caused by the interaction between the seed and ML. Since the average lattice parameter of ML(111) is smaller than that of Pt(111), the seed induces a tensile IP stress in the ML when a good epitaxy is present, shifting the ML(111) to larger angles. The stress in the Pt seed follows the pressure dependence from IP compressive to IP tensile. For the ML, the texture introduces an additional tensile component, shifting the ML(111) much more with increasing pressure with Ta, which is suppressed for the samples without Ta because of the much lower texture.

Note that the ML(111) position also depends on the individual thicknesses of Co and Pt within the ML. Changing those will also change the average lattice parameter, as it is the result of averaging the bulk values [8]. In our study, this factor can be excluded, considering the good agreement in Co/Pt bilayer thickness demonstrated by the XRR superstructure peaks in Fig. 3(a). The XRD scans display no peaks assignable to Ta, which is in agreement with the STEM images in Fig. 1 and the literature, where the amorphous nature of thin sputter-deposited Ta layers is reported [66,67].

The quality of the seed and ML texture can be inferred from the intensity and width of rocking curves for the corresponding Bragg peaks, displayed in Fig. 4(b). Lower intensity and broader peaks indicate a less pronounced texture and a wider distribution of the OOP lattice direction (mosaicity), which would correlate with a wider angular spread in the STEM FFTs (insets in Fig. 1). Because of the lower thickness of the seed layer, the intensity of Pt(111) is lower than that of ML(111). Nevertheless, trends in the rocking width for the different samples are similar for seed and ML, confirming their strongly correlated texture. Two consistent trends are visible: (1) adding an adhesion layer improves the texture considerably, and (2) increasing the pressure deteriorates the texture. This is true for both seed and ML. The Bragg peak intensities for the samples without Ta are 1–2 orders of magnitude lower for the seed and ML [note the log-scale y axes in Fig. 4(b)] and the rocking widths are considerably

wider compared to the samples with Ta. At 10 mTorr without Ta, the maximum rocking intensity is no longer at $2\theta/2$, but at significantly lower angles, confirming the mostly polycrystalline nature of the film in this case.

The satellite peaks of the ML(111) Bragg peak can present valuable information about the quality of the layering within the ML. They are the result of the artificial superstructure and are directly related to the ML period. Their positions are approximately 8° (ML(111)-1) and 16° (ML(111)-2) to the left of the laminate peak, corresponding to the ML period $d_{\text{ML}} \approx 1.1$ nm according to the Bragg equation Eq. (1), with n being the order, and 2θ the distance between ML(111) and satellite in Fig. 4(a). The satellite peaks are similar to the superstructure peaks in XRR [Fig. 3(a)], but with a sensitivity to the alignment of the atoms in their crystal structure instead of the laterally averaged electron density. For the samples with Ta, the intensity is strong, indicating a well-defined periodicity of Co and Pt in the ML. Without Ta, only the first-order satellite is visible and its intensity is much lower. At low pressure, the peak is narrow for both underlayers, with lower intensity for the sample without Ta. There is a drastic difference going from 3 mTorr to 10 mTorr for the different underlayers. The satellite peak almost vanishes without Ta, but increases in intensity and broadens when using the Ta layer. This is analogous to the behavior of the ML superstructure peak in XRR in Fig. 3(a). With Ta, increasing the sputter pressure results in two different trends for the intensity of the ML(111) laminate peak and the satellites. The ML(111) decreases in intensity, indicating more crystal defects. The rocking curves furthermore imply an increase in mosaic spread. At the same time, the intensity of the satellites increases, implying a better periodicity of the Co/Pt bilayer [1,8]. This could be caused by less displacement between the Co and Pt in the FCC(111) lattice and therefore less intermixing.

D. Magnetic characterization

Magnetic hysteresis curves in the easy-axis OOP and hard-axis IP direction are presented in Figs. 5(a) and 5(c), together with MFM images of the remanent state after OOP and IP demagnetization in Figs. 5(b) and 5(d), respectively. For comparison, the AFM images of the same area as the IP-demagnetized MFM images in Fig. 5(d) are shown in Fig. S2 with the SM [52]. Furthermore, to better describe the reversal behavior, the derivatives of the OOP loops are plotted in Fig. S4 within the SM [52]. Saturation magnetization M_s , normalized OOP remanent magnetization M_r/M_s , effective anisotropy K_{eff} , as well as OOP and IP coercive fields ($\mu_0 H_{c,\text{OOP}}$ and $\mu_0 H_{c,\text{IP}}$) are summarized in Table I.

For the 3-mTorr samples, the hysteresis loops are remarkably similar. The easy-axis OOP curves display the characteristic stripe domain field reversal [30,68]: a steep drop in magnetization followed by a section with constant slope around remanence and a steep end correspond to stripe domain nucleation and propagation, domain breathing and domain annihilation, respectively. The derivative in Fig. S4(b) within the SM [52] reflects this as a sharp peak, followed by a valley concluding in a smaller peak. After OOP demagnetization, the MFM images of these samples both show isotropic labyrinth stripe domain patterns, which is con-

TABLE I. Magnetic properties for the 3 mTorr and 10 mTorr samples with and without Ta. Presented are the saturation magnetization M_s , effective anisotropy K_{eff} , OOP normalized remanent magnetization M_r/M_s and coercive field in OOP $\mu_0 H_{c,\text{OOP}}$ and IP direction $\mu_0 H_{c,\text{IP}}$.

Pressure (mTorr) underlayer	3 w/Ta	3 w/o Ta	10 w/Ta	10 w/o Ta
M_s (kA/m)	590	582	598	464
K_{eff} (kJ/m ³)	254	207	691	156
M_r/M_s (%)	8.2	6.5	97.2	77.3
$\mu_0 H_{c,\text{OOP}}$ (mT)	25	20	447	259
$\mu_0 H_{c,\text{IP}}$ (mT)	32	40	94	99

firmed by the ring-shaped FFT image with comparable radii (domain width ≈ 220 nm). In the IP direction, the 3-mTorr samples show a hard axis loop with a slight S-shape and minimal hysteresis. The IP demagnetized state has aligned parallel stripe domains for both samples, with a smaller period visible for 3 mTorr with Ta. For such one-dimensional stripe domains [12,69], the domain size depends on the magnetic thickness, saturation magnetization and PMA. Only the PMA is slightly higher with Ta, which would favor a larger period. However, the stripe period of the sample with Ta is slightly smaller (≈ 140 nm as compared to ≈ 160 nm for the sample without Ta). This discrepancy might be resolved by following the reasoning of Kozlov *et al.* [70], who propose that different defect densities impact the formation of the stripe domain pattern during the demagnetization routine. In our cases, the better texture of the sample with Ta could also introduce more crystal defects, especially at the grain boundaries, which in turn could act as magnetic defects. We furthermore note that the domain width after OOP demagnetization is larger and also very similar for both samples (≈ 220 nm), again in agreement with the findings of Kozlov *et al.* [70]. Overall, both samples are magnetically comparable, despite the considerable structural differences discussed earlier.

At 10 mTorr, the influence of the underlayers is more pronounced. As anticipated from the literature [19], without the Ta layer, the OOP loops widen when increasing the sputter pressure. The OOP loop is slanted with an almost linear slope, starting the reversal significantly before reaching remanence. This results in reduced remanent magnetization. The sharp on- and offset of domain propagation and annihilation are absent from the loop. The reversal here is likely dominated by the switching of individual grains and grain clusters, where the intergranular exchange is reduced. Furthermore, very high OOP fields are required to completely saturate the sample, leading to a slightly lowered normalized magnetization within the displayed ± 1 T range. This is not surprising, considering the amount of off-axis pointing Co/Pt interfaces seen in Fig. 1(d), which introduce a hard-axis component in the easy-axis loop, because of the anisotropy direction being mostly determined by the Co/Pt interface normal [47]. In the IP direction, the curve for 10 mTorr without Ta is more S-shaped with notable hysteresis compared to the 3 mTorr samples. In the field range of this hysteresis, the OOP loop is also not fully saturated,

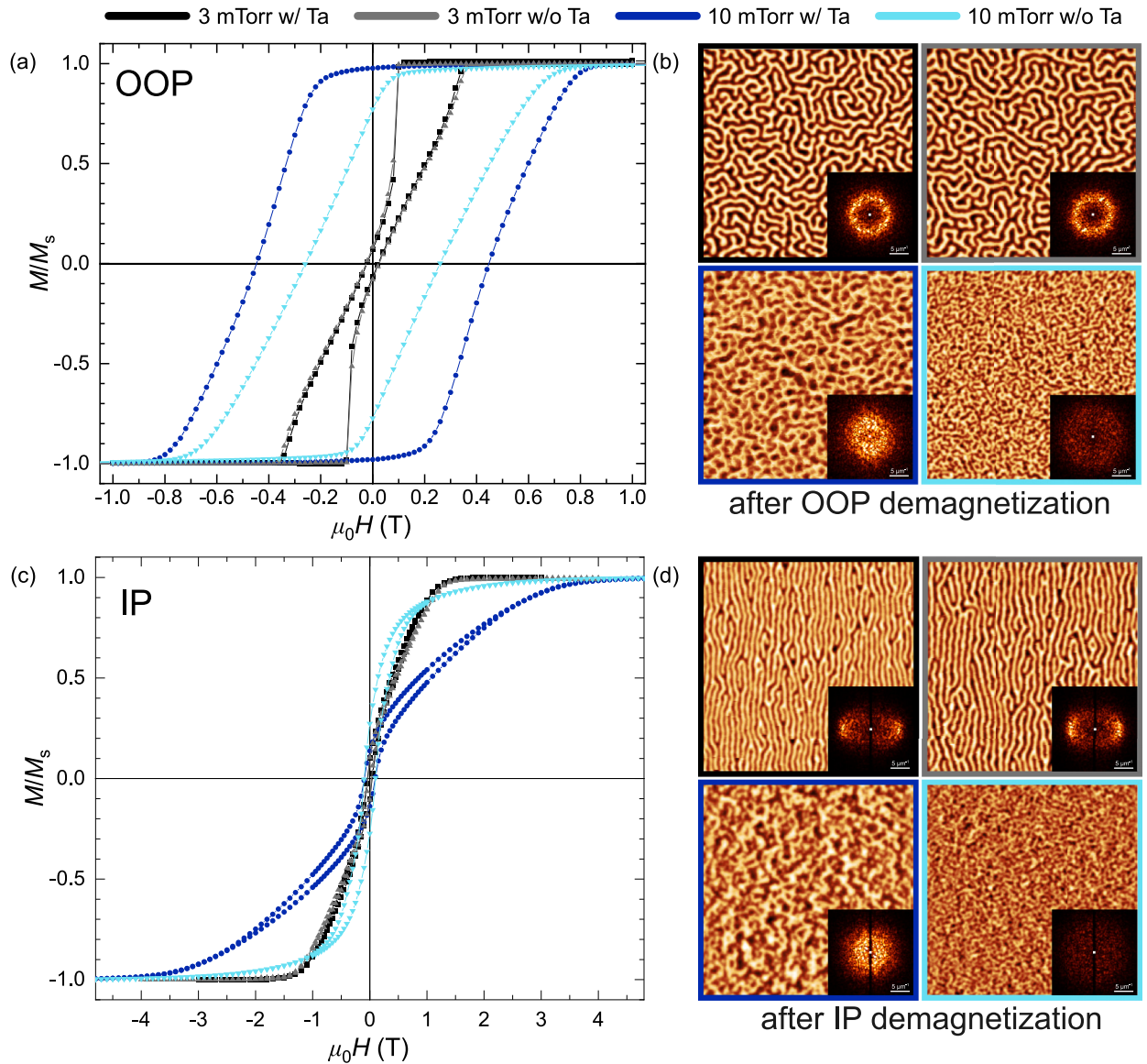


FIG. 5. Magnetic reversal behavior and remanent states of the samples with and without Ta adhesion layers sputtered at 3 mTorr and 10 mTorr. (a) Displays the OOP curves with the respective MFM images after OOP demagnetization in (b). (c) Shows the IP curves and the MFM images after IP demagnetization in (d). The MFM images are $(5 \times 5) \mu\text{m}^2$ in size, with an adjusted color scale applied per image for better visibility. The insets represent the $22 \times 22 \mu\text{m}^2$ FFTs of the MFM images and are all displayed with the same color scale.

which might be caused by grains with Co/Pt interfaces tilted relative to the film normal [see e.g., Fig. 1(d)], thus resulting in a mixture of easy and hard axis behavior.

The gradual saturation of the IP loop points to a broad distribution of anisotropy magnitude across the grains, which is supported by the disordered grain structure and Co/Pt contrast in the STEM image. Saturation magnetization decreases distinctly, consistent with the literature [19], and the effective anisotropy reduces by 25% compared to 3 mTorr without Ta. No regular labyrinth domain pattern forms after OOP demagnetization and there is no major alignment visible anymore after IP demagnetization, resulting in similar FFTs. Furthermore, the domains are much smaller than at 3 mTorr. Because of the microstructure, domain walls can follow the voids between grains, where the domain wall energy is reduced by the

lack of magnetic material and reduced intergranular coupling. This, in turn, leads to smaller and more ragged domains as a result of minimizing demagnetization energy, which is further facilitated by the lower saturation magnetization. Given the grain size from the STEM and AFM images, there are still dozens of grains per domain, indicating a smaller, but still considerable, intergranular coupling.

With Ta at 10 mTorr, the magnetic behavior changes drastically. The OOP loop widens further, increasing the coercive field and resulting in almost full remanence, as the main reversal only starts after remanence. The reversal slope is less slanted, with signs of remaining stripe domain nucleation [34], with a broad, but still defined peak in the first derivative in Fig. S4 within the SM [52]. MFM images in the OOP-demagnetized state depict large, but less-connected and more

ragged domains compared to the 3 mTorr samples. Nevertheless, a labyrinth stripe domain network is still recognizable after OOP demagnetization. The domain width is also much less defined, as is apparent from the broad ring in the FFT. In the IP direction, the loop has a shallower slope, indicating a considerably higher PMA. Specifically, at 10 mTorr, the effective anisotropy increases by 342%—or a factor of 4.4—when using the adhesion layer. Even compared to the 3 mTorr sample with Ta, the increase is still 172% (a factor of 2.7). The hysteresis opening remains similar, as evident from $\mu_0 H_{c,IP}$. The hysteresis also closes at much higher fields. MFM images after IP demagnetization consist of larger, ragged domains, lacking the parallel alignment seen at low pressure. Given the microstructure with much less pronounced grain boundaries compared to 10 mTorr without Ta, the larger domains can be attributed to the larger domain wall energy caused by the high anisotropy and better intergranular coupling. Note that the starting field of the IP demagnetization routine was below the saturation field for the 10 mTorr sample with Ta, because of the limits of the magnet.

IV. DISCUSSION

Combining all structural and magnetic results, we see the strong connection between underlayers, sputter pressure and structure. Especially, the sputter pressure is known to change the kinetic energy and flux of target atoms and reflected neutralized sputter gas ions. According to the forward sputtering model reviewed and refined by Windischmann [23,29], the structural changes in sputter-deposited films are caused by a cascade of atomic displacements, originating from a momentum-driven bombardment with particles during film growth. He defined a normalized momentum per target atom P_n^* , which implicitly assumes a negligible momentum of the target atoms themselves [23],

$$P_n^* = \frac{\Phi_g}{\Phi_t} \cdot \sqrt{2m_g E_g}, \quad (2)$$

where Φ_g and Φ_t are the particle fluxes of neutralized process gas (neutrals) and target atoms, E_g is the kinetic energy of the gas particles, transferred to the substrate and m_g is the mass of the gas atoms. Equation (2) can be generalized to include the momentum of all energetic species i ,

$$P_n^* = \frac{1}{\Phi_t} \cdot \sum_i \Phi_i \sqrt{2m_i E_i}, \quad (3)$$

which would include the gas (g) and target (t) atoms in our case. The momentum transfer of the sputter atoms can be neglected if their energy is very small. This is the case for fully thermalized sputter atoms, which happens within a couple to a dozen mean free paths [23,28], depending on the mass ratios between target and sputter gas atoms [28]. Thermalization is less pronounced for the neutrals because of their mass ratio equaling 1, enabling energy transfer through forward scattering. The heavy Pt might still contribute to the total momentum for our sputter conditions, but the main contribution should be from the neutrals [17,23]. Practically, we also cannot distinguish between the contribution of target atoms and neutrals to the normalized momentum because both are deeply connected to the same process parameters in DC magnetron sputtering. Higher target mass results in larger target atom energy, as

well as an increase in flux and energy of reflected Ar neutrals [23]. Furthermore, the contribution of the different species is not important for the forward sputtering model, since we only consider the total normalized momentum qualitatively and comparatively.

Without Ta, the evolution of the structural and magnetic properties aligns with previous literature and can be well explained by the forward sputtering model. When increasing the sputter pressure, the texture decreases, and the IP film stress transitions from compressive to tensile [23]. The density of the ML also decreases as seen from the STEM profiles and SLDs. We see a rise in roughness at the interfaces and considerably more disorder in the grain structure. As to the magnetic properties: anisotropy, saturation magnetization and domain size decrease, accompanied by an increase in coercive field strength. This is the expected behavior, bringing our samples in line with previously published results [9,18–20]. The decrease in saturation magnetization at 10 mTorr might be related to the large voids between the grains, as well as their disorder at the surface. The thin capping layer might no longer be sufficient to fully prevent oxidation of Co within the ML, as the wide grain boundaries enable a path for oxygen to react with the sides of the grains.

Based on the well-established behavior of the samples without an adhesion layer, we now discuss the changes by adding one, starting with the low-pressure case. At 3 mTorr, the only major change is the strong increase in crystal texture and moderate improvement in anisotropy. Roughness, density, layering, as well as the other observed magnetic properties, are all comparable. Since the momentum transfer to the film should be similar for both underlayers at the same pressure, we can attribute the improved texture to a less distorted substrate surface. Furthermore, the Ta layer prevents the penetration of Pt atoms into the substrate, maintaining a smooth growth surface for the seed. As a result, the momentum is not transferred deep into the substrate, which enables higher atomic mobility and facilitates better crystallization akin to a higher temperature deposition [13,61]. So, after nucleating a sufficiently thick Pt layer without Ta, the growth of the remaining film is similar, since it is dominated by the high normalized momentum. This gives rise to similar roughness and density, while the texture follows the poor template from the substrate surface.

Besides the texture, there is a respectable improvement of the anisotropy of roughly 25%. Since the Co and Pt thicknesses are in the order of a couple of monolayers, the crystal structure benefits from a good layering, the latter contributing to the higher intensity of the XRR superstructure and Bragg satellite peaks and, in turn, increasing the interface anisotropy. The higher texture could also increase the contribution from magnetocrystalline anisotropy [71], although this is expected to be a minor component in Co/Pt MLs [47]. The crystal structure might also have an impact on the angular spread of the anisotropy direction in the film. From the STEM images in Figs. 1(a) and 1(b), we can see that the chemical boundary between Co and Pt does not need to follow the lattice. The FFT insets show a comparable angular distribution for the ML period for both underlayers, but the FCC(111) planes exhibit a much stronger fiber texture in the OOP direction with a Ta underlayer. For a magnetocrystalline anisotropy component

along the FCC(111) orientation, this alone would increase the measured effective anisotropy between the OOP and IP. Still, the impact of magnetocrystalline anisotropy would be small, especially if we consider the behavior of the sample at 10 mTorr with Ta, as we will discuss next.

Increasing the sputter pressure results in vastly different structures, depending on the underlayer choice. When using the Ta adhesion layer, the texture still decreases with rising deposition pressure, but nowhere near as much as without it. Notably, the texture is still considerably better compared to the low-pressure sample without Ta. Similar to the texture, the seed/ML and cap/air roughnesses increase less strongly, although the grain height is unevenly distributed, resulting in higher RMS roughness values. We can again trace this back to the nucleation on the substrate. While the higher pressure decreases the normalized momentum per arriving target atom, it is still enough to enable forward sputtering into and through the SiO₂ layer when no Ta layer is present and a good texture when it is. The much better nucleation compensates for the lower normalized momentum, resulting in a dense and smooth seed. This also has an impact on the stress in the ML. While we see the same transition from compressive to tensile IP stress when increasing the pressure, the ML exhibits a much higher tensile IP stress with Ta than without at 10 mTorr. In this case, the much better texture, and therefore epitaxy between seed and ML, puts more tensile IP strain in the ML because of its smaller average lattice parameter. The density of the film seems to be mainly governed by the amount of voids and the density of the grain boundaries, as we can conclude from the very similar depth profiles extracted from STEM and XRR, as shown in Fig. S3 within the SM [52].

Breaking the expected pressure-dependent trend, we see an increase in the XRR superstructure and XRD satellite peaks, despite the decrease in ML(111) texture. Tied to this, the STEM images show a better contrast between Co and Pt, as well as a reduction in layer curvature, as is apparent from the FFT insets and the flatter tops of the dome-shaped grains in Fig. 1(c). The higher deposition pressure lowers the normalized momentum impinging on the film, which also reduces the amount of forward sputtering of already deposited atoms into the depth of the film. This lower intermixing between the thin layers yields higher intensity XRR superstructure and XRD satellite peaks, as well as better STEM contrast. Since the higher pressure also increases the angular distribution of the impinging atoms, self-shadowing caused by the confocal sputtering under an angle is reduced, resulting in less curved interfaces despite the reduced surface mobility, which also contributes to the intensity increase of the XRR superstructure peak.

The coercive field increases with the pressure, but is higher with Ta, which is a combination of partial grain separation caused by the voids in between the grains, as well as the improved anisotropy. The MFM images also demonstrate only a partial decoupling, enabling a disordered, but still recognizable, labyrinth domain pattern. As shown in the STEM images, wide grain boundaries only appear in the top part of the film, resulting in pinning sites located closer to the surface.

To better understand the anisotropy gains, we first need to consider the origin of the strong perpendicular magnetic anisotropy in Co/Pt MLs. As already reported earlier [40,47],

the major part of the uniaxial anisotropy is caused by the hybridization of the orbitals at the Co/Pt interface, besides additional contributions from the magneto-crystalline and magneto-elastic anisotropy [15]. Usually, there is an increase in PMA associated with the Co/Pt interfaces becoming “better” [1,40,72,73], namely: (1) There is a clear separation between Co and Pt, meaning an intermixing as low as possible. (2) The Co/Pt-interface-normal is everywhere close to the OOP direction. Any deviation there will reduce the directionality of the anisotropy. The latter condition includes the curvature of the interface within the grain, as seen in the STEM images in Fig. 1, which causes a distribution in the anisotropy direction. Short-length-scale roughness within the ≈ 20 nm grains (intragrain roughness) also affects the interface normal, resulting in a reduction of the PMA. In contrast, the longer-length-scale roughness caused by the height and shape of the grains (intergrain roughness), as visible in AFM, is less important for the anisotropy, but serves as an indicator for the domain reversal behavior, since nucleation, propagation and pinning of the domain walls strongly depend on grain-to-grain separation and variation in their height and shape [74,75]. We want to note that in general, a roughness-dependent dipolar PMA can also be present [41,76,77].

Many previous studies discussed the impact of roughness on the PMA and reversal behavior [14,20,22,47,55,78], but there is usually no distinction between intra- and intergrain roughness. Especially the 10-mTorr sample with Ta showcases the importance of this distinction, as the intergranular roughness clearly increases with rising sputter pressure, while the intragrain roughness is not affected. Note that this intragrain roughness is on the atomic scale, which is contextually very close to intermixing. In our study, intragrain roughness and intermixing denote the lateral and vertical atomic disorder, respectively.

Lateral intragrain roughness and intermixing compete with each other. This is nicely illustrated by increasing the pressure with and without Ta. At 3 mTorr, the seed roughness is comparable for both underlayers, leading to similarly curved interfaces in the ML as well, resulting in comparable magnetic reversal. With Ta, the crystal structure improves and therefore leads to better ordering on the atomic scale, resulting in a small PMA improvement, without changing the overall magnetic behavior. This changes at 10 mTorr. The normalized momentum decreases and the angle distribution of incident atoms widens because of the higher amount of scattering events. Without Ta at 10 mTorr, the roughness of the seed is cumulatively transferred to the ML, leading to high disorder. The Co/Pt interfaces are still present to some extent, but point in a wide range of directions, leading to the low PMA and wider anisotropy distribution. A considerable increase in inter- and intragranular roughness is visible. At 10 mTorr with Ta, the cumulative roughness effects are much less pronounced, resulting in less self-shadowing and therefore a reduced curvature of the interfaces, decreasing the anisotropy distribution and increasing the PMA. Furthermore, the lessened normalized momentum reduces the vertical intermixing between Co and Pt, increasing the anisotropy considerably.

Concluding our discussion, we emphasize the importance of the direction and magnitude of the normalized momentum,

as well as their interaction with substrate and underlayers. Especially the high mass ratio between target and gas atoms results in a high normalized momentum arriving at the film. If we just consider the adhesion layer, Ta bound to the substrate should be good at reflecting and dispersing the arriving moment because of its high atomic mass. We therefore expect that other often-used adhesion layers, such as Ti or Cr, are less suitable to prevent the forward sputtering of the Pt seed atoms, due to their lower mass. Based on our results, the relation between underlayers and sputter pressure should also apply to material systems containing other heavy elements, like Au and Ir. The effect of increased pressure and reduced moment also decreases with a lower target/gas mass ratio [23]. Therefore, the lower normalized momentum reduces the impact for ML systems with lighter elements, like Co/Pd or Co/Ni.

Based on the desired properties, we can use our results for different applications. The low-pressure case is and remains useful for achieving low pinning, which is cardinal for highly periodic magnetic stripe or bubble states [10,79], as well as a collective field-reversal in ML-based SAFs [32,80]. Since the crystal structure is only of minor importance, a transition of our findings to amorphous systems, e.g., CoB/Pt or CoFeB/Pt, should be possible. In such systems, we expect different grain boundary properties, making them suitable for spintronics applications by reducing spin damping. Furthermore, the understanding gained in our study paves the way for more complex tuning of the sputter deposition conditions and, with it, the sample properties. For example, PMA could be further adjusted by tuning the sputter pressure for the underlayer/seed stack and the ML separately. Higher or lower grain separation might be achievable by changing the temperature during deposition. Since grain separation effects only occur at a larger number of repetitions, different pressures can be used to vertically tune the anisotropy without changing the thicknesses or switching the material, for example, by continuously [81] or individually changing the sputter pressure for the layers.

V. CONCLUSIONS

In conclusion, the addition of a suitable adhesion layer such as Ta plays a major role in the structural and magnetic

properties of Co/Pt MLs. At a low sputter pressure of 3 mTorr, the texture and anisotropy improve, while the overall magnetic reversal behavior stays similar. At a high sputter pressure of 10 mTorr, the Ta layer results in much better texture, reduced intermixing and lower curvature of the interfaces within the grains, resulting in a considerable improvement of the effective perpendicular anisotropy, reaching 691 kJ/m^3 at 10 mTorr with Ta, which surpasses the value of 254 kJ/m^3 at 3 mTorr by a factor of 2.7. We attribute the differences between the underlayers to the implantation of Pt into the substrate in the absence of the Ta layer, resulting in considerably higher roughness and lower texture. With the Ta layer, this effect is mediated by blocking the momentum transfer into the substrate. The reduced normalized momentum and less oblique flow of atoms in our confocal sputter geometry at higher deposition pressure lead to lower intragranular roughness (intermixing) and grain curvature, as well as a better texture, thus explaining the increased PMA.

We emphasize that the role of the normalized momentum arriving at the film and its interaction with the substrate are of major importance for the magnetic anisotropy and the reversal behavior. It should therefore be considered when optimizing the magnetic behavior for certain applications.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [82].

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