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Originally published:

September 2020

Physical Review B 102(2020), 094434

DOI: <https://doi.org/10.1103/PhysRevB.102.094434>

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1 *Higher order ferromagnetic resonances in out-of-plane*
2 *saturated magnetic multilayers*

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13
14 *Abstract*

15 Artificial ferromagnetic (FM)/nonmagnetic multilayers, with large enough FM thickness to prevent
16 the dominance of interface anisotropies, offer a straightforward insight into the understanding and
17 control of perpendicular standing spin wave (PSSW) modes. Here we present a study of the static
18 and dynamic magnetic properties of $[\text{Co}(3.0\text{nm})/\text{Au}(0.6\text{nm})]_{1 \leq N \leq 30}$ multilayer systems.
19 Magnetometry reveals that the samples exhibit magnetization reversal properties typical of an
20 effective single layer with weak perpendicular anisotropy, with the distinctive thickness-dependent
21 magnetization reorientation transition from in-plane to out-of-plane. When such multilayer systems
22 are out-of-plane saturated however, the dynamic response reveals the existence of several different
23 ferromagnetic resonances in the form of PSSW modes that strongly depend on the material
24 modulation characteristics along the total thickness. These modes are induced by the layer stacking
25 itself as the effective single layer model fails to describe the complex dynamics observed in the
26 system. In contrast to most systems considered in the past, described by a dynamic model of a
27 single effectively homogeneous thick layer, the specific structures investigated here provide a
28 unique platform for a large degree of tunability of the mode frequencies and amplitude profiles.
29 We argue that the combination of periodic magnetic properties with vertical deformation gradients,
30 arising from heteroepitaxial strain relaxation, generates a vertical regular array of two-dimensional
31 pinning sites for the PSSW modes, which promotes the complex dynamics observed in the system.

I. Introduction

Multilayer (ML) systems consisting of alternating layers of ferromagnetic (FM) and non-magnetic (NM) materials have gained steadily growing interest from fundamental physical perspective as well as for technological applications [1-3]. Due to the large variety of short- and long-range interactions acting in these MLs, their characteristics can be conveniently modified through small changes in the specifics of the layer stack. Concerning the static magnetic behavior, the uniform out-of-plane (OOP) magnetic ground state may be achieved by appropriately choosing the material parameters, primarily supported by the FM/NM interfaces (surfaces) in the thin film regime [4-7]. An increase of either the thickness of the individual FM layer component or the total number of the FM/NM repetitions causes the occurrence of non-uniform microscopic magnetization states, resulting in a magnetization reversal process dominated by the collective propagation of OOP domains [8-10]. However, this change could also lead to a complete in-plane (IP) reorientation of magnetization. This may be due to either the absence of a sufficiently high OOP magnetocrystalline anisotropy or an insufficient ML volume, both causing an IP uniform state being preferred over an OOP domain state [11-16]. Indeed, these changes do not only impact the static magnetic properties, but substantially also the dynamic behavior of ML systems.

Out of many experimental methods that have been applied to dynamic studies, ferromagnetic resonance (FMR) has proven to be one of the most powerful tools to sensitively evaluate any change and evolution of magnetic properties [17-22]. While measuring the uniform or quasi-uniform resonant mode, information about the volume averaged magnetic properties can be gained. The higher order modes are more sensitive instead to exchange and anisotropy energies spatially varying across the thickness as well as to OOP magnetic inhomogeneities or modulations within the system [17-23]. Among the latter, perpendicular standing spin wave (PSSW) modes are excitations confined within the thickness of the film. Their wavelengths are usually determined by the total thickness and the magnetization pinning conditions at the top/bottom surfaces, which also govern the energy needed to access and drive the PSSW modes in FM single layers [23-27]. Although PSSW modes have been already investigated in homogeneous thick films as well as in ML systems to quite some extent, their behavior in FM/NM-systems in the regime of thick individual FM-layers—too thick to allow the interface anisotropy to dominate—has not been studied so far. In fact, FM/NM multilayers naturally provide a suitable platform to introduce systematic and regular layer-like pinning sites at the interfaces throughout the total thickness,

63 otherwise confined at the top and bottom surfaces of a magnetically homogeneous system.
64 Therefore, MLs represent an efficient means to excite and manipulate such exchange-dominated
65 spin-wave modes, which could potentially have implications for novel high-frequency spintronic
66 applications [27-29].

67 The studied system consists of $[\text{Co}(3.0 \text{ nm})/\text{Au}(0.6 \text{ nm})]_N$ MLs with a variable number
68 of Co/Au bilayer N . We have chosen the individual and total thicknesses such that the interface-
69 induced anisotropy is not primarily driving the static magnetic response. Moreover, we have grown
70 our ML structures on top of a thick Au(111) buffer layer, such that Co, which adopts at room
71 temperature the hexagonal close-packed (*hcp*) crystal structure and exhibits a magnetic easy axis
72 along the *hcp* crystal c -axis, grows with the necessary crystallographic texture to induce an OOP
73 anisotropy-axis orientation supported by the specific total thickness range [11,14-16,30]. We note
74 that similar multilayer sample structures, where the Au(0.6 nm) layers were substituted by Pt(0.6
75 nm) layers, were already fully statically characterized [30] and provided valuable knowledge for
76 the dynamic studies presented here. Finally, the proposed FMR model is based on the Landau-
77 Lifshitz equation of motion [31] in order to explain the experimental data, where variations of the
78 anisotropy fields along the thickness of the ML structure are included.

79 The paper is organized as follows. We describe the experimental details in Sec. II. Then,
80 Sec. III A details the identification of the crystal structure and the evaluation of the epitaxial
81 relationships. In Sec. III B, the room-temperature magnetometry characterization is presented,
82 whereas the FMR spectroscopy study together with analytical calculations are discussed in Sec. III
83 C. Finally, Sec. IV provides a summary of the results and general conclusions.

84 II. Experimental details

85 The samples were grown at room temperature by dc magnetron sputtering in an ultrahigh vacuum
86 system (ATC 2200 series from AJA International, Inc.) with a base pressure better than 3×10^{-6} Pa.
87 Si(001) substrates covered by a 100-nm-thick thermal SiO_x layer were used. Each layer was
88 deposited using a pure Ar pressure of 4×10^{-1} Pa. The $[\text{Co}(3.0 \text{ nm})/\text{Au}(0.6 \text{ nm})]_N$ MLs were
89 grown on Ta(1.5 nm)/Au(20 nm) seed layers. The Au (20 nm) buffer layer serves to maximize the
90 *hcp* (0001) texture within the Co/Au MLs similar as also done with Pt seed layers in Co/Pt
91 multilayers [30]. The thickness of the Au cap layer was 2.4 nm, which is sufficient to prevent
92 oxidation as well as aging effects after removal from the vacuum system. Au as capping material
93 has also been chosen to avoid breaking the spatial inversion symmetry along the OOP direction of

94 the ML [32]. The investigated periods of the ML were $N = 1, 2, 3, 4, 6, 8, 10, 14, 18, 22, 26,$ and
 95 30. A schematic representation of the sample structure, including its specific layer sequence and
 96 thicknesses, is shown in Fig. 1. The structural analysis of the samples was performed by means of
 97 x-ray diffraction (XRD) and reflectivity (XRR) utilizing a Rigaku SmartLab X-ray thin film
 98 diffractometer operated with Cu- K_α radiation. Magnetization measurements were performed using
 99 a Microsense EZ7 vibrating sample magnetometer (VSM), equipped with a 360° rotational stage.
 100 The dynamic magnetic properties were characterized by means of vector network analyzer
 101 ferromagnetic resonance spectroscopy (VNA-FMR). The samples were placed flip-chip onto a
 102 coplanar waveguide and the complex transmission parameter S_{21} was measured at constant
 103 frequency as the FMR signal, while the magnetic field was swept through resonance [33,34].

104 III. Results and Discussion

105 A. Structural characterization

106 Figure 1 illustrates XRR scans in the range of $0.5^\circ \leq 2\theta \leq 8.0^\circ$ for the entire set of Co/Au samples,
 107 with the left inset showing the specific layer sequence that we used including all template layers.

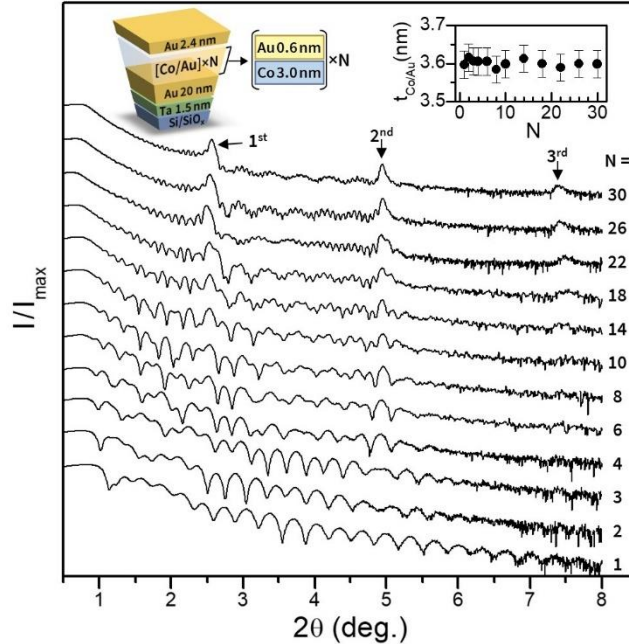


FIG. 1. XRR scans of all $[\text{Co}(3.0\text{nm})/\text{Au}(0.6\text{nm})]_N$ multilayer samples with $1 \leq N \leq 30$ repetitions. The top-left inset shows a schematic drawing of the layer stack. The top-right inset displays the average total thickness of the single Co/Au bilayer as a function of N as obtained from XRR measurements.

108 Interference-caused Kiessig fringes with two different periods are distinguishable in Fig. 1 for high
 109 N . The first type with a period inversely proportional to N can be observed at low 2θ values. These

110 short-period oscillations correspond to the total thickness of the MLs, since by increasing N (i.e.
 111 the total sample thickness) the distance between two consecutive minima or maxima decreases. At
 112 the same time their relative intensity decreases while increasing N , due to the increasing number
 113 of interfaces as well as due to the light absorption in each individual layer. In a wider 2θ -range a
 114 second set of Kiessig fringes is noticeable, whose period ($\Delta\theta \approx 0.2^\circ$) is constant as a function of N .
 115 They originate from the 20-nm-thick Au buffer layer. More importantly, the development of first,
 116 second, and third order Bragg-like superstructure peaks are observed with increasing N , giving a
 117 clear signature of a well-defined periodic elemental ML modulation. Fitting them to a Gaussian
 118 profile, the total thickness of the repeating Co/Au bilayer $t_{\text{Co/Au}}$ was evaluated [35]. The right inset
 119 in Fig. 1 shows the N -dependence of $t_{\text{Co/Au}}$, where the error bars correspond to the standard
 120 deviation values. Under the assumption of a purely statistical Gaussian distribution for the observed
 121 $t_{\text{Co/Au}}$ values, the data sets fall into the interval defined by $t_{\text{Co/Au}} = 3.61 \pm 0.02$ nm, consistent with
 122 the nominal Co/Au bilayer thickness of 3.6 nm.

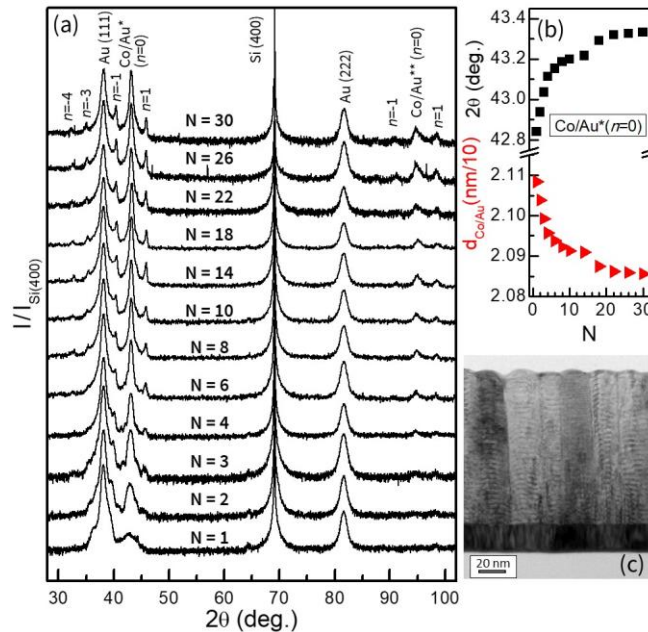


FIG. 2. (a) X-ray θ - 2θ scans of all samples. Each scan has been normalized to the intensity of its Si (400) substrate peak. (b) The top panel shows the N dependence of the Co/Au* ($n=0$) diffraction peak position (squares), while the bottom panel displays the associated average out-of-plane interplanar distance $d_{\text{Co/Au}}$ of the Co/Au heterostructure vs. N . (c) Cross-sectional TEM image of the $[\text{Co}(3.0 \text{ nm})/\text{Au}(0.6 \text{ nm})]_{30}$ sample.

123 Figure 2(a) shows the XRD scans in the full angular range $28^\circ \leq 2\theta \leq 102^\circ$ for all Co/Au
 124 samples. All the scans look very similar in their overall appearance, exhibiting only well-defined
 125 diffraction peaks corresponding to Si(004), Au(111), and Au(222) originating from the buffer
 126 layer. The superstructure reflections Co(0002)/Au(111) and Co(0004)/Au(222) are visible as well.

127 In the following they are abbreviated as Co/Au* and Co/Au** respectively. Even though
128 heteroepitaxial growth of crystalline layers is usually performed directly on top and facilitated by
129 single crystalline substrates, the presence of a thermally oxidized SiO_x thick layer is not preventing
130 the sputtered Au thin films from developing a highly oriented *fcc* (111) textured growth [30]. In
131 fact, *fcc* metal films have the largest atomic packing density along the (111) crystal plane and
132 thereby the lowest surface energy. Therefore, while using such materials as buffer layers, the
133 sample architecture studied in this work could very well be transferred to a large variety of different
134 substrates without losing the specific crystal quality. Moreover, the Au buffer layer peaks look
135 virtually the same for all samples, both in terms of angular position and peak width, verifying the
136 robustness of our fabrication process.

137 Hereby, the total X-ray scan angular range shows only well-defined *fcc*(*nnn*) and *hcp*(0002*l*)
138 peaks for Au and Co, respectively [36]. Moreover, satellite reflections around the superstructure
139 diffraction peaks [indexed by *n* in Fig. 2(a)] have been measured, which are a clear indication of a
140 perpendicular structural coherence length far larger than the thickness of the individual constituent
141 layers. It can be also observed that the negative indexed satellite diffraction peaks for the Co/Au
142 samples have higher intensity than the positive ones, with the latter being within the noise level for
143 *n* > 1. This effect, whose prominence being mainly appreciable for the first order diffraction peaks,
144 is mainly caused by the overlapping and interfering waves coming from the negatively indexed
145 satellite diffractions and the Au(111) reflection.

146 Furthermore, the diffraction conditions for the Au(111) lattice planes occur at different angles,
147 due to their different lattice dimensions resulting in $d_{111,Au} = 0.236$ nm, compared to the volume-
148 averaged Co(3.0nm)/Au(0.6nm) bilayer, corresponding to $d_{Co/Au^*} = 0.208$ nm. This would lead to
149 different magnitudes of strain throughout the ML structures depending on the number of times the
150 Co/Au bilayer building block was repeated. Thus, one could anticipate that otherwise identically
151 prepared and similar films of different thickness may have different depth-dependent strain
152 profiles. As expected, the diffraction peak intensity for Co/Au* and Co/Au** increases as the total
153 structure becomes thicker, simply due to the larger amount of epitaxially ordered material.
154 Nevertheless, upon carefully analyzing their angular position as a function of *N* [Fig.2(b)], a
155 continuous shift to higher 2θ angles is found as *N* increases. This indicates that for small *N* the
156 Co/Au bilayers are under larger tensile strain, which is partially being released as the growth
157 progresses. For samples with a larger value of *N*, the thickness dependent strain relaxation becomes
158 increasingly prominent, hereby approaching a bulk-like lattice parameter value as shown in Fig.

159 2(b). This process is occurring within each sample via the internal relaxation of the lattice mismatch
160 induced stress by the occurrence of misfit dislocations and lattice defects during growth, which
161 would then propagate towards the Co/Au interfaces. One could also conclude that the dislocations
162 are more numerous in the early stages of growth once they start to form, as the stress needed to
163 relief is larger at this point as compared to the topmost extent of the larger N samples, where a
164 significant part of the stress has already been released. From this point of view, it is reasonable to
165 assume that the Co/Au ML samples possess a gradient of strain across their vertical extent. In spite
166 of these considerations, the first and second order superstructure diffraction peaks for the Co/Au
167 MLs occur at nearly the same angle for $N > 10$ repetitions, thus indicating that the total thickness
168 is larger than the critical thickness needed to relax the corresponding strains.

169 Moreover, it cannot be neglected that the presence of several interfaces due to the alternate
170 deposition of FM/NM layers could influence the ML growth by inducing the formation of specific
171 morphological features. Fig. 2(c) shows a cross sectional bright-field TEM image for $N = 30$.
172 Without impacting the detectable and well-defined crystallographic texture measured using X-rays,
173 a columnar structure is clearly visible in the micrographs together with a precise and well-
174 established layered structure. The columns are mostly extended throughout the whole thickness of
175 the ML film and are largely perpendicular to the substrate with almost parallel walls. Nonetheless,
176 cumulative layer waviness is also observed due to the shadowing effects inherent to the room
177 temperature sputtering process and as a result of the heteroepitaxial growth initiated from the buffer
178 layer and followed by two distinct material species with different epitaxial relationships, as well as
179 due to the thickness discrepancy of the constituent layers in the ML [37,38]. Thus, both the
180 observed strain-relaxation and waviness can be regarded as important aspects related to the
181 magneto-dynamic properties of our samples, as will be discussed in Section IIIC. Hereby, our
182 structural analysis confirms the good crystallographic quality of the optimized layer growth
183 sequence resulting in well-modulated Co/Au MLs with perpendicular c-axis orientation, necessary
184 for a preferential OOP orientation of the magnetization above a critical thickness [11-16].

185 *B. Magnetostatic characterization*

186 Figures 3(a)-(l) present room temperature normalized M/M_S data as a function of the field strength
187 $\mu_0 H$ and number of Co/Au bilayer repetitions N , with M_S being the saturation magnetization. In
188 each graph, the (black) short dashed and (red) solid lines show the magnetization reversal curves
189 measured for an external magnetic field applied parallel and perpendicular to the film plane,

190 respectively. Each sample with $14 \leq N \leq 30$ shows two reversal curves that are a clear indication
 191 of an OOP preferential orientation of magnetization [8,14,16,30].
 192 For the IP field configuration, as one lowers the applied magnetic field, the saturated state becomes
 193 unstable at a critical field H_{cr} [pointed to in Fig.3(l)] and undergoes laterally alternating
 194 magnetization rotations into a tilted stripe domain state driven by the magnetocrystalline OOP
 195 anisotropy [8,14,16,30]. For $0 < H < H_{cr}$ this tilted stripe domain state becomes the system ground
 196 state [39] and the precursor to perpendicular stripe domains at remanence. Its OOP magnetization
 197 modulation increases, and the IP magnetization decreases as the applied field is further reduced,
 198 which leads to the overall curved appearance of all the in-plane $M(H)$ measurements in Fig. 3 for
 199 $14 \leq N \leq 30$. However, a final IP magnetization component still persists at remanence due to Bloch
 200 type domain walls that have been aligned during the field sequence into the in-plane external field
 201 direction and that are responsible for the IP hysteresis that occurs for low field values [14-16].

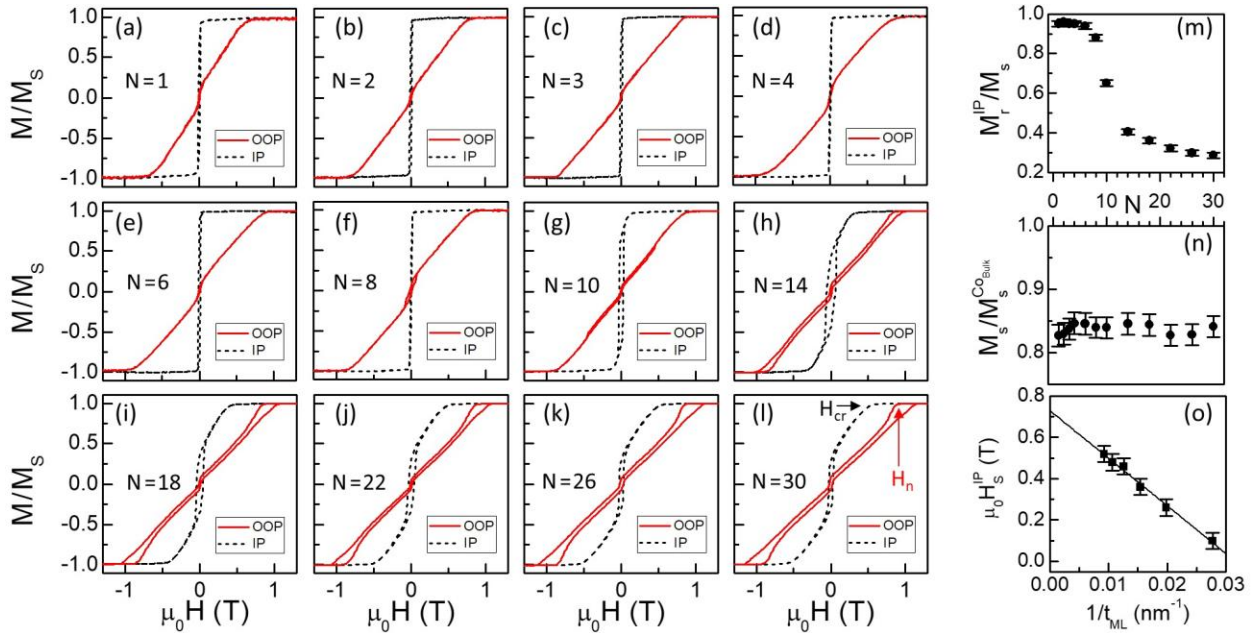


FIG. 3. (a)-(l) VSM room temperature (RT) magnetization reversal curves with the applied field along the IP (black dashed lines) and OOP (red full lines) directions for the entire set of $[\text{Co}(3.0 \text{ nm})/\text{Au}(0.6 \text{ nm})]_N$ samples. Data is normalized to its M_S in each case. (m) N -dependence of the in-plane remanence ratio M_r^{IP}/M_S [obtained from the magnetometry data of (a)-(l)]. (n) RT saturation magnetization M_S as a function of the Co/Au bilayer repetitions N . (o) RT in-plane saturation field $\mu_0 H_s^{\text{IP}}$ as a function of the inverse total multilayer thickness $t_{\text{ML}} = N \times \Lambda$, with $\Lambda = 3.6 \text{ nm}$ being the Co/Au bilayer period. The solid line represents the least-squares fits to Eq. (1).

202 For the OOP field configuration instead, once the field is lowered, the uniform state is
 203 broken by the formation of bubble domains with opposite OOP magnetization orientation driven
 204 by magnetostatic energy. This process starts rather abruptly at the nucleation field H_n^{OOP} [pointed
 205 out in Fig. 3(l)], leading to a sharp drop in the magnetization. As the field is further reduced the

206 domain dimensions increase to minimize the total magnetostatic energy resulting in the linear field
207 dependence of the magnetization down to remanence [9,40,41]. Finally, the small hysteresis effect
208 is the result of a non-uniform domain expansion and contraction, which is related to the existence
209 of slight sample imperfections. An appreciable change is noticeable as well in the two OOP-
210 hysteretic regions at high field magnitudes, associated with the initial nucleation and final
211 annihilation of domains. In fact, upon increasing N , i.e. the total thickness, their position along the
212 magnetic field axis shifts to progressively higher values as well as their hysteretic area becomes
213 gradually larger [14-16,30].

214 For the samples with $1 \leq N \leq 8$, despite the OOP orientation of the magnetocrystalline
215 anisotropy axis, an IP behavior was found. For those samples the measurements show almost
216 perfectly rectangular-shaped hysteresis loops for the IP applied field. In contrast, the OOP field
217 data in Figs. 3(a)-(f) show an almost completely reversible change in the magnetization orientation
218 where the complete alignment is reached only at a field strength of $\mu_0 H^{\text{OOP}} \approx \pm 1$ T.

219 Figure 3(g) shows the IP and OOP magnetization curves for the sample with $N = 10$. While
220 the OOP curve shows the absence of high field nucleation hysteresis near saturation, the IP
221 configuration demonstrates the persistence of a strong curvature of the loop and a very small
222 remanent magnetization. Thus, the sample is evidently not in an IP magnetization state at
223 remanence, meaning that it must undergo a perpendicular or tilted stripe domain reversal process
224 if one lowers the externally applied field [30].

225 Finally, from our experimental data in Fig. 3, we concluded that N has a profound impact
226 on the magnetization reversal characteristics of $[\text{Co}(3\text{nm})/\text{Au}(0.6\text{nm})]_N$ MLs. The overall
227 appearance of the IP and OOP magnetization reversal curves stays very similar for $14 \leq N \leq 30$
228 [Figs. 3(h)-(l)]. However, while lowering the number of repetitions, thickness-induced
229 magnetization reorientation transition takes place, which culminates with a reversal mechanism
230 characterized by IP magnetization states alone [11,15,30]. The transition from a remanent OOP
231 stripe domain state (for $N \geq 14$) to a remanent uniform in-plane state (for $N \leq 8$) is mainly driven by
232 the canting of the local magnetization [30]. Such canting angle varies monotonically with N as
233 indicated by the N dependence of the IP remanence ratio M_r^{IP}/M_S shown in Fig. 3(m). Samples with
234 $N > 14$ exhibit a low M_r^{IP} , arising mainly from the local magnetic moments confined inside the
235 domain walls. The IP remanence gradually increases when lowering N , indicating a canting of the
236 domain magnetization towards the film plane. Finally, the ratio reaches values corresponding to a

237 full IP remanent magnetization, thus confirming that the Co/Au samples (for $1 \leq N \leq 8$) are
 238 evidently in an IP-magnetized state at remanence.

239 The observed magnetic crossover from OOP to IP should be still correctly described by
 240 treating each ML as an effective single layer systems with the same saturation magnetization M_S ,
 241 anisotropy K_u , and exchange stiffness A for each different total thickness [14,15,42-47]. We have
 242 determined the room temperature saturation magnetization M_S for the entire set of samples with
 243 different N by taking into account the total magnetic thickness as $t_{ML} = N \times \Lambda$, with $\Lambda = 3.6$ nm
 244 being the Co/Au bilayer period. The results are plotted in Fig. 3(n) after having them normalized
 245 to the bulk value of the Co saturation magnetization of 1450 kA/m [48]. The data are showing that
 246 the ratios are nearly independent of N , i.e. the total thickness, with an average value of
 247 $\overline{M_S(\text{Co/Au}) / M_S^{\text{Co bulk}}} = 0.84 \pm 0.01$. The stability of M_S and other magnetic parameters (see below)
 248 allows us to ascribe any significant change in the magnetic properties to the specific number of
 249 repetitions N as opposed to the inherent Co thickness variations or substantial CoAu alloying,
 250 which would both lead to substantial variation in the saturation magnetization. However, we could
 251 see that by increasing N from 1 to 4, the volume-averaged saturation magnetization slightly
 252 increases. In an alloy material $\text{FM}_{1-x}\text{NM}_x$ the saturation magnetization monotonically decreases,
 253 while increasing the percentage of the NM element x . In our MLs the same effect is occurring at
 254 low N due to the very small magnetic moment induced into Au [49].

255 Furthermore, the invariance of effective single layer magnetic parameters while changing
 256 N [15,46], can be also verified by the linear relationship between the IP saturation field $\mu_0 H_s^{\text{IP}}$ and
 257 the inverse of the total ML thickness t_{ML} :

$$\mu_0 H_s^{\text{IP}} = \frac{2K_u}{M_S} - \frac{2K_u}{M_S} \frac{1}{\sqrt{1 + \frac{2K_u}{M_S^2} t_{ML}}} \frac{t_{cr}}{t_{ML}} \quad (1)$$

258 with t_{cr} being the critical thickness at which the reorientation transition occurs. In Fig. 3(o) we
 259 show the $\mu_0 H_s^{\text{IP}}$ experimental data for $10 \leq N \leq 30$ as a function of $1/t_{ML}$, excluding the samples
 260 that show already a preferential IP magnetization. All the data follow a linear trend and share a
 261 common intercept $2K_u/\mu_0 M_S = 0.73$ T. Furthermore, we have obtained $t_{cr} = 41.4 \pm 1.8$ nm (approx.
 262 corresponding to $N = 12$), in agreement with the experimental data. It is worth emphasizing the
 263 point that the evaluated magnetic anisotropy is integrated over the entire ML volume, and thus, the

264 total-thickness dependent (or stress release dependent) interface and magnetocrystalline
 265 anisotropies within each sample are averaged out.

266 Accordingly, despite the existence of a magnetization magnitude modulations along the
 267 thickness, whose degree depends on the nonmagnetic material, all the samples behave as single
 268 effective ferromagnetic layers with weak PMA under the application of external magnetic fields
 269 [14,16]. Nonetheless, all the OOP reversal curves are almost identical in the high field regime.
 270 Therefore, appreciable changes from sample to sample are not expected in terms of dynamic
 271 properties of their magnetization in the saturated state. However, and as already shown in Fig. 2
 272 and discussed in Sec. IIIA, the interface and magnetocrystalline anisotropy should be considered
 273 as thickness dependent due to the strain evolution within each ML sample along the thickness,
 274 which will be the focus of the following Section.

275 C. FMR study

276 The FMR was measured using a VNA-based spectrometer in a field-sweep-mode. For each field-
 277 sweep a fixed microwave frequency in the range from 0 to 35 GHz was selected.

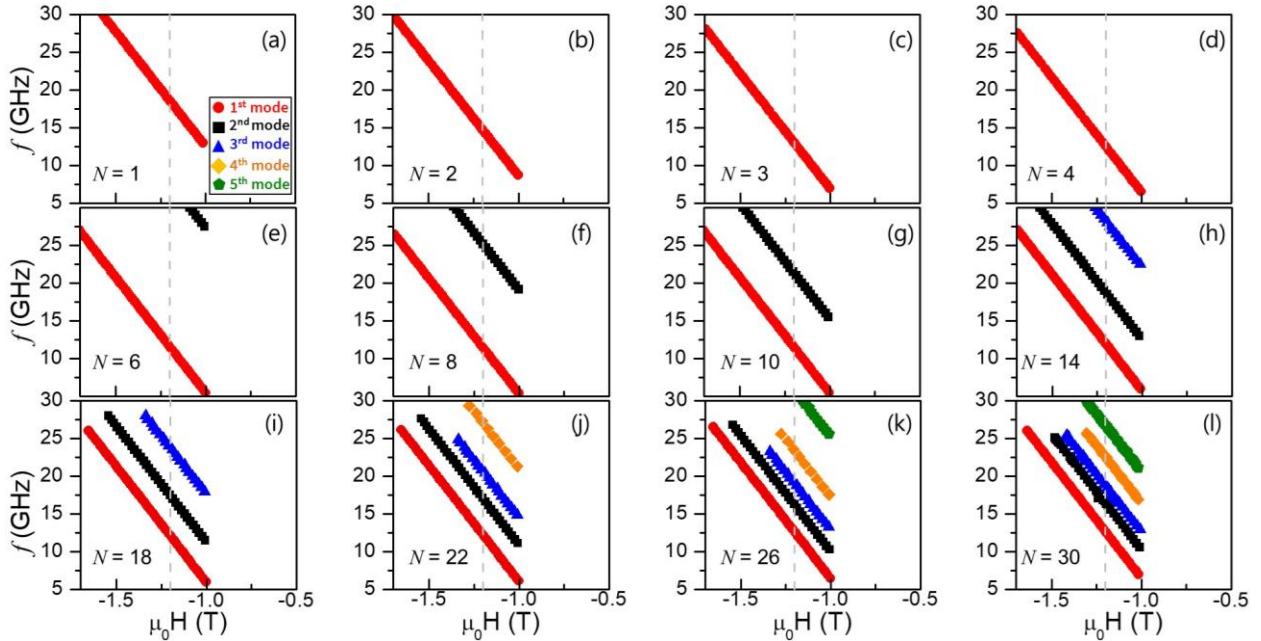


FIG. 4. (a)-(l) Frequency vs. field dependences of the ferromagnetic resonance for each sample. The external OOP field was always swept from -1.7 T to -1 T, ensuring always saturated OOP states. The vertical dashed (gray) lines mark the value $\mu_0 H = -1.2$ T corresponding to the vertical cut that is used to construct Fig. 5.

278 As we have seen from the hysteresis curves in Fig. 3, a static OOP magnetic field of $\mu_0 H = \pm 1$ T is
 279 sufficient to saturate the magnetization of each sample in the normal direction. Therefore, to ensure
 280 that all measurements start and run in the same magnetic state, the magnetic field was always swept

281 from $\mu_0 H = -1.7$ T to $\mu_0 H = -1$ T. The FMR spectra were fitted with a complex Lorentzian function
282 to retrieve the resonance fields.

283 Figure 4 shows the resonance frequency as a function of the static external magnetic field
284 for all samples (N) of our series. Figure 4(a) shows the data of the thinnest sample in this study,
285 i.e., $N = 1$ or Au(20 nm)/Co(3 nm)/Au(3 nm). Only one mode is visible in the explored frequency
286 range, which corresponds to the uniform magnetization precession mode in the single Co layer
287 characterized by a uniform dynamic magnetization across its thickness. At the other end of our
288 sample series, Fig. 4(l) displays the frequency-field dependence of the thickest $N = 30$ sample. In
289 this case, many modes are visible. The modes are always labeled from bottom to top, i.e. from
290 lowest frequency (1st mode, red dots) to highest frequency (5th mode, green pentagons). As
291 mentioned above for a uniform single layer material, the detected FMR mode may be associated
292 with the uniform precession of the magnetization. However, as we will see in detail further, the
293 ML nature of the samples introduces a more complex scenario with a distribution of FMR
294 amplitudes along the structure. The higher-order modes (2nd to 5th) can be identified as “PSSW-
295 like” along the ML thickness. The observation of both odd and even modes implies that the mode
296 amplitude profiles are asymmetric with respect to the sample thickness. Intriguingly, by decreasing
297 the number of Co/Au repetitions from $N = 30$ backward, all the PSSW-like modes could still be
298 excited at higher frequencies when the same static field is applied. Importantly, higher order modes
299 have not been measured for $N < 6$. This might suggest an apparent suppression due to the thickness
300 driven reorientation transition from OOP to IP preferential magnetization orientation. Contrary, the
301 absence of the higher-order responses is associated to their shift in frequency into a range not
302 accessible by our experimental setup (>35 GHz).

303 Moreover, the separation in frequency between the modes increases with decreasing the
304 total ML thickness, which could be understood on the basis of the perpendicularly quantized spin
305 wave vector being inversely proportional to the total film thickness. However, not only the PSSW-
306 like modes are changing their resonance conditions upon changing N , but also the 1st mode. As
307 already discussed in Sec. IIB, magnetometry has revealed a magnetic crossover from OOP to IP as
308 a function of the total thickness, with the samples $N \geq 10$ being correctly described with the same
309 values of saturation magnetization M_S , anisotropy K_u and exchange stiffness A . Nonetheless, those
310 magnetic parameters were evaluated by an integral method (VSM) and XRD characterization has
311 shown that the diffraction angle corresponding to the Co/Au superstructure Bragg peaks [displayed
312 in Fig. 2(b)] shifts towards its bulk position with a steep increase between $1 \leq N \leq 10$. The angular

313 position is directly related to the accommodation of misfits across any interface between Co and
 314 Au due to strain relaxation. This would correspond to an evolution of the interface quality as a
 315 function of N , which is dramatic for $N \leq 10$. This is therefore translated to the dynamic properties
 316 of such structures, as the 1st mode purely shows, given that it is not affected by total thickness
 317 effects. Indeed, for $N > 10$, a small change of the frequency positions of the FMR mode is still
 318 appreciable: despite corroborating that statically all these samples have the same structural
 319 properties and in particular the same Co and Au thicknesses, thus leading to nearly identical volume
 320 averaged magnetocrystalline and interface anisotropy fields, the dynamic response reveals the
 321 existence of small monotonic changes as N (and hence the total ML thickness) increases. We will
 322 argue that these changes originated in structural inhomogeneities (in particular, strain) along the
 323 vertical direction of the Co/Au ML stack.

324 In order to model the investigated samples from a dynamic point of view, a more detailed
 325 approach has been applied with respect to the single effective layer model, which was sufficient to
 326 understand the thickness-dependent magnetization reorientation transition but cannot explain the
 327 dynamic response. The model includes a bilinear interlayer exchange coupling throughout the Au
 328 interlayers [50,51]. Thus, the interaction between two consecutive Co layers ν and η is given by:

$$\epsilon_{\text{int}} = -J \frac{\mathbf{M}^{(\nu)} \cdot \mathbf{M}^{(\eta)}}{M_s^{(\nu)} M_s^{(\eta)}} \quad (2)$$

329 with J being the interlayer exchange constant. Although it is not necessary to understand the static
 330 properties of the investigated MLs, here the model includes surface anisotropies that are known to
 331 be induced by any Au/Co interface [52,53]. For the two outer Co layers (top-most and bottom-
 332 most) the effective anisotropy field is denoted by H_a^0 , while H_a corresponds to the effective
 333 anisotropy field of the inner Co layers. Moreover, given that we are interested in the OOP saturated
 334 regime promoted by sufficient external magnetic field, we assume that the equilibrium
 335 magnetization of all layers is pointing along the normal axis. The time evolution of the
 336 magnetization is determined by the Landau–Lifshitz (LL) equation of motion [31], namely

$$\dot{\mathbf{M}}^{(\nu)}(\mathbf{r}, t) = -\mu_0 \gamma \mathbf{M}^{(\nu)}(\mathbf{r}, t) \times \mathbf{H}^{e(\nu)}(\mathbf{r}, t) \quad (3)$$

337 Here, the dot denotes the time derivative, γ is the absolute value of the gyromagnetic ratio,
 338 $\mathbf{M}^{(\nu)}(\mathbf{r}, t)$ is the magnetization, and $\mathbf{H}^{e(\nu)}(\mathbf{r}, t)$ is the effective field of the Co layer ν , respectively.
 339 In the linear approximation, both the magnetization and the effective field are written as

340 $\mathbf{M}^{(v)}(\mathbf{r}, t) = M_s^{(v)} \hat{z} + \mathbf{m}^{(v)}(\mathbf{r}, t)$ and $\mathbf{H}^{e(v)}(\mathbf{r}, t) = \mathbf{H}^{e0(v)} + \mathbf{h}^{e(v)}(\mathbf{r}, t)$. Note that the z -axis is
 341 aligned parallel to the normal axis, which corresponds to the equilibrium direction. In terms of the
 342 magnetization components, the LL equations of motion are

$$i \frac{\omega}{\mu_0 \gamma} m_x^{(v)} = -m_y^{(v)} H_z^{e0(v)} + M_s^{(v)} h_y^{e(v)} \quad (4)$$

$$i \frac{\omega}{\mu_0 \gamma} m_y^{(v)} = m_x^{(v)} H_z^{e0(v)} - M_s^{(v)} h_x^{e(v)} \quad (5)$$

343 where we assumed $\mathbf{m}^{(v)}(t) = \mathbf{m}^{(v)} e^{i \omega t}$ with $\omega = 2\pi f$, where f is the frequency. The spatial
 344 dependence of the magnetization has been omitted, since we are interested in the ferromagnetic
 345 resonance response, so that the wavelength of the spin waves is supposed to be very large and
 346 hence a coherent IP motion of the magnetic moments is expected in each Co layer. Now, the static
 347 z -component of the effective field is

$$H_z^{e0(v)} = H + H_a^{(v)} - M_s^{(v)} + \sum_{\eta} \left(d \mu_0 M_s^{(v)} \right)^{-1} J (\delta_{v-1} \delta_{\eta} + \delta_{v+1} \delta_{\eta}). \quad (6)$$

348 where H is the external field, $H_a^{(v)}$ is the effective anisotropy field (encompassing surface magnetic
 349 anisotropy and magnetocrystalline anisotropy contributions), $H_a^{(v)} = (H_K^{(v)} + H_S^{(v)})$, $M_s^{(v)}$ is the
 350 demagnetizing field of the uniform state, d is the thickness of the v -th Co layer (3 nm) and $\delta_{i,j}$ is
 351 the Kronecker delta function (0 if $i \neq j$ and 1 if $i = j$). The latter function indicates that the
 352 interlayer exchange coupling [see Eq. (2)] is approximated to be active on the next nearest
 353 neighbors only. On the other hand, the dynamic components of the effective fields are:

$$h_{x,y}^{e(v)} = \sum_{\eta} \left(d \mu_0 M_s^{(v)} M_s^{(\eta)} \right)^{-1} J m_{x,y}^{(\eta)} (\delta_{v-1} \delta_{\eta} + \delta_{v+1} \delta_{\eta}). \quad (7)$$

354 By inserting Eqs. (6) and (7) into Eqs. (4) and (5), the system can be solved numerically.

355 Figure 5 shows a direct comparison of FMR experimental data (dots) and theory (lines)
 356 evaluated at $\mu_0 H^{\text{OOP}} = -1.2$ T (saturated regime for any N), where the external field is applied
 357 normal to the sample. A systematic analysis was performed from the theoretical point of view to
 358 reach a good agreement with the FMR data and to justify the use of the proposed model. Typically,
 359 FMR is modeled under the assumption that the measured signal is coherent, i.e., the spatial extent
 360 of OOP features is small enough that the absorption corresponds to a single average OOP sample
 361 structure. By following this line of action, first we use a simple macrospin model to fit the
 362 experimental data [see Fig. 5(a)], where each Co layer has the same effective anisotropy field $\mu_0 H_a$

363 that is set by fitting the frequency of the 1st mode for $N=1$, while the outer layers have a different
 364 effective anisotropy field $\mu_0 H_a^0$ to take into account the effect of having a single neighboring
 365 exchange coupled layer.

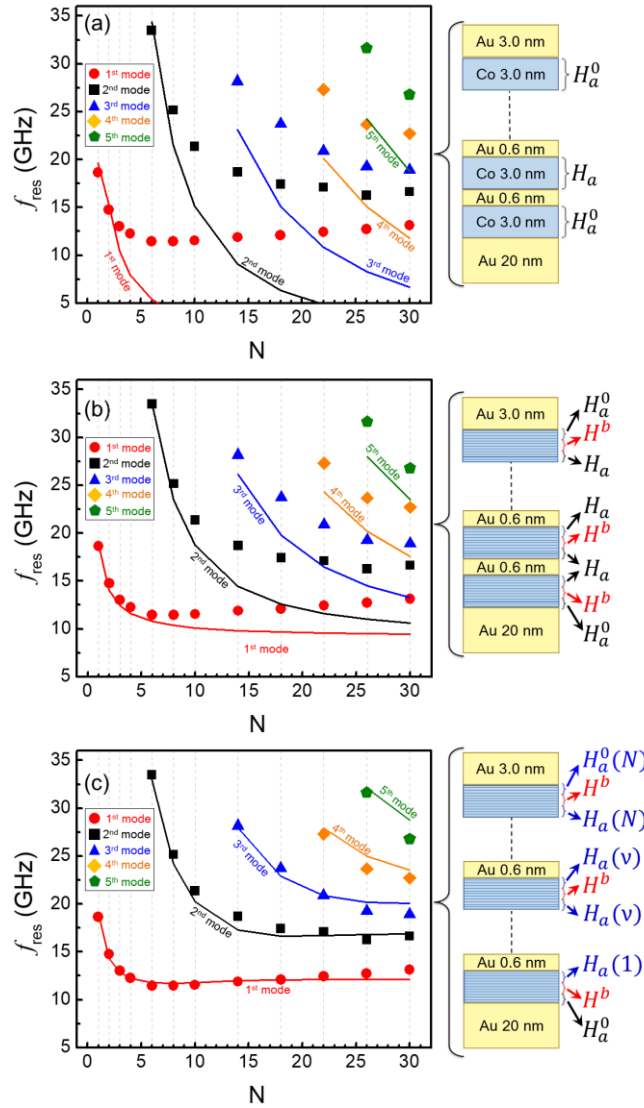


FIG. 5. (a)-(c) FMR measurements (dots) and theory (lines) evaluated at $\mu_0 H = -1.2$ T. Cases (a)-(c) show different approximations of the theoretical model (see text for details) that are summarized by the layer sketches on the right.

366 However, attempting to model the FMR data with this simple model results in a truly
 367 incorrect fit, as shown in Fig. 5(a). Still, we observe that the model could be useful to explain the
 368 behavior of the low-frequency mode (red dots) for the cases of one, two or three Co layers ($N = 1,$
 369 2, 3) since a reasonable agreement with the experimental results is obtained. Nevertheless, if N
 370 increases it is not possible anymore to find a good agreement between the FMR data and the
 371 theoretical calculation implying that such a simple macrospin model is not able to capture all the

372 details of the dynamic magnetization for larger N values. Moreover, this first simple approach
373 considers the surface anisotropy fields to be weak within the effective field $\mu_0 H_a$ of the entire 3-
374 nm-thick Co layer in the LL equation of motion, namely they are not assumed as located solely at
375 the Co/Au interfaces.

376 In order to consider the variations of the dynamic magnetization along the thickness and
377 the interfacially confined nature of the surface anisotropy fields, we use an approach that
378 subdivides the FM layers [54], so that $\mathbf{m}^{(v)}$ varies along the $d = 3$ nm thick individual Co layer.
379 Here, we define the anisotropy field $\mu_0 H_a$ uniquely within the sublayers that are next to the Au/Co
380 interfaces, where both magnetocrystalline and surface anisotropy contributions are acting, while
381 within the central region of the FM layer a different bulk anisotropy field parameter ($\mu_0 H^b$) is
382 considered. This latter term originates mainly from the magnetocrystalline anisotropy, which we
383 assume to dominate over the surface anisotropy in the central 3-nm-thick Co film region and is
384 enabled by the crystalline orientation of the ML stack as confirmed by the X-ray characterization
385 and the magnetometry measurements. This case is shown in Fig. 5(b) and the sketch next to it,
386 where we have subdivided each individual FM layer into 12 discrete sublayers with index SL .
387 Twelve SLs is the best compromise between accuracy of the fitting procedure and optimization of the
388 computational time. Thus, sublayers $SL = 1 - 4$ and $9 - 12$ possess an effective field $\mu_0 H_a$, whereas
389 the inner sublayers $5 \leq SL \leq 8$ are dominated by the effective field $\mu_0 H^b$ of magnetocrystalline
390 origin. Once again, the effective fields of the sublayers adjacent to the bottom-most and top-most
391 Co/Au interfaces are represented by a distinct fit parameter $\mu_0 H_a^o$, to account for the distinct
392 boundary conditions. The agreement between the calculations (lines) and the experimental data
393 (symbols) improves in general and substantially for repetition numbers up to $N = 6$ but fails to
394 reproduce the FMR modes for samples with $N > 6$.

395 Overall, we noted that for both models the theoretical results always underestimate the
396 experimentally determined FMR frequencies for high N . To account for this effect, we further
397 include a variation of the anisotropy field as a function of the repeating Co film unit within the ML
398 samples, such that the magnetic anisotropy varies as we move from the bottom Co layer ($v = 1$)
399 towards the topmost Co layer ($v = N$). This type of modeling is challenging, as without significant
400 constraints on the individual magnetic profiles, the number of free parameters can become
401 prohibitively large, rendering the results meaningless. Thus, we have chosen to build the model
402 based on useful information already determined from the previously discussed magnetometry
403 characterization as well as fitting the entire FMR data set with the same set of fitting parameters.

422 to assume that as more Co/Au bilayers are grown, the stress that builds up gradually attenuates. In
423 fact, we found that most of the strain relaxation occurs upon growth of the first ten Co/Au bilayers
424 [see Fig. 2(b)]. Thus, the density of the aforementioned misfit dislocations would naturally decrease
425 when going from the Co/Au interfaces located in the bottom part of the ML stack towards the top-
426 most interfaces.

427 It has been reported that misfit dislocations at interfaces in *hcp* Co films often consist of
428 *fcc*-like inclusions, thus locally impacting the magnetocrystalline anisotropy energy of Co films
429 due to the lower anisotropy of the *fcc*-like ordering as compared to *hcp* [58-60]. Thus, we can now
430 connect the gradient in the magnetic anisotropy field obtained from the fit to the dynamic model
431 with a gradient in the concentration of misfit dislocations at the Co/Au interfaces. In the bottom
432 region of the multilayer stack, a larger number of dislocations appears due to the larger stress build-
433 up, which then effectively lowers the magnetocrystalline anisotropy contribution to the $\mu_0 H_a(v)$
434 term. As growth progresses and strain is gradually released, fewer dislocations are introduced, and
435 thus, the magnetocrystalline anisotropy term to $\mu_0 H_a(v)$ being less affected by the presence of *fcc*-
436 like inclusions in the interfacial region, such that a larger magnetic anisotropy develops towards
437 the top region of the Co/Au ML stack.

438 It is interesting to consider that a variation of the $\mu_0 H_a(v)$ along the vertical direction of the
439 ML stack could also originate from a different surface anisotropy contribution as we go from one
440 Co/Au interface to another along the vertical direction. In fact, the surface waviness observed in
441 the TEM micrograph in Fig. 2(c) might suggest that the surface anisotropy is influenced by the
442 non-ideal, wavy interface geometry. TEM imaging does not show, however, a clear gradient of the
443 waviness when moving from the bottom-most to the top-most part of the stacks, such that we
444 associate the main contribution to the anisotropy field gradient from a perspective of strain
445 relaxation induced magnetocrystalline anisotropy reduction by the presence of dislocations.

446 The proposed model also allows for evaluating the depth profile of the dynamic response
447 amplitude. The corresponding dynamic magnetization profiles are shown in Fig. 7. In Fig. 7(a) the
448 case of one isolated FM layer ($N=1$) is shown where the orbits of the sublayers are depicted. The
449 red dots denote the $t=0$ point, i.e., they mark the excitation amplitude and phase for all modes at
450 the same time, while the orbit highlighted with yellow thick dots corresponds to the orbit of the 7-
451 th sublayer. As the number of layers increases, the orbits change radically as depicted in Figs. 7(b)-
452 (d), introducing an amplitude distribution throughout the layers while keeping them in phase. Due
453 to the different anisotropies at the top and bottom parts of the ML, the higher amplitude is mainly

454 located at the bottom part of the structure, where the local anisotropies are smaller. This behavior
 455 explains why the first mode (or low-frequency mode) does not significantly change at higher values
 456 of N . Indeed, by increasing N the first mode becomes gradually a mode that is confined at the
 457 bottom part of the sample. The excited mode profiles for $N = 30$ are illustrated in Figs. 7(e)–(f),
 458 which are showing the nonmonotonic amplitude profiles of the high order modes, whose
 459 distribution depends on the energy profile along the total thickness.

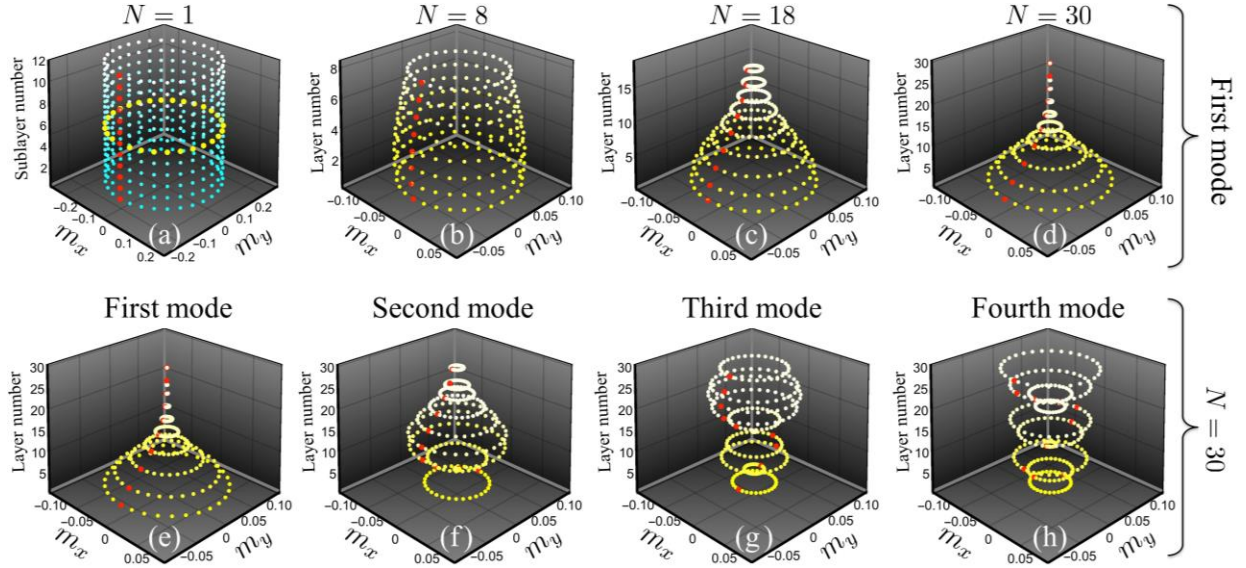


FIG. 7. In-plane dynamic magnetization orbits in z -direction across the sample thickness as a function of N . In cases (a)–(d) the first mode is illustrated for different N values, while the first four excited modes are shown in (e)–(h) for the case $N=30$. Dynamic magnetization components $m_{x,y}$ are expressed in arbitrary units. The rod dots mark the excitation amplitude and phase for all modes at the same time ($t=0$).

460 For high mode numbers the phase changes by 180° , giving them a spin wave character. Therefore,
 461 a PSSW-like mode in thinner ML films needs dramatically more energy, due to higher exchange
 462 energy needed for the same kind of wave.

463 IV. Conclusions

464 In this paper, we have successfully fabricated $[\text{Co}(3.0 \text{ nm})/\text{Au}(0.6 \text{ nm})]_N$ multilayer films with a
 465 magnetic anisotropy axis perpendicular to the ML plane. The RT magnetometry measurements for
 466 the samples with $N > 10$ reveal two very different magnetization reversal processes for external
 467 applied fields within and perpendicular to the film plane: namely (i) an instability-driven reversal
 468 process leading to the generation of parallel stripe domains for IP field orientation and (ii) a domain
 469 nucleation process that is hysteretic in nature for OOP field orientation [30]. Our N -dependent
 470 study shows a gradual shrinking of the nucleation regime with decreasing N , so that at sufficiently
 471 low N only the instability-driven second-order phase transition occurs even for OOP applied fields.

472 The disappearance of the nucleation regime is driven by the strongly thickness-dependent balance
473 between magnetic anisotropy and magnetostatic energies and occurs before the effective OOP
474 anisotropy energy becomes too weak to support a stripe domain state altogether, i.e., it occurs while
475 the stripe domain instability is still dominating the magnetization reversal process by
476 spontaneously forming stripe domains at a critical applied field without a real bubble nucleation
477 and stripe propagation process. By further reducing the number of Co/Au repetitions below $N < 10$,
478 we observe a characteristic in-plane easy-plane magnetic behavior.

479 Moreover, we have demonstrated that such MLs provide an efficient platform for the
480 excitation of PSSWs, whose characteristics turns out to be strongly dependent on the material
481 modulation along the total thickness induced by the multilayering itself. Specifically, we identify
482 various dynamic excitations for statically OOP saturated multilayer state, which are excited under
483 rf fields. Therefore, in contrast to homogeneous thick films or thin multilayer systems, the
484 investigated structures allow for a full comprehension of the conversion of periodic material and
485 magnetic properties along the sample stack into a vertical array of two-dimensional (exchange-
486 based) pinning sites for the PSSW modes within the film. In this context, in particular the decreased
487 exchange across the Au interlayers seems to be responsible for the more complex dynamic
488 behavior, as compared to systems with a less strong vertical exchange modulation, such as for
489 example in typically investigated Co/Pt and Co/Pd ML systems. Moreover, we demonstrate that
490 PSSW modes provide a handle to study the magnetic interactions and their modulation along the
491 thickness in such ML systems, offering a unique platform for full tunability of the mode frequencies
492 and amplitude profiles. Our observations can be generalized for different multilayered
493 ferromagnetic materials exhibiting OOP preferential orientation of magnetization. Overall our
494 work illustrates the substantial relevance of PMA thin films for a very detailed understanding of
495 high-frequency spin wave excitations in artificially multilayered systems.

ACKNOWLEDGMENTS

L. F. is very thankful to K. Potzger for the support with experimental facilities. R. A. G. acknowledges the financial support from Fondecyt Iniciación, Grant 11170736 and Basal Program for Centers of Excellence, Grant FB0807 CEDENNA, CONICYT. CzechNanoLab project LM2018110 funded by MEYS CR is gratefully acknowledged for supporting the experimental work at CEITEC Nano Research Infrastructure. This work has received funding from the European Union's Horizon 2020 research and innovation program under the Marie Skłodowska-Curie Actions and it is co-financed by the South Moravian Region under grant agreement No. 665860. The use of the HZDR Ion Beam Center TEM facilities is gratefully acknowledged.

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- [36] Despite our XRD investigation not being able to exclude the presence of *fcc* stacking faults in Co, the *hcp* lattice of Co was additionally verified via in-plane diffraction measurements, by setting the incident and diffracted beams nearly parallel to the sample surface, in which only Co(11 $\bar{2}$ 0), Co(10 $\bar{1}$ 0) and Au(220) reflections were measured for our samples. The in-plane diffraction experiments confirmed a fiber-textured ordering of the multilayers, with randomly in-plane oriented crystallographic planes.
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