

UC Berkeley

UC Berkeley Previously Published Works

Title

Coexistence of soft and hard magnetic phases in single layer amorphous Tb-Co thin films

Permalink

<https://escholarship.org/uc/item/01v746cb>

Journal

Journal of Applied Physics, 131(3)

ISSN

0021-8979

Authors

Ceballos, A
Charilaou, M
Molina-Ruiz, M
[et al.](#)

Publication Date

2022-01-21

DOI

10.1063/5.0074314

Copyright Information

This work is made available under the terms of a Creative Commons Attribution-NonCommercial License, available at <https://creativecommons.org/licenses/by-nc/4.0/>

Peer reviewed

1 Coexistence of soft and hard magnetic phases in single layer amorphous 2 Tb-Co thin films

3 A. Ceballos,¹ M. Charilaou,² M. Molina-Ruiz,³ and F. Hellman^{1,3,4}

4 ¹*Department of Materials Science and Engineering, University of California, Berkeley, California 94720,*
5 *USA*

6 ²*Department of Physics, University of Louisiana, Lafayette, Louisiana 70504, USA*

7 ³*Department of Physics, University of California, Berkeley, California 94720, USA*

8 ⁴*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720,*
9 *USA*

10 (*Electronic mail: michalis.charilaou@louisiana.edu)

Amorphous(*a*-), ferrimagnetic Tb-Co thin films prepared with a thin Ta underlayer and either a Ta or Pt overlayer show evidence of both a soft and a hard magnetic phase. At room temperature the films exhibit conventional ferromagnetism, but low temperature magnetometry measurements reveal the decoupling of the two magnetic phases with decreasing temperature due to increased anisotropy energy of the hard layer at lower temperatures. Decreasing the film thickness to 2 nm, slightly above the superparamagnetic limit found at 1 nm, a soft, low density phase was isolated and found to be present in all the films as confirmed with x-ray reflectivity and Rutherford backscattering spectrometry measurements. For greater thicknesses, the bottom layer retains its soft magnetic nature, while the remainder of the film is denser and has strong perpendicular magnetic anisotropy, leading to exchange-spring behavior when the anisotropy becomes large, either at low temperatures or via a Pt overlayer which adds a strong interfacial anisotropy to the layer. Micromagnetic simulations of a soft/hard bilayer model with the experimentally-determined anisotropy and magnetization parameters into a soft/hard bilayer model, reproduced the experimental hysteretic behavior very well. These findings demonstrate how the magnetic state and the response of *a*-Tb-Co films to external fields can be controlled, providing a high degree of tunability that is promising for high-performance nanoscale devices.

11 I. INTRODUCTION

12 With increasing pressure for faster, denser and more
13 energy-efficient magnetic data and logic devices comes the
14 necessity to study reduced dimensions and interfacial phe-
15 nomena in magnetism. Amongst the many magnetic mate-
16 rials being proposed, ferrimagnetic rare-earth (RE) - transi-
17 tion metal (TM) alloys have been receiving increasing atten-
18 tion due to their wealth of magnetic properties, situating them
19 at the center of many proposed magnetic devices. Studies
20 of these systems include spin-orbit torque measurements of
21 ferro(i)magnet/heavy metal (HM) bilayer systems;¹⁻⁵ imag-
22 ing of exotic spin textures such as skyrmions in multilay-
23 ered HM/RE-TM alloys,⁶ and ultrafast magnetization dynam-
24 ics in Gd-Fe-Co and Gd-Co alloys.⁷⁻¹¹ All of these stud-
25 ies take advantage of amorphous RE-TM alloys' growth-
26 induced perpendicular magnetic anisotropy (PMA), relatively
27 high Curie temperature, and tunability of their magnetic prop-
28 erties, including the magnetic anisotropy, dipolar coupling,
29 Dzyaloshinskii-Moriya interaction, damping and spin-orbit
30 coupling by modifying the composition, growth temperature,
31 anneal temperature and by engineering the interface.^{12,13}
32 This tunability makes these systems highly desirable testbeds
33 at the forefront of applied and basic research.

34 The amorphous structure of these RE-TM alloys introduces
35 an added challenge when it comes to characterizing the struc-
36 ture of ultra thin films. In addition, very thin films may exhibit
37 superparamagnetic effects, and interfacial or surface effects be-
38 come important and may dominate the magnetism of the film
39 itself.

40 This study shows that amorphous, ferrimagnetic Tb-Co thin
41 films, which appear to be conventional ferrimagnets at room

42 temperature, exhibit unexpected magnetic hysteretic behav-
43 ior at low temperatures, manifested in the form of magnetiza-
44 tion steps, due to the coexistence of a low/high density bilayer
45 that results from growth effects. This finding highlights how
46 the growth conditions can be tuned to deliberately obtain a
47 tailored exchange-spring type magnetic state in RE-TM thin
48 films, which is promising for nanoscale applications.

49 II. GROWTH OF THIN FILMS

50 The thin films were deposited using a confocal DC mag-
51 netron sputtering system by co-sputtering separate Tb, Co,
52 Ta and Pt targets. The samples consisted of trilayers of Ta(2
53 nm)/*a*-Tb₁₈Co₈₂(*t* nm)/Pt or Ta(7 nm) with *t* ranging from 2
54 to 16 nm for the Ta-capped samples, and 2 - 4 nm for the
55 Pt-capped samples. All samples were grown at room temper-
56 ature in 1 mTorr of Ar and a base pressure of 8×10^{-8} Torr at
57 a growth rate of ~ 1.0 Å/s. Samples were grown on 300 nm of
58 amorphous silicon nitride (*a*-SiN_x) on Si substrates, coated
59 with 50 nm thermal oxide (SiO₂), and were determined to be
60 amorphous via electron diffraction in a Transmission Electron
61 Microscope. The deposition of Ta onto *a*-SiN_x ensures that the
62 Ta underlayer is amorphous, as determined by fluctuating-
63 electron microscopy¹⁴, and it serves as an adhesion layer to
64 enable the growth of flatter *a*-Tb-Co films¹⁵.

65 The magnetic properties of the films were investigated us-
66 ing a SQUID magnetometer in a temperature range of 10-400
67 K. The layer thickness and densities were determined via X-
68 ray reflectometry (XRR) and Rutherford backscattering
69 (RBS). XRR measurements were conducted with a PANalyt-
70 ical X-Ray diffractometer (Cu-K α radiation, 0.154 nm). The

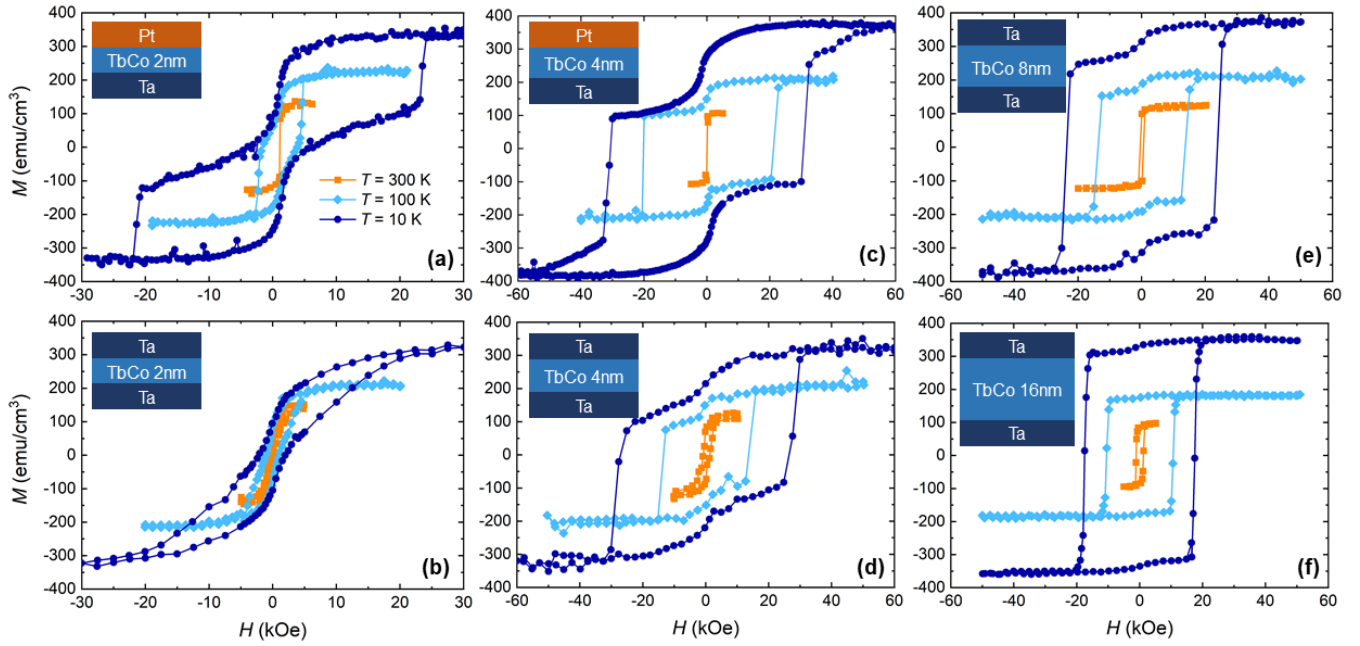


FIG. 1. Out of plane magnetization hysteresis for α -Tb₁₈Co₈₂ samples at room temperature (orange squares), 100 K (light-blue diamonds) and 10 K (dark-blue circles). Panels (a) and (c) correspond to the 2 and 4 nm Pt-capped samples while (b), (d), (e) and (f) correspond to the 2 – 16 nm Ta-capped samples, respectively, as shown in the inset schematics for each panel. Coercivity increases with decreasing temperature and steps are seen in all samples at 10 K except the Ta-capped 2 nm film.

71 RBS spectra were recorded in the Cornell geometry with a 3
 72 MeV $^4\text{He}^+$ ion beam incident at 35° from the sample's normal
 73 and backscattered into a detector located at 165° from the inci-
 74 dent beam direction. Higher angles of incidence were tested
 75 in order to improve the surface sensitivity but it was found that
 76 the Ta or Pt peak considerably overlapped with the Tb peak.
 77 Thus 35° was chosen as the optimal angle of incidence.

78 III. RESULTS AND DISCUSSION

79 The α -Tb-Co films' chemical composition was measured
 80 to be 18.0 ± 0.5 at.% Tb, 82 ± 1 at.% Co via RBS. At this
 81 composition the ferrimagnetic samples possess a compensa-
 82 tion temperature of $T_{\text{comp}} \approx 150\text{--}200$ K¹⁵; at room tempera-
 83 ture the net magnetization is parallel to the Co sublattice and
 84 is said to be Co-dominant, while below T_{comp} the magnetiza-
 85 tion is Tb-dominant. Figure 1 shows the out of plane (OOP)
 86 magnetic hysteresis for all samples. At 300 K every sam-
 87 ple exhibits hysteretic behavior indicative of a homogeneous
 88 magnetic film. However upon decreasing the temperature to
 89 100 K a step in the hysteresis is observed which is indicative
 90 of the coexistence of a decoupled soft and a hard magnetic
 91 phase. All samples exhibit this low temperature step except
 92 for the Ta-capped 2 nm film as shown in Fig. 1b. The mag-
 93 nitude of the step in the hysteresis is significantly reduced in
 94 the Ta-capped 8 and 16 nm samples as seen in Figs. 1e and 1f
 95 respectively. This will be shown to be associated with a soft
 96 magnetic layer that is unaffected by the total film thickness.
 97 Figs. 1a and 1c show the magnetization of the Pt-capped sam-

98 ples which show similar behavior to their Ta-capped counter-
 99 parts. These measurements also show that the coercivity in-
 100 creases with decreasing temperature and that the films possess
 101 perpendicular magnetic anisotropy. The value $M_s = 100 \pm 15$
 102 emu/cm^3 at room temperature is in good agreement to values
 103 reported in the literature for the same composition^{16,17}. Im-
 104 portantly, the fact that all samples have the same M_s at each
 105 temperature, despite the different thickness, indicates that all
 106 films have the same composition, considering that M_s changes
 107 strongly with composition¹⁶.

108 The in-plane (IP) and OOP magnetization loops of the Ta-
 109 capped 2 nm film at 10 K are shown in Fig. 2. It reveals
 110 that unlike all other samples in this study, this sample pos-
 111 sesses no effective magnetic anisotropy as it has no preferred
 112 magnetization direction. Film thickness of 1 nm results in
 113 superparamagnetism (See Supplemental Fig. S1), and at 2
 114 nm the film overcomes the superparamagnetic limit. How-
 115 ever, despite achieving ferromagnetic order the sample lacks
 116 the strong PMA seen in the other samples as shown clearly in
 117 Fig. 3.

118 The intrinsic anisotropy constant (K_{ui}) is extracted from
 119 IP and OOP magnetization measurements using the relation
 120 $K_{\text{ui}} = H_u M_s / 2 + 2\pi M_s^2$ where H_u is the anisotropy field and M_s
 121 the saturation magnetization, where H_u is determined from the
 122 external field needed to saturate the samples in the hard-axis
 123 direction. It is seen that the anisotropy is influenced by three
 124 factors: increasing film thickness increases PMA, decreasing
 125 temperature increases PMA, and capping with Pt slightly en-
 126 hances the PMA. The zero effective anisotropy seen in the Ta-
 127 capped 2 nm sample is not due to K_{ui} being zero, but rather to

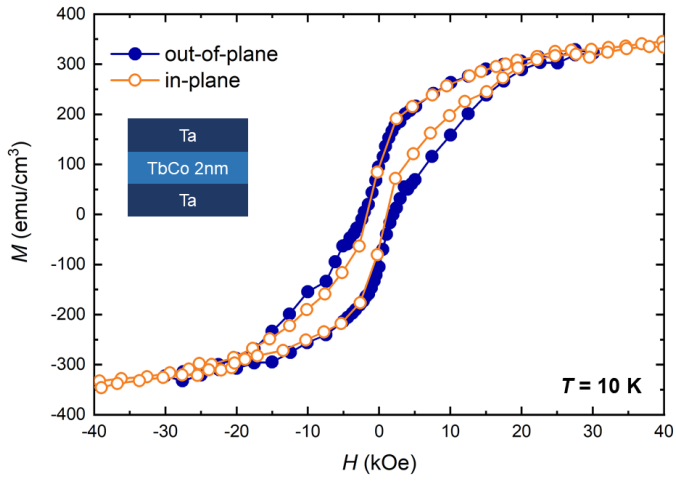


FIG. 2. Out of plane and in plane magnetization loops of the Ta-capped 2 nm sample shows no effective magnetic anisotropy, meaning that shape anisotropy is balanced by an intrinsic growth induced PMA of magnitude approximately 7×10^5 ergs/cc.

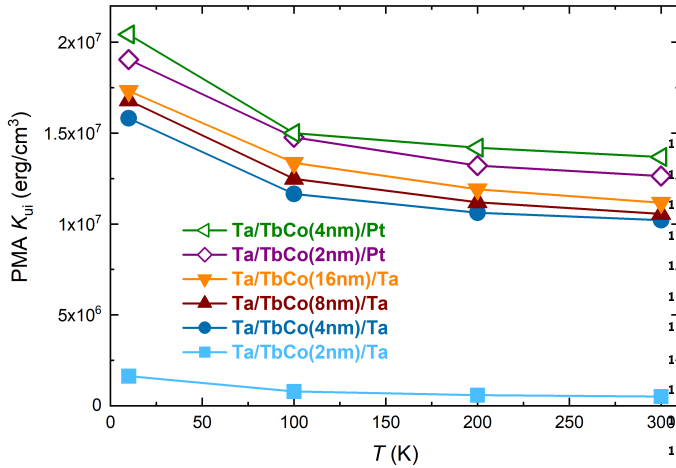


FIG. 3. Intrinsic anisotropy constant (K_{ii}) as a function of temperature. Decreasing the temperature results in increased PMA. PMA also increases with film thickness as well as by capping with Pt as seen in the Pt-capped series.

128 K_{ii} being very close in value to the shape anisotropy energy.

129 Structural characterization via XRR allows determination
 130 of sample thickness, roughness and density. XRR curves
 131 were analyzed using the GenX software,¹⁸ in which a layered
 132 model of the samples is composed as input and the GenX pro-
 133 gram varied the thickness, roughness and density of each layer
 134 until the difference between the simulation and the experi-
 135 mental data was minimized. For the Ta-capped samples the
 136 layered model consisted of air/TaO_x/Ta/*a*-Tb-Co/Ta/*a*-SiN_x
 137 whereas for the Pt-capped samples it consisted of air/Pt/*a*-Tb-
 138 Co/Ta/*a*-SiN_x. Fig. 4 shows the XRR curve of the Ta-capped 4
 139 nm sample and its accompanying simulation. Fig. 4a presents
 140 the simulation done with a single layer of *a*-Tb-Co and it is
 141 evident that it does not yield a very good fit. Fig. 4b shows
 142 the result of simulating a bilayer consisting of a lower density

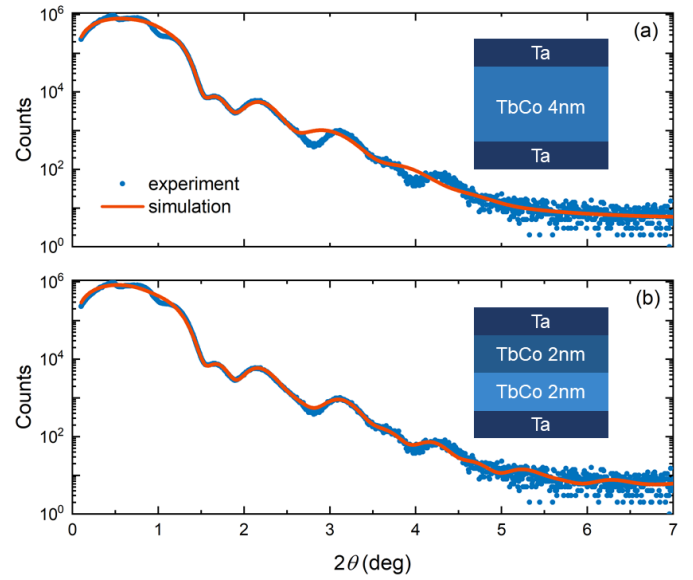


FIG. 4. XRR measurements on the Ta-capped 4 nm sample. a) Shows the inadequate fitting resulting from using a single 4 nm Tb-Co layer. b) Shows the improved fit when breaking the nominal 4 nm Tb-Co film into a bilayer of different atomic densities.

bottom layer and a higher density top layer of *a*-Tb-Co, result-
 ing in a much improved fit. This bilayer approach had to be
 implemented in all films except for the 2 nm Ta-capped and
 Pt-capped samples in order to achieve good fits. Comparing
 the densities of the bottom layers across all samples revealed
 that the bottom layer was always ~ 2 nm thick with a density
 that matched that of the 2 nm films as will be discussed below.

To further characterize the low/high density bilayer, the
 samples were probed with RBS. RBS uses MeV helium ions
 to depth profile information about a thin film. The helium
 ions recoil from classical collisions with the film's atoms, and
 the energy of the recoiled atoms can be modeled to tell the
 atomic composition and thickness or density of a thin film.
 Either thickness or density is required for proper RBS fitting
 and calculating density requires a precise measure of a film
 thickness which in this case is provided by the XRR analysis.
 Thus XRR and RBS are powerful and complementary
 techniques. The RBS spectra were analyzed using the SIM-
 NRA software¹⁹ and the densities from both XRR and RBS
 are plotted in Fig. 5, where the "Bottom Layer" and "Top
 Layer" labels denote the separate densities of the bilayer. To
 obtain the density from RBS the SIMNRA simulation's areal
 density was divided by the film thickness as extracted from
 XRR. The XRR bilayer thickness, roughness and density are
 tabulated in Table I as well as the RBS density and RMS sur-
 face roughness as measured with an atomic force microscope
 (AFM) as seen in supplementary materials.

The XRR data in Fig. 5 shows that a bilayer was found for
 all samples except the 2 nm Ta-capped and Pt-capped sam-
 ples, which possess a density that is similar to that found in
 the initial ~ 2 nm in all the other films as seen within the
 region labeled "Bottom Layer". Table I shows that the thick-
 ness of this lower density bottom layer is in the 1.4 - 1.8 nm
 range.

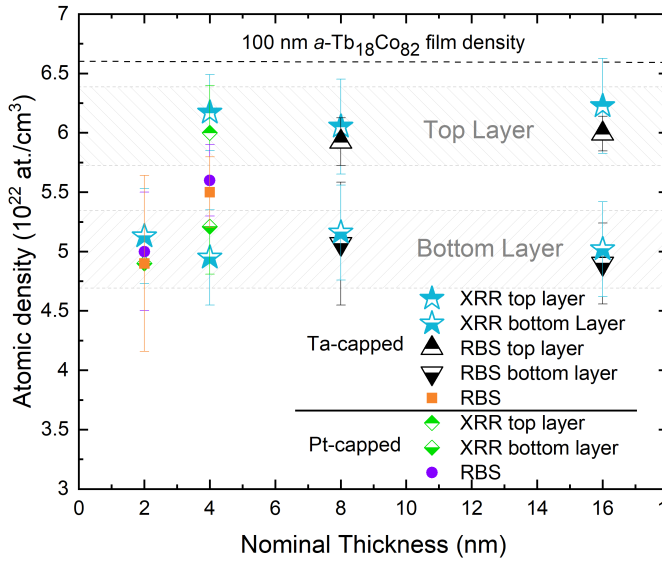


FIG. 5. Atomic density as a function of the nominal film thickness as extracted from both XRR and RBS. The rectangular regions are guides to the eyes to show the agreement of both XRR and RBS in determining the film density and confirming the bilayer model. The RBS density of the 4 nm samples lies between the two regions because it is averaging the low/high density values of the bilayer as discussed in the main text. Points labeled “RBS” indicate that a single layer was used for the fitting.

The RBS data confirmed the presence of a bilayer in the 8 and 16 nm Ta-capped samples, and a single layer fit was sufficient for the Ta-capped and Pt-capped 2 - 4 nm samples. The density extracted from RBS, is in good agreement with the density found via XRR. The RBS density found for the 4 nm samples is in between that found with a bilayer model in XRR, suggesting that a bilayer is present and its densities are being averaged in RBS. Both XRR and RBS also found a near constant density for the top layer, as seen in the gray box labeled “Top Layer” depicting a somewhat flat density trend with increasing film thickness. The dotted line at the top of Fig. 5 is the measured density of a 100 nm thick $a\text{-Tb}_{18}\text{Co}_{82}$ film, marking the density limit and showing that the density increases only slightly with increasing thickness beyond a thickness of 10 nm.

In short, these experiments show that at the growth conditions discussed in section II, amorphous Tb-Co films grow with an initial low-density layer followed by a layer that is 20% denser. The increase in density, as the film thickness increases above 2 nm, yields a bilayer system with a soft base layer and hard top layer with PMA. At room temperature, the two layers are coupled and exhibit conventional ferromagnetic hysteresis, but at low temperature they effectively decouple and the $M(H)$ curve exhibits distinct magnetization steps, the magnitude of which decreases with increasing thickness. Importantly, it should be noted that the steps in the $M(H)$ curves appear only below the ferrimagnetic compensation temperature (T_{comp}) where the Tb moments are dominant. Considering that the magnetic moment of Tb is roughly 7 times larger than that of Co¹⁶, the Zeeman energy becomes larger at $T < T_{\text{comp}}$

therefore the soft layer follows the external field at the cost of the exchange energy between the layers. That, and the porosity found in the films, accounts for the observation of the steps in the $M(H)$ curves below T_{comp} . In order to support this interpretation, we calculated an estimate of the external field strength required to justify the penalty in the exchange energy at the interface. The energy contributions for the soft layer can be described as

$$E = -Jx_{\text{Co}}^2S^2 - \tilde{J}x_{\text{Co}}x_{\text{Tb}}S\sigma - g_{\text{Co}}x_{\text{Co}}SB\mu_B - g_{\text{Tb}}x_{\text{Tb}}\sigma B\mu_B + \tilde{K}(x_{\text{Co}}^2S_z^2 + x_{\text{Tb}}^2\sigma_z^2) \quad (1)$$

where S and σ is the spin of Co and Tb, respectively, J is the Co-Co exchange interaction, \tilde{J} is the Co-Tb exchange interaction, g is the Landé factor, μ_B is the Bohr magneton, \tilde{K} is an effective shape anisotropy due to dipole-dipole interactions, B is the external field, and $x_{\text{Co}} = 0.82$, $x_{\text{Tb}} = 0.18$ reflect the composition of the system. For the soft layer to rotate towards the external field, there is a penalty in the exchange interaction at the interface with the hard layer, whereas the Zeeman energy of the N atomic layers in the soft layers is satisfied, with σ in the direction of B and S in the opposite direction, i.e., ferrimagnetic order is maintained in the soft layer. This yields $B \geq Jx_{\text{Co}}^2S^2 / (g_{\text{Tb}}x_{\text{Tb}}\sigma - g_{\text{Co}}x_{\text{Co}}S)\mu_B N$, and based on values from our experiments and from the literature¹⁶ ($S = 0.7$, $\sigma = 6.0$, $J = 1 \times 10^{-14}$ erg, $g_{\text{Co}} = 2.2$, $g_{\text{Tb}} = 1.75$, $N \approx 7$), we obtain a value of $B \approx 8$ kOe, which is in reasonable agreement with the field strength at which the step in the $M(H)$ curve was observed in the experiments. At room temperature, where Co is the dominant magnetic moment, the field strength needed to rotate the soft layer becomes 28 kOe, and therefore no steps in the $M(H)$ curves would occur.

Hence, this effect is a result of the ferrimagnetic order in the films and the fact that energy contributions change with temperature as the relative sublattice magnetizations change. To further support our interpretation that the steps in the $M(H)$ curves is the result of the soft/hard layer system, we performed micromagnetic simulations.

IV. MICROMAGNETIC SIMULATIONS

In the micromagnetic continuum approximation, the total energy density in our system can be written as

$$E = A \sum_{k=x,y,z} (\nabla m_k)^2 - K_{\text{ui}}(m_z)^2 - 2\pi M_s \mathbf{m} \cdot (2\mathbf{H} + \mathbf{H}_{\text{dm}}) \quad (2)$$

where $\mathbf{m} = \mathbf{M}/M_s$ is the local magnetization unit vector, A is the exchange stiffness, \mathbf{H} is the external field, and \mathbf{H}_{dm} is the local demagnetizing field due to dipole-dipole interactions. Hysteresis loops were simulated by sweeping the external field and minimizing the energy using the steepest conjugate gradient method in Mumax3²⁰.

The modeled system consists of two 40 nm by 40 nm cuboids of and a fixed thickness of 2 nm for the bottom layer to represent the low density layer, and variable thicknesses for

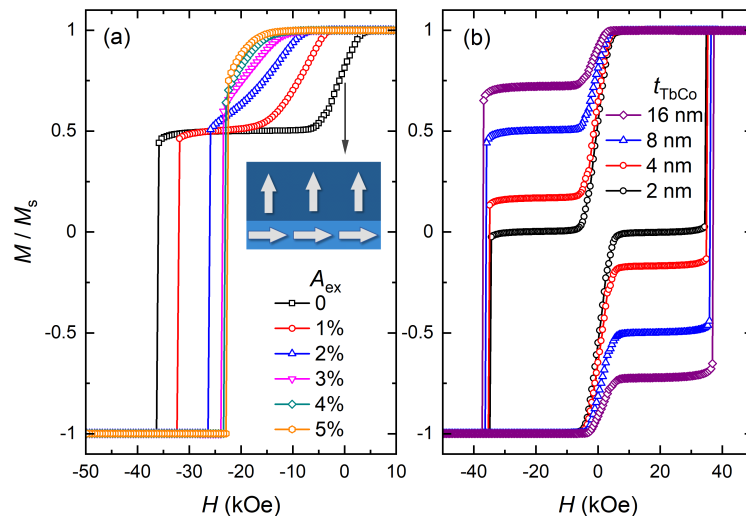


FIG. 6. Simulated magnetization reversal of a Tb-Co bilayer consisting of a soft 2 nm base layer and a hard top layer: (a) shows the $M(H)$ for a film with $t_{\text{TbCo}} = 8$ nm with different strengths of the coupling between the two layers, and (b) shows the $M(H)$ loop of four different Tb-Co thicknesses, where the two layers are fully decoupled. The hysteresis loops reproduce the experimental observations.

the upper layer ranging from 2 - 16 nm. The size of 40 nm is based on the average grain size as measured via AFM on an uncapped 2 nm thick a -Tb₁₈Co₈₂ film as shown in supplementary materials. The two regions were modeled to have the same value of $M_s = 400$ emu/cm³ (4×10^5 A/m) and exchange stiffness of $A = 2 \times 10^{-6}$ erg/cm (2×10^{-11} J/m). The layers only differed in the anisotropy constant, with the upper layer having a PMA of $K_{\text{ui}} = 10^7$ erg/cm³ (10^6 J/m³) and the bottom layer having $K_{\text{ui}} = 0$. These values were extracted from the experiments discussed above, with the exception of A , which was estimated based on the Curie temperature of the films, which was extrapolated from Ref.¹⁶ to be $T_C \sim 900$ K, and comparing to the values for pure Co ($T_C = 1400$ K, $A_{\text{Co}} = 2.8 \times 10^{-11}$ J/m)²¹, we scaled the exchange stiffness with $A = A_{\text{Co}}(T_{\text{TbCo}}/T_{\text{Co}})$. While this is only a rough estimate, it provides the correct order of magnitude. The cell size was kept at 0.5 nm, but occasional checks with different cell sizes were done to verify numerical stability.

In such a soft/hard bilayer, the exchange between the two layers plays a very important role in the magnetization reversal process. Even though micromagnetics, as a continuum approximation, cannot account for the ferrimagnetic atomistic effects discussed above, it offers a platform to investigate the effective magnetic state in such a system. As seen in Figure 6(a), which shows the simulated $M(H)$ for a film with $t = 8$ nm, when the bilayer is fully decoupled, a step occurs, similar to those observed experimentally. Specifically, as the field decreases, the magnetization of the soft layer starts decreasing before the field direction is reversed, due to the shape anisotropy of the soft layer. At zero field, the magnetization of the soft layer is in the in-plane configuration, where that of the hard layer is along the perpendicular direction, as shown by the inset to Fig. 6(a). When the field direction is reversed ($H < 0$), the magnetization of the soft layer gradually rotates towards the direction of H , and the net magnetization decreases until the field is $H = -6$ kOe, which is in good

agreement to the estimate noted above. The magnetization remains nearly constant with further increasing the opposing field, until it reverses at a coercive field of -36 kOe. With increasing inter-layer exchange, the step in the $M(H)$ curve becomes smoother and with 4–5% inter-layer exchange, it vanishes completely.

Comparing these findings with the measured $M(H)$ curves shown in Fig. 1, where a step is evident and the magnetization decreases before the external field is zero, we conclude that the bilayers are effectively decoupled at low temperature. Given that, we simulated decoupled bilayer systems with different thickness of the top layer, as shown in Fig. 6(b), which again agrees with the experimental observations, i.e., with increasing thickness the magnitude of the step decreases, as the contribution of the soft layer becomes increasingly smaller.

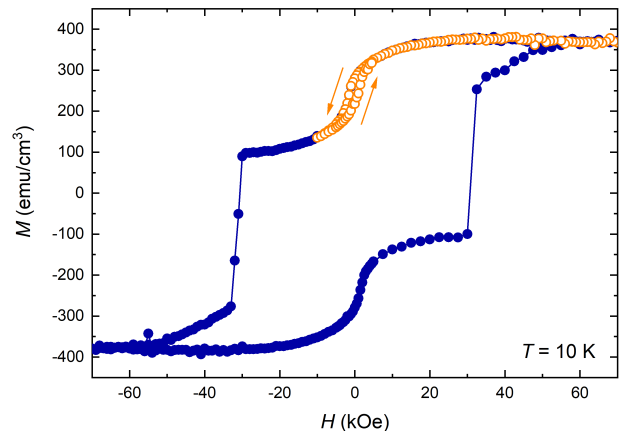


FIG. 7. Magnetization recoil curves illustrating the reversal of the soft, low density, layer of a 4 nm Pt-capped a -Tb₁₈Co₈₂ film at 10 K.

A soft/hard bilayer is a common recipe for exchange-spring systems which have aroused much interest in the

303 past two decades because of their potential in achieving gi- 358
 304 ant energy products as well as their rich variety of mag- 359
 305 netic behaviors.^{22–26}. Exchange-spring systems are based 360
 306 on interfacial exchange coupled soft and hard ferromagnetic 361
 307 nanocomposites or layers, that combine the high magnetiza- 362
 308 tion of the soft phase with the high anisotropy of the hard 363
 309 phase to attain a high energy product. 364

310 Considering that the soft and hard phases are only exchange 365
 311 coupled at the interface, the reorientation of the soft, low 366
 312 density layer should be fully reversible for fields below the 367
 313 switching field of the hard layer. This is the namesake of the 368
 314 “exchange-spring” term, indicating that as long as the applied 369
 315 field is below that of the hard layer, the magnetization of the 370
 316 soft layer will spring back to orient itself with the hard phase 371
 317 once the field is removed. In order to confirm that the soft/hard 372
 318 bilayer found in the films make up an exchange-spring system, 373
 319 magnetization recoil curves were measured. Recoil curves 374
 320 are obtained from the successive removal to remanence and 375
 321 reapplication of an increasingly negative field from the ma- 376
 322 jor demagnetization. The recoil curve shown in Fig. 7 was 377
 323 measured on the Pt-capped 4 nm α -Tb₁₈Co₈₂ sample, up to 378
 324 an opposing field strength of -10 kOe (1 T), and illustrates 379
 325 that the soft layer is reversible as expected of an exchange- 380
 326 spring system. A minor hysteretic behavior can be seen in 381
 327 the recoil curve, which is attributed to grain-boundary effects. 382
 328 Even though other engineered structures exhibit a stronger 383
 329 exchange-spring effect, it is striking to observe this behavior 384
 330 for a single-phase amorphous metal.

331 The successful reproduction of the experimental hystereses 385
 332 via micromagnetic simulations of a soft/hard bilayer, in con- 386
 333 junction with the density findings from XRR and RBS, leads 387
 334 to the following interpretation of the growth effect: the initial 388
 335 1–2 nanometers of the film grow at low density, whereas upon 389
 336 further growth the structure densifies, and the low/high den- 390
 337 sity is responsible for the soft/hard magnetic behavior of the 391
 338 films. The bilayer model is an approximation of what is bound 392
 339 to be a more gradual change as the deposition progresses. It 393
 340 is very likely that this type of growth effect is present in many 394
 341 other systems but its effects remain hidden due to experiments 395
 342 at room temperature, where the anisotropy is low enough to 396
 343 couple both phases so the magnetization reverses coherently 397
 344 and often on thicker films. This model accounts for the for- 398
 345 mation of the step in all films except for the 2 nm Pt-capped, 399
 346 which exhibits a formidable step at low temperatures. It is 400
 347 quite possible that the Pt interface is responsible for the for- 401
 348 mation of the step. Since it was seen that both Pt-capped sam- 402
 349 ples exhibit higher anisotropy than the Ta-capped ones, likely 403
 350 due to the Tb-Co/Pt interface. Thus the bilayer here would 404
 351 consist of soft Tb-Co and a hard Tb-Co/Pt interface. In the 405
 352 Pt-capped 4 nm sample, three steps are not observed because 406
 353 both the hard Tb-Co and the hard Tb-Co/Pt phases are close 407
 354 enough in anisotropy to reverse together.

355 V. CONCLUSION

356 In conclusion, the coexistence of a soft/hard bilayer in com- 408
 357 positionally homogeneous, single layer, amorphous, ferrimag- 409

netic Tb-Co thin films has been found to arise from a growth 410
 effect that creates a low density/high density bilayer which 411
 behaves as an exchange-spring magnet at low temperatures. 412
 This low density initial growth layer is likely a materials ef- 413
 fect associated with nucleation of clusters in the early phase 414
 of film growth. The lower density bottom layer is magneti- 415
 cally soft, and for film thickness near 1 nm, the film is su- 416
 perparamagnetic. For film thickness near 2 nm, the film is 417
 a soft ferromagnet. For thicker films, the 2 nm thick lower 418
 soft ferromagnet continues to influence the overall magnetic 419
 properties, causing steps in the magnetic hysteresis loops at 420
 temperatures below room temperature when the anisotropy of 421
 the hard layer becomes large. XRR and RBS provided di- 422
 rect structural evidence of this bilayer structure for all films 423
 thicker than 2 nm. This hidden low density layer is likely 424
 present in many other material systems, but the associated 425
 soft and hard magnetic phases will decouple only when the 426
 difference in anisotropy energy is large enough, which here 427
 was enhanced by the Pt interface, typically occurring at low 428
 temperatures. These findings demonstrate how even a single 429
 phase Tb-Co thin film can exhibit exchange-spring behavior 430
 due to differences in density, and interface, and suggest that 431
 such effects might be present in a wide variety of other sys- 432
 tems and have remained undetected. Hence, these observa- 433
 tions suggest that growth-induced density-differential struc- 434
 tures have the potential to yield high-performance nanoscale 435
 exchange-spring magnets.

385 SUPPLEMENTARY MATERIAL

386 See supplementary material for $M(H)$ curves of a 1 nm 436
 387 thick α -Tb-Co film at $T = 300$ K and 50 K showing su- 437
 388 perparamagnetic behavior (Fig. S1), and Atomic Force Mi- 438
 389 croscopy images of the Ta- and Pt-capped Tb-Co films (Fig. 439
 390 S2) as well as of a 2 nm un-capped film (Fig. S3).

391 ACKNOWLEDGMENTS

392 This work was primarily supported by the Director, Office 440
 393 of Science, Office of Basic Energy Sciences, Materials Sci- 441
 394 ences and Engineering Division, of the U.S. Department of 442
 395 Energy under Contract No. DE-AC02-05-CH11231 within 443
 396 the Nonequilibrium Magnetic Materials Program (KC2204). 444
 397 A.C. acknowledges support by the National Science Founda- 445
 398 tion under Grant No. DGE 1106400. M.C. acknowledges 446
 399 support from the Louisiana Board of Regents [contract Nr. 447
 400 LEQSF(2020-23)-RD-A-32].

401 DATA AVAILABILITY

402 The data that support the findings of this study are available 448
 403 from the corresponding author upon reasonable request. 449

404 ¹N. Roschewsky, T. Matsumura, S. Cheema, F. Hellman, T. Kato, S. Iwata, 450
 405 and S. Salahuddin, Appl. Phys. Lett. **109**, 112403 (2016).

- 406 ²J.-H. Kim, D.-J. Lee, K.-J. Lee, B.-K. Ju, H. C. Koo, B.-C. Min, and O.-J. 433
 407 Lee, *Sci. Rep.* **8**, 6017 (2018). 434
- 408 ³S.-G. Je, J.-C. Rojas-Sánchez, T. H. Pham, P. Vallobra, G. Malinowski, 435
 409 D. Lacour, T. Fache, M.-C. Cyrille, D.-Y. Kim, S.-B. Choe, M. Belmegue- 436
 410 nai, M. Hehn, S. Mangin, G. Gaudin, and O. Boulle, *Appl. Phys. Lett.* **112**, 437
 411 062401 (2018). 438
- 412 ⁴K. Ueda, M. Mann, C.-F. Pai, A.-J. Tan, and G. S. D. Beach, *Appl. Phys.* 439
 413 *Lett.* **109**, 232403 (2016). 440
- 414 ⁵H. W. Seung, S. Kim, D.-H. Kim, K.-J. Kim, T. Okuno, H. Yoshikawa, 441
 415 A. Tsukamoto, T. Moriyama, and T. Ono, *Appl. Phys. Lett.* **110**, 242405 442
 416 (2017). 443
- 417 ⁶R. Streubel, C.-H. Lambert, N. Kent, P. Ercius, A. T. N'Diaye, C. Ophus, 444
 418 S. Salahuddin, and P. Fischer, *Adv. Mater.* **30**, 1800199 (2018). 445
- 419 ⁷I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H. A. Dürr, T. A. 446
 420 Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, A. Tsukamoto, A. Itoh, 447
 421 A. Kirilyuk, T. Rasing, and A. V. Kimel, *Nature* **472**, 205 (2011). 448
- 422 ⁸C. D. Stanciu, A. Tsukamoto, A. V. Kimel, F. Hansteen, A. Kirilyuk, 449
 423 A. Itoh, and T. Rasing, *Phys. Rev. Lett.* **99**, 217204 (2007). 450
- 424 ⁹T. Ostler, J. Barker, R. Evans, R. Chantrell, U. Atxitia, O. Chubykalo- 451
 425 Fesenko, S. El Moussaoui, L. Le Guyader, E. Mengotti, L. Heyderman, 452
 426 F. Nolting, A. Tsukamoto, A. Itoh, D. Afanasiev, B. Ivanov, A. Kalash- 453
 427 nikova, K. Vahaplar, J. Mentink, A. Kirilyuk, T. Rasing, and A. Kimel, 454
 428 *Nature Comm.* **3**, 666 (2012). 455
- 429 ¹⁰M. L. M. Laliou, M. J. G. Peeters, S. R. R. Haenen, R. Lavrijsen, and 456
 430 B. Koopmans, *Phys. Rev. B* **96** (2017), 10.1103/PhysRevB.96.220411. 457
- 431 ¹¹J. Gorchon, C.-H. Lambert, Y. Yang, A. Pattabi, R. B. Wilson, S. Salahud- 458
 432 din, and J. Bokor, *Appl. Phys. Lett.* **111**, 042401 (2017). 459
- ¹²F. Hellman, M. Messer, and E. N. Abarra, *J. Appl. Phys.* **86**, 1047 (1999).
- ¹³F. Hellman, A. Hoffmann, Y. Tserkovnyak, G. S. Beach, E. E. Fullerton, 434
 C. Leighton, A. H. MacDonald, D. C. Ralph, D. A. Arena, H. A. Dürr, 435
et al., *Rev. Mod. Phys.* **89**, 025006 (2017). 436
- ¹⁴E. Kennedy, N. Erynolds, L. R. DaCosta, F. Hellman, C. Ophus, and M. C. 437
 Scott, *Appl. Phys. Lett.* **117**, 091903 (2020). 438
- ¹⁵A. Ceballos-Sanchez, *Characterization and manipulation of temperature-* 439
driven magnetic phenomena: magnetic transition in FeRh thin films and ul- 440
trafast magnetization reversal in α -Gd_{22-x}Tb_xCo₇₈ thin films, Ph.D. the- 441
 sis, University of California, Berkeley (2019). 442
- ¹⁶P. Hansen, C. Clausen, G. Much, M. Rosenkranz, and K. Witter, *J. Appl.* 443
Phys. **66**, 756 (1989). 444
- ¹⁷S. Alebrand, M. Gottwald, M. Hehn, D. Steil, M. Cinchetti, D. Lacour, E. E. 445
 Fullerton, M. Aeschlimann, and S. Mangin, *Appl. Phys. Lett.* **101**, 162408 446
 (2012). 447
- ¹⁸M. Björck and G. Andersson, *J. Appl. Cryst.* **40**, 1174 (2007).
- ¹⁹M. Mayer, Germany: Garching (1977).
- ²⁰A. Vansteenkiste, J. Leliaert, M. Dvornik, M. Helsen, F. Garcia-Sanchez, 450
 and B. Van Waeyenberge, *AIP Advances* **4**, 107133 (2014). 451
- ²¹A. Guimaraes, *Principles of Nanomagnetism* (Springer, 2009).
- ²²E. E. Fullerton, J. Jiang, and S. Bader, *J. Magn. Magn. Mater.* **200**, 392 452
 (1999). 453
- ²³W. Si, G. P. Zhao, N. Ryan, Y. Peng, F. J. Morvan, and X. L. Wan, *Sci.* 454
Rep. **5**, 16212 (2015). 455
- ²⁴A. Bill and H. B. Braun, *J. Magn. Magn. Mater.* **272-276**, 1266 (2004). 456
- ²⁵J. S. Jiang and S. D. Bader, *J. Phys.:* *Condens. Matter* **26**, 064214 (2013). 457
- ²⁶J. Jiang and S. Bader, *Scripta Mater.* **47**, 563 (2002). 458

	XRR			RBS	AFM
	Thickness (nm)	Roughness (nm)	Density (10^{22} at/cm ³)	Density (10^{22} at/cm ³)	Roughness (nm)
Ta-capped	lower layer / top layer	lower layer / top layer	lower layer / top layer	lower layer / top layer	
2 nm	1.6 ± 0.3	0.64 ± 0.07	5.1 ± 0.4	4.9 ± 0.7	0.49 ± 0.07
4 nm	$1.4 \pm 0.3 / 2.2 \pm 0.2$	$0.41 \pm 0.08 / 0.52 \pm 0.06$	$4.9 \pm 0.4 / 6.2 \pm 0.3$	5.5 ± 0.3	0.45 ± 0.06
8 nm	$1.6 \pm 0.3 / 6.2 \pm 0.2$	$0.47 \pm 0.09 / 0.61 \pm 0.08$	$5.2 \pm 0.4 / 5.1 \pm 0.4$	$5.1 \pm 0.5 / 5.9 \pm 0.2$	0.39 ± 0.05
16 nm	$1.5 \pm 0.3 / 14.2 \pm 0.2$	$0.39 \pm 0.06 / 0.48 \pm 0.07$	$5.0 \pm 0.4 / 6.2 \pm 0.4$	$4.9 \pm 0.3 / 6.0 \pm 0.2$	0.54 ± 0.07
Pt-capped					
2 nm	1.8 ± 0.3	0.59 ± 0.08	4.9 ± 0.4	5.0 ± 0.5	0.41 ± 0.06
4 nm	$1.4 \pm 0.3 / 2.3 \pm 0.3$	$0.42 \pm 0.07 / 0.63 \pm 0.08$	$5.2 \pm 0.4 / 6.0 \pm 0.4$	5.6 ± 0.3	0.52 ± 0.05

TABLE I. The thickness, roughness and density of the *a*-Tb-Co bilayers as obtained from XRR. Densities obtained from RBS are consistent with those found with XRR and confirmed the presence of a low density/higher density bilayer. The RMS surface roughness measured in AFM is consistent with that obtained via XRR.