Topologically stable helices in exchange coupled rare-earth/rare-earth multilayer with superspin-glass like ordering

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Existence of $2\pi$-planar domain walls (DWs) are often reported for transition metal-rare-earth (TM/RE) layered systems. The magnetization process of such two-dimensional randomly anisotropical system in the form of $2\pi$-DWs is directly correlated with topologically stable helices. Here, instead of TM/RE, we have investigated [Dy/Tb]$_{10}$ multilayers involving two different anisotropic layers of rare-earth/rare-earth (RE/RE). Using magnetization and susceptibility as function of temperature along with thermo-remanent magnetization measurements we have confirmed superspin-glass type of behavior within this RE/RE system. Additionally, an exchange bias field up to $-0.88$ kOe ($-88$ mT) was also revealed for such rare-earth based multilayers. Interestingly, using detailed analysis of the polarized neutron reflectometry profiles, we find evidences of superimposed helical magnetic configurations within both materials of Dy and Tb associated with spin-frustrated interfaces. Furthermore, magnetization fluctuations around the mean magnetization from vertically uncorrelated domains were observed with polarized off-specular neutron scattering. We believe that coexistence of helical ground states with superspin-glass-like ordering are fundamentally instrumental for topologically stability in RE/RE systems, which in principle, can be exploited in all-spin-based technology.

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thin films composed of rare-earth (RE) metals are known to form a helical antiferromagnetic (AF) structure due to competing magnetoelastic, crystalline anisotropy, and exchange interactions\textsuperscript{1,2}. These interactions can lead to topologically stable spin configurations, which are virtually indispensible in all-spin-based technology\textsuperscript{3}. Numerically, Vedmedenko et al. have shown that magnetic states corresponding to modulated helices with integer number of twists, commensurate with the chain’s length, are topologically stable\textsuperscript{4}. Such stability comes without the presence of chiral Dzyaloshinskii-Moriya interaction relevant in skyrmions, where its shape protects it from trivial unwinding. The quantized energy spectrum within finite magnetic chains results in stabilizing topologically protected configurations of helices. Such helices, initially twisted in an external magnetic field, stay stable even without the presence of a field as they are created by exchange interaction and does not allow a trivial unwinding\textsuperscript{5}.

Using polarized neutron reflectivity (PNR) measurements, it was shown earlier that AF-coupling at a pure transition metal-rare earth (TM/RE) (e.g., Fe/Tb) interface facilitates the formation of $\pi$ or $2\pi$ planar domain walls (DWs)\textsuperscript{6,7}. Within a multilayer, formation of such $2\pi$-DWs resemble a helical form leading to double hysteresis loop (DHL). DHLS are associated with exchange-bias (EB)-like shifts along and opposite to the field cooling axis below the ordering temperature of RE. The energy it takes to form a DW within the soft layer, which remains frozen upon cooling, results in the EB shift. Subsequently, exchange coupling and helical phase characteristics were also reported within both layers of Fe and Dy, comprising the multilayer\textsuperscript{8}. At low fields, the TM/RE layers exhibit ferromagnetic spin alignment, which form twisted magnetic helices in the form of planar $2\pi$-DWs. However, at higher fields, a more complicated but stable and continuous helical arrangement was observed. In another case, evidence of such stable helices within single layer Er films was shown using PNR\textsuperscript{9}.

Even though TM/RE multilayers are commonly studied, RE/RE multilayers remain scantily probed. Some combinations of epitaxial RE/Y multilayers showing modified helical moment configurations had been reported in the past using neutron diffraction\textsuperscript{9}. Epitaxial erbium thin films and Er-Y superlattices were reported to show the effects of basal-plane strain on the modulated spin structure by Borchers et al.\textsuperscript{10}. However, all strain related phenomena are expected in epitaxial films and are insignificant in polycrystalline ones. Moreover, Y is a transition metal but is often classified as RE due to its similar electronic behavior. Note that exchange coupling has also been observed in several TM/RE systems\textsuperscript{11,12,13}, but not in RE/RE systems. In contrast to the direct exchange in the transition elements, RE magnetism finds its explanation in an indirect exchange interaction known as the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Experimentally, it is found that in addition to normal ferromagnetism, some of the rare earths exhibit a helical AF state over a limited temperature range. On application of an external magnetic field, the helical state is radically distorted and eventually collapses into a ferromagnetic configuration. Presence of large orbital momentum in Tb, Dy, and Ho leads to strong spin-orbit coupling and larger magnetic anisotropy\textsuperscript{14}. Moreover, a large difference in spin-orbit coupling for different RE elements, would consequently have a significant influence on the overall demagnetization processes. In a recent example of RE/RE system, Er-Tb superlattice has been shown to possess long range magnetic order despite their competing anisotropy directions. Probing the vertically correlated magnetic structures by off-spectular polarized neutron scattering the existence of magnetic vortex-like domains associated with magnetic helical ordering within the Er layers was evidenced\textsuperscript{5}.

Here, we investigate polycrystalline [Dy\textsubscript{10}/Tb\textsubscript{10}]\textsubscript{10}: [Dy(10.0 nm)/Tb(10.0 nm)]\textsubscript{N=10} multilayers on Si substrates where N is the number of bilayer repetitions. The temperature ranges where Tb and Dy show AF helical magnetization, are distinctly different in bulk. For Tb, this range hovers around 220 K, whereas for Dy it ranges approximately between 180–80 K, below which both are ferromagnetic\textsuperscript{15,16}. We found that the multilayer is magnetically very hard, with a saturation magnetization beyond 50 kOe (5000 mT). Most interestingly, we observed an EB coupling up to ~0.88 kOe (~88 mT) due to strong pinning at spin-frustrated interfaces. Direct current (DC) magnetization as a function of temperature and alternating current (AC) susceptibility measurements for various frequencies and temperature confirmed such superspin-glass (SSG) type of behavior for the multilayer. Additionally, thermo-remanent magnetization (TRM) confirms the dynamics in the SSG phase and test its scaling behavior. Using PNR and polarized off-specular neutron scattering, we determined the magnetic profile of the multilayer indicating spin configurations like helices and associated magnetic fluctuations within the stack. Double spin spirals within Dy and Tb leads to spin-frustrated SSG-like ordering at the interfaces. Stability of the $2\pi$-DWs at a higher field ensures their topological stability. Our study is expected to open up a path for a broader base of researches as it describes novel phases of long range helical ordering in RE/RE polycrystalline system across spin-frustrated interfaces with innumerable possible combinations to explore in the future.

Results

X-ray characterization. For details on the initial characterization such as X-ray reflectivity (XRR) and X-ray diffraction (XRD), see the discussion in Supplementary Note 1 and Supplementary Fig. 1.

Magnetization measurements. In order to characterize the magnetic properties of the multilayer, the magnetization $M(T)$ was measured as a function of temperature for [Dy\textsubscript{10}/Tb\textsubscript{10}]. We applied different magnetic field strengths $H_{\text{irr}} = 0.25, 0.5, 1.0, 2.5, 5.0, 10.0,$ and $50.0\text{ kOe (25, 50, 100, 250, 500, 1000, and 5000 mT)}$ during the measurements. All samples were either initially cooled down to 5 K in presence of a field $H_{\text{FC}} = 70\text{ kOe (7000 mT)}$ to obtain the field cooled (FC) curves or with no magnetic field to obtain the zero field cooled (ZFC) curves. The $M(T)$ curves are shown in Fig. 1a, b. The ZFC curves show a broad peak which can be referred to as blocking/freezing temperature ($T_{\text{F}}$) whereas the FC curves do not show a temperature independent platform-like behavior. A well-defined irreversibility temperature ($T_{\text{irr}}$), which is indicated by the temperature where FC and ZFC curves diverge, could be identified. The gradual convergence of ZFC and FC curves with increasing field, indicates the attainment of similar type of magnetic configuration near equilibrium as exemplified for 50 kOe (5000 mT). As both 5/5 ([Dy\textsubscript{5}/Tb\textsubscript{5}]) and 10/10 ([Dy\textsubscript{10}/Tb\textsubscript{10}]) multilayers show a similar behavior (see Supplementary Figs. 2 and 3), we restrict further analysis to the [Dy\textsubscript{10}/Tb\textsubscript{10}] sample only. Note that for neutron measurements, sub-loops were measured for a limited FC condition of 10.0 kOe (1000 mT) as shown in Supplementary Fig. 4.

The transition from ferromagnetism to superparamagnetism (SPM) or super spin glass (SSG) behavior is generally expected for discrete nano-clusters (collection of nanoparticles) where individual magnetic moments within clusters are thermally unstable. The SSG state is believed to result from frustration generated by dipole–dipole interactions among superspins (magnetic moments of nano-clusters) and from disorders in the system (e.g., the random distributions of clusters, positions, sizes and anisotropy-axis orientations)\textsuperscript{15}. The irreversibility temperature ($T_{\text{irr}}$) and the...
Fig. 1 Field cooled (FC) and zero field cooled (ZFC) measurements. a, b The plot of in-plane magnetization versus temperature for the [Dy10/Tb10]10 sample. The measurements were done on heating at various fields (indicated in the figure) after ZFC and FC in 70 kOe (7000 mT). A broad maximum can be observed for the ZFC curves ($T_d$) followed by a furcation point ($T_m$) with the FC curves. Presence of a maximum for the ZFC curves at fields of 10 kOe (1000 mT) and lower indicates a freezing-like state. c The plot of $T_m$ and $T_d$ as estimated from the ZFC and FC curves versus increasing fields of measurement $H$. The lines are guide to the eye. Error bars represent standard error of the measured temperature value. d The plot of $H_{irr}$ versus $T_m$, showing $T_m(O) = 205$ K. The blue line is the linear fit to the data following the Almeida-Thouless (AT) equation. The inset of (d) shows phase diagram of the same in a plot of $H_{irr}$ as a function of $1 - T_m(H)/T_m(O)$. The region below the line is SSG while it is SPM above.

The blocking or freezing temperature $T_b$ as a function of field is shown in Fig. 1c for the [Dy10/Tb10]10 multilayer. $T_b$ is slightly below $T_{irr}$, which in some respects resembles cluster-glassy behavior. They both show a gradual decrease which suggests that the frozen state is suppressed by the field (a similar behavior is seen for [Dy5/Tb5]10); see Supplementary Fig. 2.

The shift of $T_{irr}$ to lower temperatures can follow the Almeida-Thouless (AT) line indicating a SSG-like behavior. The AT line is expressed as

$$H_{irr}/\Delta J \propto \left(1 - \frac{T_{irr}(H)}{T_{irr}(0)}\right)^{2/3} \tag{1}$$

where $T_{irr}(0)$ is the zero field freezing temperature and $\Delta J$ is the width of the distribution of exchange interactions. The plot of the measuring field $H_{irr}^{2/3}$ as a function of $T_{irr}$ has been shown in Fig. 1d, which fits to a straight line following the AT equation. The line cuts the $x$ axis at around $T_{irr}(0) = 205$ K for [Dy10/Tb10]10 ($T_{irr}(0) = 210$ K for [Dy5/Tb5]10); see Supplementary Fig. 2) which is similar to $T_b$. AT line separates a nonergodic (SSG) phase from an ergodic (SPM) one. In the inset of Fig. 1d we additionally plot $H_{irr}^{2/3}$ as a function of $1 - T_{irr}(H)/T_{irr}(0)$, showing the transition line separating the SPM and SSG phases.

In-plane magnetic field hysteresis loops were measured at seven different temperatures after field cooling in 70 kOe (7000 mT) from 300 K and are shown in Fig. 2a for [Dy10/Tb10]10. Apparently, there are no distinct DHLs as observed earlier for TM/RE multilayers. The hysteresis loops show high saturation fields of 50 kOe (5000 mT), large increments in their coercive fields ($H_c$) up to 1.1 kOe (110 mT) and an increase in exchange bias fields ($H_{EB}$) up to -0.65 kOe (-65 mT) for [Dy10/Tb10]10 ($H_{EB} = -0.88$ kOe or -88 mT for [Dy5/Tb5]10), particularly at temperatures below 75 K.

The temperature dependence of $H_{irr} = (H^{+}_c - H^{-}_c)/2$ (blue triangles) and $H_{EB} = (H^{+}_c + H^{-}_c)/2$ (black squares), where $H^{+}_c$ and $H^{-}_c$ are the coercive fields for the positive and negative field axes, are shown in Fig. 2b for the [Dy10/Tb10]10 multilayer. One can see a gradual decrease of the coercivity $H_{irr}$ with increasing temperature. The $H_{EB}$ value decreases gradually with increase in temperature and vanishes at around 200 K. Note that the temperature dependence of the remanent magnetizations $M = (|M| + |M_{-}|)/2$ (red circles in Fig. 2b) goes to zero also at 200 K (a similar behavior was seen for the [Dy5/Tb5]10 multilayer as well).

Dy exhibits a rich magnetic phase diagram, including a few modulated magnetic phases and also posses one of the highest intrinsic magnetic moments (bulk value of 10.6 $\mu_B$/atom). The magnetic modulations propagate coherently over a long range, even with intervening nonmagnetic layers, via RKKY interaction. The magnetic moment of Tb (with bulk value of 9.0 $\mu_B$/atom and 6.4 $\mu_B$/atom at 10 K in epitaxial thin film) is slightly lower than Dy. The range of stability of the helical state in Tb is quite restricted compared to Dy. An applied field of less than 1 kOe (100 mT) will produce a transition to the ferromagnetic state in Tb, while in Dy the reminiscent of the helical state can be found even at lower temperatures.

Moreover, due to basal–plane anisotropies existing both in Dy (a axis) and Tb (b axis), a combination of these two RE elements may lead to an exchange coupling as well. With an increase in spin-frustration in the pinning layers at the interfaces, a transition from a helical AF domain state to a spin-glass type state might be expected which can eventually increase the $H_{EB}$ field.

If we assume that randomly oriented nano-clusters are following the usually observed $T^2$ law of nucleation and propagation for an ensemble of non-interacting clusters, it
temperatures. The loops are corrected for their diamagnetic contributions. (empirical equation 7,30.

The shift gives the activation energy and is characteristic of the supermagnetic dynamics. The models are employed in the (i) Néel-Arrhenius law 25,26, (ii) Vogel-Fulcher law27,28. Phenomenologically, the dynamical behavior, three common models are employed in discerning the supermagnetic dynamics. The models are embodied in the (i) Néel-Arrhenius law25,26, (ii) Vogel-Fulcher law27,28 and (iii) power law29 in addition to the (iv) empirical equation7,30.

Noninteracting SPM clusters generally obey the Néel-Arrhenius law25,26.

\[ \tau_m = \tau_0 \exp \left( \frac{\Delta E}{k_B T_{AC}^{\text{max}}} \right) \]  

(2)

where \( \tau_0 = (2\pi f_0)^{-1} \) is the relaxation time for the attempt frequency \( f_0 \) and \( \tau_m = (2\pi f_m)^{-1} \) is the measuring time (~10^2 s for the DC magnetization measurement) for the measuring frequency \( f_m \). Here, \( \Delta E = (-K_A V) \) is the activation energy, \( K_A \) is the anisotropy constant, \( k_B = 1.38 \times 10^{-16} \text{erg/K} \) is the Boltzmann constant and \( V \) is the average cluster volume critical for SPM or SSG state. The value of \( \tau_0 \) typically ranges from ~10^{-9} to ~10^{-13} s for a SPM behavior31. In Fig. 3b, the dependence of \( (T_{AC}^{\text{max}})^{-1} \) on the natural logarithm of the measurement frequency is plotted. A linear fit to the data yields a slope of \( \Delta E/k_B = 190.344 \text{K} \), which implies an unreasonably small value of \( \tau_0 \). Failure of the Néel-Arrhenius expression in describing the dynamics suggests the presence of non-ignorable magnetic dipole–dipole interactions.

Weak interactions among spin nano-clusters are taken into account within the Vogel-Fulcher law27,28. Phenomenologically, it describes the frequency response of the relaxation time and is expressed by

\[ \tau_m = \tau_0 \exp \left( \frac{\Delta E}{k_B T_{AC}^{\text{max}} - T_0} \right) \]  

(3)

where \( T_0 \) is the characteristic temperature that accounts for the static interaction field of the surrounding clusters. In Fig. 3c, a plot of \( \ln(\tau_m) \) versus \( T_{AC}^{\text{max}} \) is shown. The values obtained from a fit to the equation 3 are \( \Delta E/k_B = 112 \text{K} \), \( T_0 = 203 \text{K} \) and \( \tau_0 = 10^{-13} \text{s} \). From the linear fit to the plots of \( T_{AC}^{\text{max}} \) versus \( 1/\ln(\tau_m/\tau_0) \), there can be a few possible combinations where \( \tau_0 \) can be within a reasonable range yielding different \( \Delta E/k_B \) values32. For example, \( \tau_0 = 10^{-17} \text{s} \) corresponds to \( \Delta E/k_B = 214 \pm 12 \text{K} \) and \( T_0 = 201 \pm 0.4 \text{K} \) while \( \tau_0 = 10^{-9} \text{s} \) corresponds to \( \Delta E/k_B = 35 \pm 2 \text{K} \) and \( T_0 = 205 \pm 0.2 \text{K} \) (see the discussion under AC susceptibilities within Supplementary Note 2 and subsequent Supplementary Fig. 5a).

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These values are reasonably comparable with the conventional spin-glass behavior for which the typical value of $\tau_0 \sim 10^{-10}$ to $10^{-13}$ s or even lower. A longer $\tau_0$ yields dynamics on a shorter accessible time scale and are probed within the experimental time window available for a SSG type of behavior. The maxima observed in $\chi'(T)$ would therefore correspond to the freezing of weakly interacting moments of the nano-clusters.

The power law relies on the relation of critical slowing down of the relaxation time near the transition temperature. It is also known as the scaling hypothesis, which predicts the existence of an equilibrium phase transition. The relaxation behavior corresponding to a characteristic temperature $T_{\text{SSG}}$ for superspin-glass behavior is expressed by

$$\tau_m = \tau_s \left( \frac{T_{\text{AC}} - T_{\text{SSG}}}{T_{\text{SSG}}} \right)^{-z\nu}$$

where $\tau_s$ is a relaxation time for each nano-cluster, $z\nu$ is the dynamical scaling critical exponent constant related to the correlation length $\xi$, which is a measure of the size of the lateral coarsening ($\nu$ describes the divergence while $z$ is involved in the dynamical scaling hypothesis $\tau_m \sim \xi^z$). Tholen criterion requires a very small value for $\eta = (T_{\text{AC}} - T_{\text{SSG}})/T_{\text{SSG}}$ in order to comply with the SSG type of behavior. In Fig. 3d, we plot the variation of $\tau_m$ (in log scale) versus $T_{\text{AC}}$. A fit to the Eq. (4) yields $z\nu \sim 5.0$ (typical range for SSG is between 4 to 12). $\tau_s \sim 3 \times 10^{-15}$ s, and $T_{\text{SSG}} \sim 206.8$ K. Considering this $T_{\text{SSG}}$ value, the maximum value $T_{\text{max}}$ is the difference in $\chi'(T)$, whereas $\Delta T_{\text{AC}}$ is the difference in $T_{\text{AC}}$ over the frequency interval $\Delta \log_{10}(f_{\text{AC}})$. Typically, for SSG phase, the parameter $\rho$ assumes values of 0.0045–0.06 whereas for SPM phase, it is between 0.1–0.30. The parameter $\rho$ for the SPM phase is typically an order or two higher than that for the SSG phase. Here, we obtain $\rho = 0.0028$, which again points towards an interacting SSG-type of behavior. The obtained lower value of $\rho$ indicates that the accessible time scale for SSG dynamics is indeed shorter.

For TRM measurements, the sample is heated to a temperature above the superspin-glass transition temperature ($T_{\text{SSG}} = 205$ K) and subsequently cooled to a measuring temperature ($T_{\text{m}} = 143$ K) in the presence of a small excitation field, $H_{\text{TRM}} = 0.005$ kOe (0.5 mT). After a waiting time $t_w$, the field is switched off and the relaxation of the magnetization is measured over $t = 30,000$ s. A waiting time dependence was examined to evaluate the aging character of the dynamics. The field should remain small enough so that the response of the sample remains in the linear regime (non-perturbative probe) to avoid an influence of the field on aging.

Figure 4a shows the relaxation of magnetization $M_{\text{TRM}}(t)$ for [Dy10/Tb10]$_{10}$ normalized to the field cooled magnetization ($M_{\text{FC}}$) for values of waiting times $t_w$ varying from 30 s, 300 s, 1,000 s, 3,000 s to 30,000 s. The plot for $t_w = 0$ (at $t_0$) is used for reference. It can be seen that the relaxation depends on the value of $t_w$: this illustrates the aging character of the dynamics. Longer the $t_w$, slower is the relaxation, indicating a stiffening of the sample response during the waiting time. The relaxation trend $S(t) = \Delta M_{\text{TRM}}(t)/\log(t)$ can be used to show the changes in $M_{\text{TRM}}(t)$ more clearly, where $\Delta M_{\text{TRM}} = M_{\text{TRM}}(t_w) - M_{\text{TRM}}(t_0)$. Note that $S$...
is slightly different from the commonly defined derivative term. Nevertheless, the quantity $S$ is equivalent to the relaxation time distribution, a wide breadth of which is the reason behind the slow and non-exponential relaxation of the response function in a SSG state. The inflection point $t_{\text{inf}}$ can be found from the $S(t)$ versus $\log(t)$ plot in Fig. 4b.

Two features distinguish a SSG behavior from an atomic spin glass behavior. Firstly, a departure from approximate scaling in the TRM curves versus the scaling variable $\lambda/t_{\text{w}}$ plot in Fig. 4c where the effective time $\lambda = t_{\text{w}}[(1 + t_{\text{w}})^{-\mu} - 1]/[1 - \mu]$ with $\mu = 0.5$, which is much smaller than is normally found for atomic spin glasses (0.7–0.9) and is similar to that of SSG (0.4)$^{38}$. Values of $\mu \neq 1$ indicate by how much the effective age of a SSG deviates from the nominal age; i.e., the experimental waiting time, $t_{\text{w}}$.$^{45}$ Secondly, $\log(t_{\text{inf}}) \neq \log(t_{\text{w}})$, which is unlikely for an atomic spin glass where an approximate scaling is usually observed (Fig. 4d). Thus, the SSG behavior is well established for the multilayer under investigation.

**Polarized neutron scattering measurements.** For the investigation of the magnetic structure of the sample, PNR measurements were carried out. While specular scattering is used for depth information, off-specular scattering can deliver information on lateral structure. A sketch of the neutron scattering geometry is shown in Fig. 5a. Figure 5b shows the sketch of lateral magnetic domains with respect to the lateral projection of the neutron coherence length $l$, which is deterministic in observing specular and off-specular scattering. For details see the “Methods” section.

PNR measurements were performed for different magnetic histories of [Dy10/Tb10]$_{10}$. Figure shows the specular polarized neutron intensity profiles along $Q_z$ (see the “Methods” section for details) and their fits. The measurements were done after cooling the sample to 10 K in zero field (ZFC) (Fig. 6a) and in $+10.0$ kOe or $+1000$ mT (FC) (Fig. 6b). Both measurements took place at an external field $H_e = -0.01$ kOe ($-1$ mT). The corresponding hysteresis sub-loops measured at 10 K are shown for the FC condition in Supplementary Fig. 4. The fits were initialized using simple models of block-potentials. The parameters used for fitting were the individual layer thicknesses along with the nuclear and magnetic scattering length densities (SLDs) of individual layers. The errors in the thickness of the layers are ±0.2 nm, while that for the nuclear and magnetic SLDs $\rho_n$ and $\rho_m$ are $\pm1 \times 10^{-6}$ Å$^{-2}$ and $\pm0.05 \times 10^{-6}$ Å$^{-2}$, respectively. As the top layer is susceptible to oxidation, we assumed a different $\rho_n$ and $\rho_m$ values when compared with the stack. The interface roughness is 2.0(±0.5) nm. Fitted parameters were obtained using the minimization of the reduced chi-squared ($\chi^2$ or the goodness of fit) value.

Due to the limitation in providing the maximum applied field at the experimental station, the saturation moments could not be determined. The ZFC data (Fig. 6a) at $H_e = -0.01$ kOe ($-1$ mT) (virgin state) was found similar to the room temperature data with no net magnetic moment. Absence of any net magnetization is due to the formation of domain states in accordance with the ZFC data in Fig. 1a. The peaks at $Q_z = 0.038$ Å$^{-1}$ and $Q_z = 0.109$ Å$^{-1}$ are the 1st order and 3rd Bragg peaks due to the structural periodicity. The 2nd order structural Bragg peak is expected to be significantly reduced since the layer thicknesses are almost equal. For the structural SLDs, the fits yielded the following values: $\rho_n = 2.5 \times 10^{-6}$ Å$^{-2}$, $\rho_m = 4.8 \times 10^{-6}$ Å$^{-2}$. We also found an interdiffused layer of around 0.5 nm and 1.0 nm at the Dy/Tb and Tb/Dy interfaces, respectively. This diffusion layer was also evident in the XRR measurements (estimated from the fits to the XRR data in Supplementary Fig. 1). In the case of a random distribution of domain magnetization directions, the dispersion is $\langle \cos^2 \phi_A \rangle - \langle \cos \phi_A \rangle^2 \neq 0$. Here, $\phi_A$ is the angle that the magnetization $M_{FM}$ subtends with the applied field $H_e$. For a coherent
Specular scattering are observable here, for magnetic domains (green arrows) with respect to the coherence ellipse (shaded green area) or the lateral projection of the neutron coherence length \( l_c \). Here, for \( \alpha \neq \alpha_0 \), one may observe off-specular scattering where \( \alpha_i \) and \( \alpha_f \) are incident and exit angles for scattering. When the magnetic domains are smaller than the coherence ellipse length \( l_c \), both specular and off-specular scattering are observable. When the domains sizes are larger than \( l_c \), mainly specular scattering are observable.

The nuclear SLDs, thus obtained, were used to fit the FC data (Fig. 6b) at \( H_a = -0.01 \) kOe (\(-1 \) mT) and obtain the magnetic SLD parameters close to the reduced-remanence field of the sub-loop hysteresis. Here again, the fit was started with a simple model of block-like-potentials with average magnetic SLD values to fit the profiles. At this field, the magnetic SLD values obtained from the fits are: \( \rho_{m,Tb} = -1.4 \times 10^{-6} \) Å\(^{-2} \) and \( \rho_{m,Dy} = -1.6 \times 10^{-6} \) Å\(^{-2} \). The corresponding magnetic moments are Tb: 1.7 \( \mu_B/\)atom and Dy: 1.9 \( \mu_B/\)atom. These values are found to be much lower than those known for bulk or values reported earlier (polycrystalline Tb: 2.4 \( \mu_B/\)atom\(^6\) and polycrystalline Dy: 7.1 \( \mu_B/\)atom\(^8\)) for thin films. This lowering of moment is caused by the fact that the FC process was done not in a saturation field but in an intermediate field. The top layer was found to posses a similar average moment and was associated with a small rotation, oriented 10(±5) away from the applied field direction. This disorientation gave rise to a small SF signal. Such an increase in the SF signal also was reported earlier for another multilayer system\(^{39}\). The difference between the two SF curves is basically due to a difference in the instrumental (polarizer and analyzer) inefficiencies. The plot in the inset show the \( \rho_n \) and \( \rho_m \) values for the two layers as function of thickness.

Similarly, fits to the FC profiles (Fig. 7) at \( H_a = -10.0 \) kOe (\(-1000 \) mT) were used to obtain the magnetic SLD parameters near the coercive field. Like that for the ZFC measurement at \( H_a = -0.01 \) kOe (\(-1 \) mT), no true SF signal could be observed at this field, which indicates existence of either domain states or compensated moments\(^{40}\). We have investigated several models representing different possible scenarios in an attempt to simulate the intensity profiles (see the discussion under Model description within Supplementary Note 3, Supplementary Table 1 along with Supplementary Figs. 6 and 7). Out of these, two of the most probable scenarios are discussed below.

Fig. 5 Neutron scattering geometry. a Illustration of the in-plane neutron scattering geometry. Here, \( M_{FM} \) is the in-plane magnetization vector making an angle \( \phi_A \) with respect to the field \( (H_a) \) along the \( y \) axis. The spatial fluctuation of \( M_{FM} \) from region to region is designated by \( \Delta \phi_A \). B Sketch of lateral magnetic domains (green arrows) with respect to the coherence ellipse (shaded green area) or the lateral projection of the neutron coherence length \( l_c \). Here, for \( \alpha \neq \alpha_0 \), one may observe off-specular scattering where \( \alpha_i \) and \( \alpha_f \) are incident and exit angles for scattering. When the magnetic domains are smaller than the coherence ellipse length \( l_c \), both specular and off-specular scattering are observable. When the domains sizes are larger than \( l_c \), mainly specular scattering are observable.

Fig. 6 Polarized neutron reflectivity (PNR) measurements. Specular neutron reflectivity patterns (solid symbols) along with their best fits (open symbols) as a function of \( Q_z \) for the non-spin flip (NSF) [\( R_{-z} \) (black) and \( R_{+z} \) (red)] and spin flip (SF) [\( R_{-x} \) (blue) and \( R_{+x} \) (green)] channels measured at a reduced-remanence field \( H_a = -0.01 \) kOe (\(-1 \) mT) after a zero field cooled (ZFC) and b field cooled (FC) and measured at 10 K for the [Dy10/Tb10]\( n \) sample. Error bars represent standard error of the estimated intensity. Shown alongside are the nuclear \( (\rho_n) \) and magnetic \( (\rho_m) \) scattering length densities (SLDs) versus thickness of the multilayer.

NSF specular signal. The plot in the inset show the depth profiles in terms of \( \rho_n \) values for the two layers while the \( \rho_m \) value is zero for both.

Rotation this dispersion is essentially zero. One should not expect here any SF specular signal due to the formation of domain states and no net magnetism, therefore the SF specular intensity visible is attributed purely to the inefficiencies (2–5\%) of the optical elements. Note that the so-called leaked SF specular signal is at least two orders of magnitude smaller than the corresponding parameters close to the reduced-remanence field of the sub-loop hysteresis. Here again, the fit was started with a simple model of block-like-potentials with average magnetic SLD values to fit the profiles. At this field, the magnetic SLD values obtained from the fits are: \( \rho_{m,Tb} = -1.4 \times 10^{-6} \) Å\(^{-2} \) and \( \rho_{m,Dy} = -1.6 \times 10^{-6} \) Å\(^{-2} \). The corresponding magnetic moments are Tb: 1.7 \( \mu_B/\)atom and Dy: 1.9 \( \mu_B/\)atom. These values are found to be much lower than those known for bulk or values reported earlier (polycrystalline Tb: 2.4 \( \mu_B/\)atom\(^6\) and polycrystalline Dy: 7.1 \( \mu_B/\)atom\(^8\)) for thin films. This lowering of moment is caused by the fact that the FC process was done not in a saturation field but in an intermediate field. The top layer was found to posses a similar average moment and was associated with a small rotation, oriented 10(±5) away from the applied field direction. This disorientation gave rise to a small SF signal. Such an increase in the SF signal also was reported earlier for another multilayer system\(^{39}\). The difference between the two SF curves is basically due to a difference in the instrumental (polarizer and analyzer) inefficiencies. The plot in the inset show the \( \rho_n \) and \( \rho_m \) values for the two layers as function of thickness.

Similarly, fits to the FC profiles (Fig. 7) at \( H_a = -10.0 \) kOe (\(-1000 \) mT) were used to obtain the magnetic SLD parameters near the coercive field. Like that for the ZFC measurement at \( H_a = -0.01 \) kOe (\(-1 \) mT), no true SF signal could be observed at this field, which indicates existence of either domain states or compensated moments\(^{40}\). We have investigated several models representing different possible scenarios in an attempt to simulate the intensity profiles (see the discussion under Model description within Supplementary Note 3, Supplementary Table 1 along with Supplementary Figs. 6 and 7). Out of these, two of the most probable scenarios are discussed below.

In Fig. 7a uniform average magnetizations within both layers were considered. This simple model of block-like-potentials with average magnetic SLD values failed to fit the profiles. The magnetic SLD values which could be obtained were: \( \rho_{m,Tb} = 0.25 \times 10^{-6} \) Å\(^{-2} \) and \( \rho_{m,Dy} = 0.7 \times 10^{-6} \) Å\(^{-2} \). The magnetic SLDs and thereby the corresponding magnetic moments of Tb: 0.3 \( \mu_B/\)atom and Dy: 0.8 \( \mu_B/\)atom are much lower than in bulk or thin film at saturation. The plot in the inset show the \( \rho_n \) and \( \rho_m \) values versus thickness obtained for the two layers. This drastic lowering
of layer magnetizations indicate that they allow $2\pi$-DWs formation within the layers which are stabilized by the in-plane magnetocRYstalline anisotropy and exchange coupling at the interfaces.

Furthermore, the magnetization vector of each layer to rotate in-plane ($x-y$) by an angle $\phi_{X,Y}$ within the multilayer stack is considered, which mimics the existence of $2\pi$-planar DWs within both layers of Dy and Tb. The scenario is shown in Fig. 7b. The plot in the inset shows the $\rho_{\parallel}$ and $\rho_{\perp}$ values obtained for the two layers as a function of the depth. The larger Dy and Tb moments are confined within a range of a few nanometers from the core of the layers. The planar DWs, which are compressed with an increase in field, are spanned on both sides of the remaining layer while they penetrate on either side of the adjacent layer. Extension of continuous helices, from one layer to the other, is owed to ferromagnetic coupling of the core layers and formation of DWs due to the pinning at interfaces. Within each layer, integration of magnetic moments show little or no variation around the core, whereas around the interfaces the DWs being laid spanned results in a lowering of magnetization. The improved quality of the fit with planar DWs indicates a possibility of our helical modulation model. Since no additional parameters were included other than those mentioned we believe that this is the most logical consequence. Earlier, helical magnetic structures or $2\pi$-DWs have been reported for similar systems.

Unlike TM/RE systems, here the layers are neither ferrimagnetically or antiferromagnetically aligned but are ferromagnetically aligned to each other. Signatures of magnetic helices in Dy have been reported earlier in presence of low fields (0.03–0.1 kOe or 3–10 mT) even at 5 K. The magnetic ordering of helical modulation is sensitive to the cooling history and interface clamping. Thus, one can expect a similar helical phase within Dy and Tb, which are merely reminiscent of the helical ordering realized at higher temperatures. The magnetic stability of the confined helices, demonstrated at $-10.0$ kOe ($-1000$ mT), can be attributed not only to the internal field that is created by exchange interaction which stores the magnetic energy but also to the strong anisotropy of the Dy layer (axial: $10^9$–$10^8$ erg/cm$^3$; basal-plane: $10^7$–$10^6$ erg/cm$^3$). Note the difference in the basal-plane anisotropy constant for Tb ($2.4 \times 10^6$ erg/cm$^3$) and Dy ($7.5 \times 10^6$ erg/cm$^3$) at 4 K. This difference in anisotropy leads to planar DW formation whose energy is modified by exchange interaction. The anisotropy in both Tb and Dy keeps the magnetic moments confined to the basal plane.

We show the specular and off-specular NSF and SF intensity maps in Fig. 8a, b for the multilayer with two different field histories, ZFC and FC and measured at $H_z = -0.01$ kOe (1 mT) and $=-10.0$ kOe ($-1000$ mT), respectively. The scenario for FC condition and measured at $-0.01$ kOe (1 mT) also showed a similar intensity map (not shown). The measurement channels shown are the NSF channels $I_{++}$, $I_{+-}$ and the SF channel $I_{--}$. The intensity along the diagonal $\alpha_1 = \alpha_2$ is the specular reflection along the scattering vector $Q_{\parallel}$ and has been discussed in the last section. Apart from the multilayer Bragg peaks along the specular ridge depicting the periodicities of the multilayer, we could observe off-specular intensities in form of sheets of intensities perpendicular to the specular line (Bragg sheets) emanating across the Bragg peaks ($I_{++}$ and $I_{--}$ channels) and intensities near the critical edge and along the Yoneda wings. Additional intensities along the Bragg peaks, parallel to $\alpha_{sf}$ (diffuse streaks in the $I_{--}$ channel) indicate configuration of a domain state for sample magnetization perpendicular to the $H_z$ direction.

Similar intensities of the specular Bragg peaks in the NSF channels indicate that they were measured either at $-0.01$ kOe (1 mT) after ZFC (Fig. 8a) or at the coercive field of $-10.0$ kOe ($-1000$ mT) after FC (Fig. 8b). A slight imbalance in intensity along the specular ridge at $-10.0$ kOe ($-1000$ mT) between $I_{++}$ and $I_{--}$ channels indicate a preferential alignment of the magnetization, which is also in accordance with the fits to the specular profiles shown in Fig. 7b.

In the SF channel, there exists no off-specular Bragg sheet. However, off-specular diffuse streaks exist in the NSF channels as well as in the SF channels, indicating a domain state configuration of the sample magnetization also along and perpendicular to the $H_z$ direction (parallel and perpendicular components). The Bragg sheets in the NSF channels remain similarly intense with a change in the $H_z$ value while the diffuse streaks (indicated by arrows) are more intense in the $I_{++}$ channel for ZFC and in the $I_{--}$ channel for FC. Similarly, in the SF channel $I_{--}$, it is more intense along $\alpha_2$ at $-0.01$ kOe (1 mT) as compared to that along $\alpha_1$ at $-10.0$ kOe ($-1000$ mT), which obviously indicates that the diffuse streaks are of magnetic origin while the Bragg sheets, which remain similar at both fields, are not of magnetic origin. Such a temperature or field dependence of off-specular SF scattering from vertically correlated domains was reported earlier by Paul et al. in epitaxial Er/Tb system. Owing to the reciprocity theorem, SF channels intensity maps, $I_{+-}$ and $I_{--}$ are interchangeable by a magnetic field directional switch.

**Fig. 7** Polarized neutron reflectivity (PNR) measurements. Specular neutron reflectivity patterns (solid symbols) along with their best fits (open symbols) as a function of $Q_z$ for the non-spin flip (NSF) $R_{+(+)}$ (black) and $R_{+-}$ (red) and spin flip (SF) $R_{+-}$ (blue) and $R_{-+}$ (green) channels measured at a coercive field $H_z = -10.0$ kOe ($-1000$ mT) after field cooled (FC) and measured at 10 K for the [Dy10/Tb10]10 sample. Error bars represent standard error of the estimated intensity. The simulations are done using models considering cases a without any 2\( \pi \)-domain wall and b with 2\( \pi \)-domain walls in both layers of Dy and Tb. Shown alongside are the nuclear ($\rho_{\parallel}$) and magnetic ($\rho_{\parallel}$) scattering length densities (SLDs) versus thickness of the multilayer.
certain field without any change in the field direction, they are supposed to remain asymmetric with respect to the diagonal intensities, i.e., $I_{+ +} \neq I_{+ -}$. Thus, the change in asymmetry of the scattered intensity in the respective SF channel $I_{+ +}$ for ZFC and FC in Fig. 8a, b, accounts for the fact that the spin states of the incoming and outgoing neutron waves are inversely populated indicating that they have been reversed by the field. This interchange of intensity obviously conveys magnetic origin.

On the one hand, the Bragg sheets occur due to pronounced structural and/or magnetic vertical correlation along the stack of the multilayer. On the other hand, diffuse streaks run parallel to $\alpha_d$ axes. Due to some instability in the system, one often witnesses fluctuations of the magnetization $(\Delta \phi_A)$ from vertically uncorrelated domains around the mean magnetization in the form of diffuse scattering. Perpendicular fluctuations are characterized by the mean value $\sin^2(\Delta \phi_A)$, which are commonly seen in the SF channels below a saturation field.\(^{40,43}\)

Distorted wave Born approximation (DWBA) has been applied in simulating the corresponding SF intensity maps in the SF $I_{+ +}$ channel (Fig. 8a, b). The parameters for the off-specular data simulations follow from the fit parameters obtained from the fitting of the corresponding specular data. Thus the inefficiencies of the polarizer and analyzer are intrinsically incorporated in the simulations. One can see an increase in the off-specular SF intensities near the critical edge in the simulated maps. These are typical signatures of random non-collinear arrangement of small-scale domains with sizes ranging from $1.5 \pm 0.5$ \(\mu\)m and $0.5 \pm 0.5$ \(\mu\)m accompanied by fluctuations of magnetization $\Delta \phi_A = 4$ and 15, for ZFC and FC conditions measured at $-0.01$ kOe ($-1$ mT) and $-10.0$ kOe ($-1000$ mT), respectively. The domain sizes are typically smaller than the neutron coherence length along the $x$ axis (few microns), i.e., as and when the coherence ellipse covers several domains.\(^{40,43}\) The change in asymmetry for ZFC and FC has duly been accounted for in the respective simulations.

**Noncollinear configuration of planar spin chain.** Theoretically, Popov et al.\(^{44}\) treated in-plane exchange interaction within mean field approximation, the energy of which can be described by a finite size 1D model for $N$ planar classical spins

$$
\varepsilon = -\sum_{i=1}^{N} \cos(\phi_{A,i}^{i+1} - \phi_{A,i}^{i}) - \sum_{i=1}^{N} (h_A \cos^2 \phi_{A,i}^{i} + 2h \cos \phi_{A,i}^{i})
$$

Here, $h_A = H_A/H_{EB}$ and $h = H_{EB}/H_A$, where $H_{EB}$, $H_A$, and $H_2$ are the effective exchange field, effective uniaxial anisotropy field and external applied field, respectively. A spin on the side of the chain forms an angle $\phi_A^{i}$ perpendicular to the chain axis. A number of collinear ground state and noncollinear equilibrium stable configurations were numerically calculated. Even though the theoretical discussion is meant for epitaxial films,\(^{44}\) we could find it relevant for polycrystalline films as well emphasizing the robustness of spin configurations and thereby the generality of results.

A multilayer stack can relax into different equilibrium states depending upon the energy density introduced by an external magnetic field. With removal of the external field, the multilayer typically relaxes into a ground state as all magnetic moments remain aligned and attains a global minimum. However, for an intermediate field, the stack relaxes into a metastable state as the spins remain misoriented with the attainment of a number of local energy minima.\(^{5,8}\) In the present context, Fig. 6b represents a scrambled ground state at $-0.01$ kOe ($-1$ mT) where all spins are randomly oriented while Fig. 7b represents a state of noncollinear equilibrium configurations at $-10.0$ kOe ($-1000$ mT) where spins are appreciably misoriented with respect to the hard axis as expected for an intermediate field.

Supplementary Fig. 8a shows a schematic of the noncollinear spin configurations containing two $\bar{\pi}$-DWs (topologically protected non-trivial structure) so that the overall rotation of the spins along the chain is $2\pi$ within the layers $\left(\frac{\pi}{2} > 2\pi\right)$ of Dy and
Tb. Alternatively, a combination of $\pi$-DW and $\pi$-antiDW (topologically unstable trivial structure) of width $\xi$ is possible. It may be noted that PNR alone cannot distinguish between the two configurational states. However, magnetic stability of the 2$\pi$-DWs or confined helices at $-10.0$ kOe ($-1000$ mT) exemplifies the fact that a non-trivial topological structure cannot be easily destroyed by a field.

In order to reduce the cost of uniaxial anisotropy the spin orientations remain close to an integer multiple of $\pi$ at the end points of the chain. Due to halved number of nearest neighbors, the spins at the surface/interface (centers of DW nucleation) were shown to be two times less, which reduces the energy ($E_n = 4a/(J_{ab} - K)$) near the end points by a factor of $\sqrt{2}$. The DW width, which is given by $\xi = \frac{\sqrt{2}}{a} \approx \pi \sqrt{2}$, leads to an uneven distribution of spins within an individual layer with relative compression at the interfaces and decompression in the inner layers. Here, $y = \frac{\xi}{a}$ as $J_{ex}$, $a$ and $K$ are the exchange coupling constant, lattice constant and uniaxial anisotropy constant, respectively. Also, $J_{ex} = \frac{4a}{\xi}$ where $A$ is the exchange stiffness. Assuming the parameter $A = 8 \times 10^{-8}$ erg/cm for Dy and Tb and $K_{Dy} = 7.5 \times 10^6$ erg/cm$^3$ and $K_{Tb} = 2.4 \times 10^6$ erg/cm$^3$, the basal plane DW widths are $\xi_{Dy} \sim 3.2$ nm and $\xi_{Tb} \sim 5.7$ nm. Thus, a difference in the reduced number of spins at the end points due to unequal $\xi$ along with structural interdiffusion ($\sim 2.0 \pm 0.5$ nm) at the interfaces of Dy and Tb layers would contribute to vertically uncorrelated domains when the two layers are stacked as a multilayer.

Based upon the temperature-dependent neutron-scattering intensity, a picture of the spin-glass state coexisting with the helical spin ordering was discussed earlier by Sato et al.45. An interdomain interaction should be pronounced when the competition of two types of modulated spin fluctuation becomes significant, particularly at the interfaces. A frustrated interdomain magnetic interaction would lead to the coexistence of spin-glass-like ordering and helical spin modulation.

Model interpretation of spin-glass as a short-range spin density wave (SDW) has been proposed earlier by Myoshin17. Therefore, magnetic configurations of topologically stable modulated helices can be visualized as a superposition of the double spin spirals or SDW within Dy and Tb with similar propagation vector $q$ ($\sim 2\pi/nN$) and pitch, and separated by a phase factor. A resultant of superposition of two helices represented by the magnetic SLD profile is shown in Supplementary Fig. 8b, for clarity. The number of nodes/anti-nodes (modulation period) in the modulation of the envelope lines forming the double helices, which being commensurate with the chain’s length, determines the number of twists in the chain. Increase in the number of twists increases the helix modulation, which in turn enhances the energy stored in the system.

Discussion
We have demonstrated the realization of magnetic helices in form of 2$\pi$-planar DWs in an unprecedented multilayered system involving two hard rare-earth materials (Dy/Tb), instead of a more familiar soft-hard (Fe/Tb or Fe/Dy) combination. Using depth sensitive PNR, DWs have been shown to exist within both Dy and Tb. Since both materials are anisotropically hard and their respective helical and ferromagnetic ordering temperatures are also significantly different in bulk form, it was hardly expected for them to establish exchange coupled helices. Thus, our work realizes a novel state of spin configuration even within such unusual candidates.

The robustness of the helices at an applied field of $-10.0$ kOe ($-1000$ mT) ensures their topological stability. The helices within both RE/RE layers are stabilized by the strong in-plane magnetocrystalline anisotropy of the Dy and relatively softer Tb layers induced by the growth process. Exchange bias coupling of around $-0.88$ kOe ($-88$ mT) and $-0.65$ kOe ($-65$ mT) were revealed in two multilayers with different $t_{Dy}$ and $t_{Tb}$ thicknesses combinations, which can be owed to the enhanced pinning at spin-frustrated interfaces.

Rigorous magnetic data analysis suggests the existence of superspin-glass-type of freezing of weakly interacting disordered spins. Additionally, such SSG type of behavior with $T_{SG} \approx 206$ K, was found to be coexistent with the helical spin configurations. The dynamics in the superspin glass phase and its scaling behavior was further confirmed by TRM measurements. Such a coexistence of SSG and helical phases, is due to the spin-frustrated interdomain magnetic interaction as indicated by polarized off-speccular neutron scattering.

We believe, that topological stability for the effective periodicity of the modulation and the internal effective field in the helix, which stores the magnetic energy density, can be interesting for exploiting their applicability in all-spin-based technology.

Methods
Sample preparation. The samples were prepared by DC magnetron sputtering using Si(100) as substrate, of compositions $[\text{Dy}_{10}/\text{Tb}_{10}]_{10}$, $[\text{Dy}(10.0 \text{nm})/\text{Tb}(10.0 \text{nm})]_N \times 10 = 10$ and $[\text{Dy}/\text{Tb}]_{10}$. $[\text{Dy}(5.0 \text{nm})/\text{Tb}(5.0 \text{nm})]_N \times 10 = 10$. Two different multilayers, with different thicknesses of $t_{Dy}$ and $t_{Tb}$, were prepared in order to investigate the effect of changing Dy and Tb magnetization. Thicknesses of the layers were carefully chosen such that the volume anisotropy is confined to the film plane overcoming the surface anisotropy.

The substrates were single crystalline Si wafers of $20 \times 20$ mm$^2$, which were used after cleaning them in isopropyl alcohol. The targets were disks of 2 inch diameter and 0.125 inch thickness with a purity of 99.9%. They were bonded to a copper backing plate. The targets were cleaned by pre-sputtering for 10–15 min in Ar atmosphere. The deposition rates were about 0.215 nm/s for Tb and 0.282 nm/s for Dy and were precalibrated. During deposition, the Ar pressures in the magnetron sputtering chamber were $3.9 \times 10^{-5}$ mbar for Tb and $2.9 \times 10^{-5}$ mbar for Dy and the base pressure was $1.5 \times 10^{-8}$ mbar. The magnetron sputtered samples were expected to grow with a high degree of crystallographic orientation (texture) and in-plane easy axis as confirmed earlier from the XRD patterns (see Supplementary Fig. 1).

X-ray characterization. XRR and XRD measurements were performed on an Empyrean diffractometer from PANalytical which provides information on the structure, thickness and interface roughness of the individual layers (see Supplementary Fig. 1). The individual layers and multilayers show broad polycrystalline peaks when investigated by XRD as observed earlier for similarly grown films.46 X-ray contrast between Dy and Tb is very poor due to their adjacent atomic numbers, hence a multilayer structure could not be properly studied using XRR.

Magnetometry. Conventional in-plane magnetization measurements were performed at various temperatures and fields using a superconducting quantum interference device (SQUID) from Quantum Design (MPMS-XL). Conventional AC field susceptibility measurements were acquired at various temperatures and frequencies using a physical property measurement system (PPMS) from Quantum Design.

Polarized neutron scattering. PNR measurements were performed at the reflectometer MARI A at MLZ, Germany47. In-plane magnetization fields of $10 \text{KOE}$ ($1000$ mT) and $0 \text{Oe}$ ($0$ mT) were used before the samples were cooled to $10 \text{K}$ separately in a cryostat and measured at applied fields $H_\parallel = -0.01 \text{KOE}$ ($-1$ mT) and $-10.0 \text{KOE}$ ($-1000$ mT). From the neutron polarization analysis we resolve different components of the magnetization within the film plane. The saturation field of approximately $50.0 \text{KOE}$ of the system exceeded the experimentally available field of $10.0 \text{KOE}$ ($1000$ mT) at MARI A. The SLDs of a specimen are given by the nuclear ($\rho_n$) and magnetic ($\rho_m$) components. Four different cross sections were measured, namely non-spin flip (NSF) scattering: ($\rho_n \pm \rho_m \cos \phi_m$) represented by $R_+ \text{ and } R_-$ and spin flip (SF) scattering: ($\rho_m \sin^2 \phi_m$) represented by $R_+ \text{ and } R_-$. Here, $\pm$ signs are used to distinguish the intensity contributions $R$ representing a polarization component parallel or anti-parallel to the guiding field, respectively. The angle $\phi_m$ is between the magnetization $M_{\parallel}$ and the applied field $H_\parallel$. The spin dependent NSF reflectivities are given by

$$R_+ = \frac{1}{2} [R_+(1 + \cos \phi_m) + R_-(1 - \cos \phi_m)]^2$$ (7)
The source codes used for neutron scattering code availability.

The NSF approves the code availability.

The original source codes used for neutron scattering.

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Author contributions
J.Y. assisted T.B. in sample preparation. N.P. and A.P. did the X-ray measurements. T.B. and J.Y. did the DC and AC magnetization measurements while A.P. did the TRM measurements. J.Y. and S.M. did the PNR measurements from a beamtime proposal of A.P. All authors reviewed the manuscript. A.P. conceived and designed the work, analyzed all datasets, coordinated the work, and wrote the manuscript.

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