# SUPPLEMENTAL MATERIAL

## Chiral coupling between magnetic layers with orthogonal magnetization

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#### SM 1. Calibration protocol of $\theta_B$

The field sweep and angle scan measurements reported in Fig. 3(a) and 3(c) require an accurate calibration of the angle  $\theta_B$  between the z-axis and external field. To perform such a calibration, we apply a large out-of-plane field in excess of the saturation field of the in-plane top Co layer (e.g., 1.5 T). With such field, the magnetizations of both layers are aligned with the external field along the z-axis. We then rotate the field within a small range of angles, e.g.,  $\pm 10^{\circ}$ . Since the anomalous Hall effect signal is maximum at  $\theta_B = 0^{\circ}$ , we find the maximum signal and set it as a new reference for  $\theta_B = 0^{\circ}$ . We repeat the same procedure by decreasing the range ( $\pm 5^{\circ}$ ,  $\pm 3^{\circ}$ , etc.) and the angle steps progressively until achieving a precise calibration of the angle as shown in Fig. S1.



**Figure S1**. Schematic representation of the calibration procedure of  $\theta_B$  and representative data of the anomalous Hall resistance in a field of 1.5 T after the calibration.

### SM 2. Measurement of the DMI field as a function of angle

Here we demonstrate an alternative experimental scheme to measure  $B_{\rm DMI}$  acting on  $M_{\rm TbFe}$  or, more in general, on a layer with OOP magnetization. In this measurement, the magnetic field is rotated about D, as shown in Fig. S2(a). The amplitude of the field is set to 200 mT, which is larger than  $B_c$  but much lower than the OOP saturation field of  $M_{\rm C0}$ . In Fig. S2(b) we report  $R_{\rm H}$  for the clockwise (cw) and counterclockwise (ccw) rotation of  $\theta_B$  about D measured on TbFe/Pt(1.2 nm)/Co. During the clockwise rotation, we expect that  $M_{\rm TbFe}$  reverses from up to down (down to up) upon crossing  $\theta_B =$  $90^{\circ}$  ( $\theta_B = 270^{\circ}$ ) when the OOP component of the external field overcomes  $B_c$ . For the counterclockwise rotation, the sign of the reversal should simply invert. Moreover, in the absence of DMI, the reversal events for clockwise and counterclockwise scans should be symmetric with respect to  $\theta_B = 90^\circ$  and 270°. Instead, we observe a clear shift of the reversals towards smaller angles, which indicates that the system switches from an unfavored to a favored configuration when the magnetization rotates in the clockwise direction. This shift agrees with the smaller coercivity observed along  $D \times z$  in the field sweep data, as reported in Fig. 3(a). The information obtained from the angle scan measurements is consistent with that derived from the field sweeps discussed in the main text, thus confirming the influence of the interlayer DMI coupling on  $M_{\text{TbFe}}$ .



**Figure S2**. (a) Schematic representation of the angle scan in the clockwise (cw) and counterclockwise (ccw) directions. (b) Hall resistance of TbFe(8)/Pt(1.2)/Co(3) measured during the rotation of the magnetic field about D.

#### SM 3. Macrospin simulations of the influence of the interlayer DMI on magnetization reversal

We performed macrospin simulations of the interlayer DMI coupling between a perpendicularly magnetized layer (1) and an in-plane layer (2) with a custom-written code using MATLAB. Here, layers 1 and 2 represent the TbFe bottom film and the top Co film, respectively. The total energy of the system comprises the contributions from the Zeeman, magnetic anisotropy, demagnetization, RKKY and DMI energies, and reads:

$$E = -\mathbf{M}_{1} \cdot \mathbf{B} - \mathbf{M}_{2} \cdot \mathbf{B} - \frac{1}{2}k_{1}\sin^{2}\theta_{1} - \frac{1}{2}k_{2}\sin^{2}\theta_{2} - \frac{1}{2}\mu_{0}M_{S,1}^{2}\cos^{2}\theta_{1} - \frac{1}{2}\mu_{0}M_{S,2}^{2}\cos^{2}\theta_{2} + \frac{1}{2}k_{3}\cos^{2}\varphi_{2} - \sigma \mathbf{m}_{1} \cdot \mathbf{m}_{2} - \mathbf{D} \cdot \mathbf{m}_{1} \times \mathbf{m}_{2}.$$
(S1)

Here,  $M_i = M_{S,i} m_i = M_{S,i} [\sin\theta_i \cos\varphi_i, \sin\theta_i \sin\varphi_i, \cos\theta_i]$  is the magnetization of the *i*<sup>th</sup> layer, with saturation magnetization  $M_{S,i}$ ,  $k_{1,2}$  are the perpendicular magnetic anisotropy energy densities of the two layers, and  $k_3$  is the uniaxial in-plane anisotropy energy density of the 2<sup>nd</sup> layer.  $\sigma$  is the RKKY energy density and **D** is the DMI vector, which we assume to be along – *y*.

For each magnetic field magnitude and direction, the equilibrium orientation of  $M_1$  and  $M_2$ was obtained by iteratively minimizing Eq. (S1). The simulations presented here were obtained with the following set of parameters:  $M_{S,1} = 0.45 \times 10^6$  A/m,  $M_{S,2} = 1.1 \times 10^6$  A/m,  $k_1 = 350$  kJ/m<sup>3</sup>,  $k_2 = -50$ kJ/m<sup>3</sup>, D = 10 kJ/m<sup>3</sup>. For simplicity, we neglected the RKYY coupling, which is zero for orthogonal magnetizations, and the uniaxial in-plane anisotropy ( $k_3 = \sigma = 0$ ). Representative results simulating the field sweeps shown in Fig.3 of the main text and the angle scan in Fig. S2 are presented in Fig. S3. The simulations closely reproduce the measurements and provide a better understanding of the orientation of the two layers. As an example, let us consider a field sweep with the field tilted at  $\theta_B =$  $15^{\circ}$  along  $-D \times z$  [initial  $\varphi_B = 0^{\circ}$ , black curve in Fig. S3(b)]. At 150 mT,  $M_1$  and  $M_2$  are oriented upright, a configuration that frustrates the DMI because the latter favors the orientations up-left and downright (we define the orientation of  $M_2$  by looking at the *xz* plane from -y to +y). When the field is reduced,  $M_2$  rotates towards -x driven by the DMI, which enforces the configuration up-left. Thus, a large field is required to switch  $M_1$  from up to down and bring back the coupled layers in a configuration that opposes the DMI. As the field is increased from -150 mT to 0,  $M_2$  is released and turns to +x. Since the new down-right orientation is promoted by the DMI, a large field must be applied to switch  $M_1$  upward, symmetrically to the down-to-up switching. The opposite situation is realized if the field is swept at  $\theta_B = 15^\circ$  and tilted along  $D \times z$  [initial  $\varphi_B = 180^\circ$ , black curve in Fig. S3(a)]. In this case,  $M_1$ and  $M_2$  are initially oriented up-left, which is a configuration favored by the DMI. As the field becomes negative,  $M_2$  is forced to rotate towards x, which brings the system in a configuration that is unfavored by the DMI. Thus, a smaller field is required to switch  $M_1$  from up to down and bring back the coupled layers in a configuration that favors the DMI. A similar situation occurs as  $M_1$  switches from down to up as the field returns positive. We can interpret by the same argument the simulations of the angle scan and AMR in Fig. S3(c,d).



**Figure S3.** (a) Definition of the reference system for field and angle sweep simulations. (b) Macrospin simulation of field sweeps tilted at  $\theta_B = 15^\circ$  along  $-D \times z$  (initial  $\varphi_B = 0^\circ$ , black curve) and along  $D \times z$  (initial  $\varphi_B = 180^\circ$  red curve). The black (gray) arrow shows the orientation of layer 1 (2). (c) Coercivity difference  $\Delta B_c$  as a function of  $\varphi_B$ . (d) Simulation of an angle scan of the external field about  $D \parallel -y$ . (e) Simulation of the anisotropic magnetoresistance during a field scan at  $\theta_B = 89.5^\circ$  and  $\varphi_B = 0^\circ$ , with  $M_1$  oriented along +z (blue curve) and -z (red curve).

### SM 4. Pt Spacer thickness dependence of the TbFe coercivity

In an effort to understand the deviations in the DMI measured on the TbFe layer (main text, Fig. 4 (a)) we plot the coercivity ( $B_c$ ) of TbFe as a function of the Pt spacer thickness (Fig. S4). As evident from the data, the samples with Pt(1 nm) and Pt(2 nm) spacer show significant deviations from the overall linearly increasing  $B_c$  trend as a function of  $t_{Pt}$ . Such deviations suggest that the properties of these TbFe layers might be different than the remaining samples in the batch. The magnetic properties as well as the interlayer DMI is known to be highly sensitive to the interfaces of the ferromagnets with Pt, which might exhibit local fluctuations due to the influence of substrate, sputtering process and fabrication-related issues. We believe that one or several of such factors played a role for the mentioned two samples and resulted in a different coercivity and interfacial DMI simultaneously.



**Figure S4.** Coercivity  $(B_c)$  of the TbFe layer as a function of the Pt spacer thickness.