Supplementary Information

Extraordinary Chiral Exchange-Bias Phenomenon: Engineering the Sign of the Bias Field in Orthogonal Bilayers by a Magnetically Switchable Response Mechanism

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The novelty of the studied phenomenon, i.e. the unprecedented possibility of isothermally inducing a bias field with control on both magnitude and sign, has driven us to carry a systematic study in order to understand the causes behind its origin and unveil the main conditions required to set it up. On this basis, we demonstrate in the following material a fundamental requirement: the exceptional exchange bias (EB)-like phenomenon is the result of the combination of the uniaxial anisotropy of the CoFeB layer (induced by oblique growth) and the unidirectional anisotropy resulting from the magnetostatic coupling between the magnetically hard and soft layers.

In addition, this Supplementary Information also presents x-ray diffraction pattern (XRD) of the reference $SmCo_5$ film confirming c-axis growth of the hexagonal phase and AFM/MFM images measured for both $SmCo_5$/spacer/CoFeB systems under study.

Texture of the magnetically hard $SmCo_5$ layer

Fig. S1 shows the XRD pattern in standard Bragg-Brentano geometry (Bruker Advance D8) of a reference $SmCo_5$ sample as shown in Fig. 1a. The (0006) peak of the Al_2O_3 substrate is clearly visible on which Ru grows with (0002) orientation. Reflections corresponding to the presence of Sm-oxide are also found as previously reported by coauthors of this work, which did not have any influence on the texture of the film.[27] The $SmCo_5$ phase is identified by its c-axis oriented (0.0.l) lattice planes. Other $SmCo_5$ crystal orientations are not detectable, confirming the expected c-axis growth of the hexagonal phase.

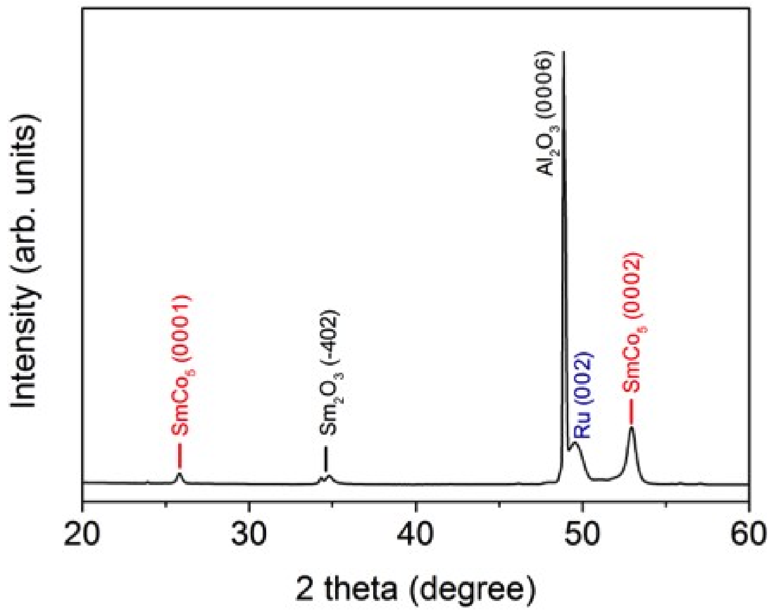

Fig. S1 XRD measurement of the $SmCo_5$ reference film (as shown in Fig. 1a).

The thickness (30 nm) of the $SmCo_5$ film has been chosen based on previous results by coauthors of this work,[27,28] which showed that excessively thin (below 10 nm) and thick (100 nm) films resulted in a detrimental effect on texture and magnetic properties. In the first case authors explained that partial structural disorder of the 1:5 phases might be responsible of the achieved moderate perpendicular permanent magnet properties, while the formation of nanocrystalline, randomly oriented Sm–Co grains might be the reason for not achieving optimized properties when going to excessively thick films. A thickness of about 30 nm allowed for achieving optimized perpendicular permanent magnet properties due to a single epitaxial orientation to the Ru buffered Al_2O_3 (0001) substrate with perpendicular alignment of the c –axis.[27]

Study of CoFeB/spacer/$SmCo_5$ multilayers with different initial magnetic configurations

With this objective in mind, specific samples were designed and grown following the preparation conditions described in the main manuscript and using identical substrates as there indicated for the reference samples and the complete hard/spacer/soft structures.
Two complementary samples were grown to proof that the observed EB-like phenomenon is the result of the combination of the uniaxial anisotropy from the CoFeB layer and the dipolar field created by the out-of-plane magnetization of the $SmCo_5$ layer:

- $SmCo_5$/spacer/CoFeB structure with $SmCo_5$ saturated out-of-plane and the CoFeB layer grown with vertical incidence (in comparison with the rest of samples where the soft magnetic layer was grown with oblique incidence) [Fig. S2]. Structure of the sample:

  Al_2O_3(0001)/Ru(20nm)/$SmCo_5$(30nm)/spacer(12.8nm)/CoFeB(3nm)-*VERTICAL*/Pt(1.8nm)

  Firstly, a reference CoFeB layer [structure: Si/SiO_2/Pt(1.8nm)/CoFeB(3nm)/Pt(1.8nm)] was grown with vertical incidence. Figure S1a shows the isotropic nature of this CoFeB film when grown vertically. Afterwards, and also with vertical incidence, a CoFeB film with same thickness was grown as part of $SmCo_5$/spacer/CoFeB structure, with $SmCo_5$ previously saturated out-of-plane. As shown in Fig. S2b, a practically isotropic behavior is observed in the angular evolution of coercivity. This result allows concluding that inducement of a uniaxial anisotropy in the CoFeB is a first requirement to attain a preferential orientation in the multilayer system.

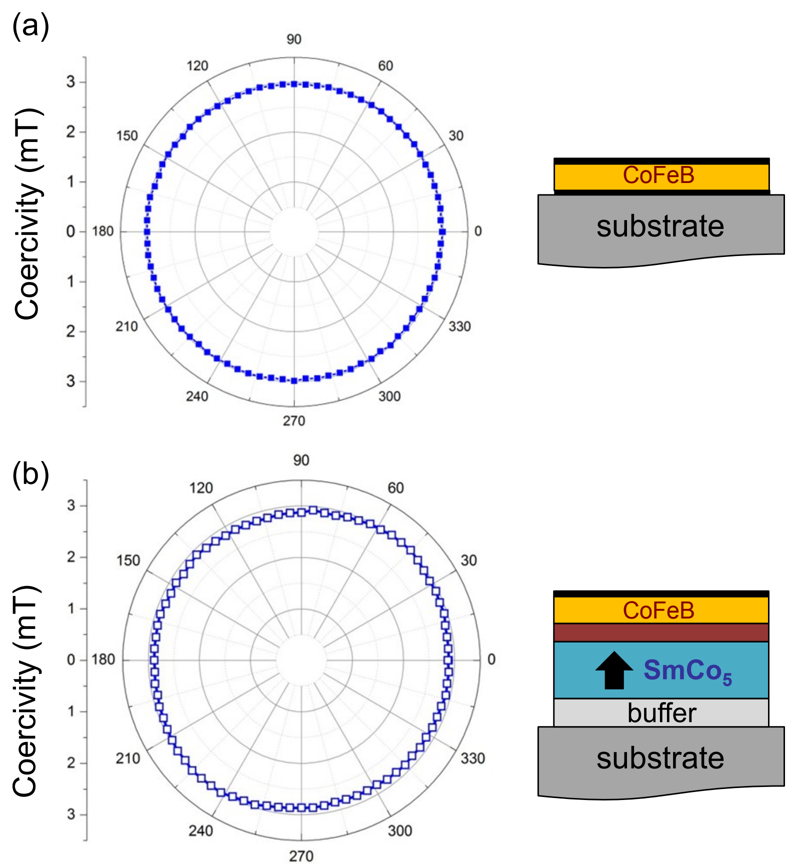

Fig. S2 Angular evolution (polar-plot representation) of coercivity and schematic representation of the corresponding multilayer structure (right-hand side) for: (a) CoFeB (3 nm) reference film grown with vertical incidence with the structure [Si/SiO_2/Pt(1.8nm)/CoFeB(3nm)/Pt(1.8nm)]; and (b) $SmCo_5$(30nm)/spacer(12.8nm)/CoFeB(3nm) with $SmCo_5$ saturated out-of-plane prior to growth of the CoFeB film (vertical incidence).

- $SmCo_5$/spacer/CoFeB structure with the $SmCo_5$ layer in a demagnetized state (in comparison with the rest of samples where it was saturated out-of-plane) and the CoFeB layer grown with oblique incidence [Fig. S3]. Structure of the sample:

Al_2O_3(0001)/Ru(20nm)/$SmCo_5$(30nm)-*DEMAG.*/spacer(12.8nm)/CoFeB(3nm) /Pt(1.8nm)

Figure S2a shows the result of the uniaxial anisotropy induced in a reference CoFeB layer [structure: Si/SiO_2/Pt(1.8nm)/CoFeB(3nm)/Pt(1.8nm)] grown by oblique incidence.

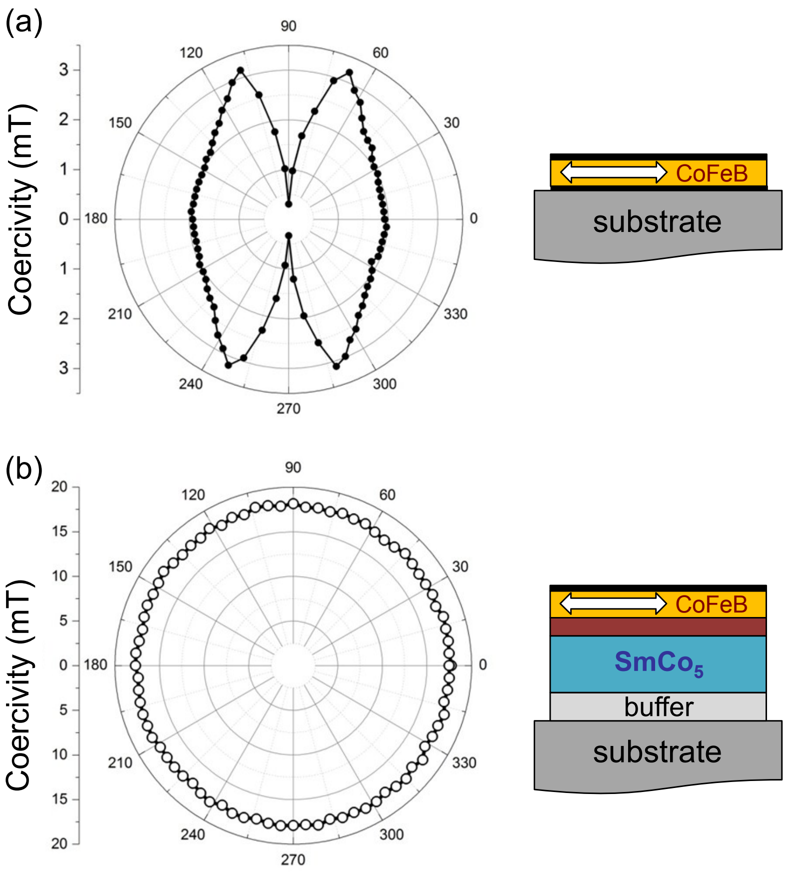

Fig. S3 Angular evolution (polar-plot representation) of coercivity and schematic representation of the corresponding multilayer structure (right-hand side) for: (a) CoFeB (3 nm) reference film grown with oblique incidence with the structure [Si/SiO_2/Pt(1.8nm)/CoFeB(3nm)/Pt(1.8nm)]; and (b) $SmCo_5$(30nm)/spacer(12.8nm)/CoFeB(3nm) with $SmCo_5$ in a demagnetized state and CoFeB grown with oblique incidence.

However, growth of a CoFeB layer under identical conditions on a demagnetized $SmCo_5$ layer (spacer-mediated) results in isotropic behavior of the system (Fig. S3b). This result allows concluding that saturation of the $SmCo_5$ layer is a second requirement to attain a preferential orientation in the multilayer system. Moreover, it is worth remarking the high coercivity of 18 mT (the highest obtained among the samples under study) achieved in this system, which is consistent with much larger stray fields in the close vicinity of the CoFeB layer, as a consequence of the breakage into magnetic domains (fine-scaled domain structure) in the demagnetized $SmCo_5$ layer.[28]

Magnetic force microscopy results

Magnetic force microscopy (MFM) images are shown for $SmCo_5$/spacer/CoFeB with a thickness of the spacer of 4.3 nm (Fig. S4a) and 12.8 nm (Fig. S4b) measured at remanence. Atomic force microscopy (AFM) images of both samples are show on the left-hand side for the sake of completeness. The length scale of the MFM patterns is poorly defined but the possibility of observing magnetic contrast variations for both samples suggests some tilting of the magnetization, which could originate from the dipolar field of the epitaxial $SmCo_5$ film (strong out-of-plane magnetic anisotropy). An increased thickness of the spacer between the magnetically hard and soft layers will weak the strength of the dipolar field at the position of the soft magnetic layer and thus will have an increased in-plane component.

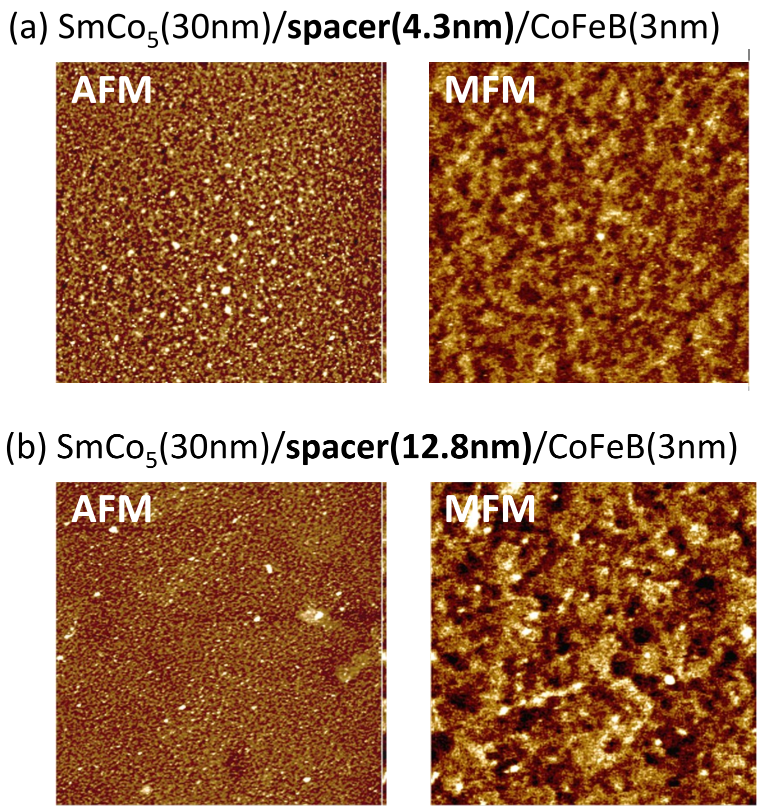

Fig. S4 AFM/MFM images for $SmCo_5$/spacer/CoFeB with a thickness of the spacer of (a) 4.3 nm and (b) 12.8 nm at remanence. Scan area: 20 μm x 20 μm.