Supplementary Material

Interfacial Dzyaloshinskii-Moriya interaction in the epitaxial W/Co/Pt multilayers

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1. Description of the RHEED pattern

The RHEED pattern is depicted in Figure 1a in the manuscript. The sharp and uniform streaks from the Pt buffer confirm its high quality fcc $Fm\bar{3}m$ 111 crystalline structure. A more complex zig-zag-like RHEED pattern from W layer, containing blurred two sub-patterns with the dashed streaks, originates from texture growth [1] in bcc $Im\bar{3}m$ 110 direction and rotated azimuthal orientation. This superposition of the two patterns is caused by the randomly distributed crystallites domains rotated in the sample plane by 30° (assuming symmetry of the layers

below) or 90° (since 60° is off diffraction angle for the low index atom rows and only 90° may result in the zig-zag diffraction pattern). Each diffraction pattern originates from the two perpendicular azimuths: $<1\bar{1}1>$ with $<\bar{1}12>$ and $<002>$ with $<1\bar{1}0>$ (in the sample plane) every 30° respectively. The more blurred and dashed pattern may result from scattering at the surface roughness and defects with dimensions smaller than the coherent length of the RHEED e-beam. The blurry and elongated streaks from the Co layer (fcc $Fm\bar{3}m$ 111) are also visible every 30° (along azimuths <112> and <110> every 60°, respectively) evidencing the closed packed surface. However, due to similarity of the RHEED pattern, the bulk-like hcp-$P6_3/mmc$ 0001 structure occurrence cannot be confirmed unambiguously. Finally, growth of the Pt cap layer resembles that of the buffer. The slightly blurred streaks reflect the less perfect crystalline structure due to the smaller thickness and the different layer located underneath. The specific crystal symmetry parameters deduced from the described above characteristics are listed in the manuscript in Table 1 for all the evaporated component layers. This layer growth scenario allows for an insight into the structure of each component layer of the W/Co/Pt multilayer stack.

2. Determination of DMI

Using the calculation scheme proposed by Legrand *et al.* [2], we assume that the configuration of parallel stripe domains observed by MFM at remanence corresponds to the minimum energy of the investigated systems. Then it is possible to determine the domain wall energy and DMI strength from the domain structure periodicity. K_{eff} anisotropy coefficient is estimated from the H_k field value required for magnetization saturation of the multilayers in their plane. The relation between K_{eff} and K_u is the following:

$$K_{eff} = K_u - {}^{1}/_{2}\mu_0 M_s^2 \qquad \text{(S1)}$$

where K_{eff} is effective anisotropy coefficient, K_u– uniaxial anisotropy coefficient and M_s – magnetization saturation.

All the conditions satisfying applicability of the model are fulfilled: $\Delta \ll W$, $P \ll W$ and $P \ll 2\pi\Delta$, where: Δ is the domain wall width, P is periodicity of the multilayer stacks and W is the domain width [3]. To calculate the parameters describing the multidomain state, the whole multilayered structure is treated as an effective magnetic medium with the uniformly distributed dilute moments. According to this approach the parameters characterizing magnetic structure are scaled by a factor $\frac{t_m}{t_{stack}} = f$ = 0.24 (=0.69/2.87):

$$\frac{M_S^{'}}{M_S} = \frac{A^{'}}{A} = \frac{D^{'}}{D} = \frac{K_u^{'}}{K_u} = \frac{K_{eff}^{'}}{K_{eff}} = f \qquad \text{(S2)}$$

where A is the exchange stiffness constant, D is the DMI strength, t_m is the Co layer thickness and t_{stack} is the total thickness of the basic trilayer stack. The symbols with apostrophe correspond to the scaled values.

In the analysed case, the domain wall is considered as a monodomain magnetic body with the infinite length, expanding across the multilayered structure. A shape of elliptic cylinder [4,5] is assumed as a good approximation for calculating demagnetizing fields. Then the domain wall width is given by the expression:

$$\Delta = \sqrt{\frac{A^{'}}{K_u^{'} + \frac{\mu_0 M_s^{'2}(t' - 2\Delta)}{2 \quad (t' + 2\Delta)}}} \qquad \text{(S3)}$$

where, $t^{'} = (T * f)$ and T is total thickness of the magnetic multilayer starting from the bottom ferromagnetic layer to the top ferromagnetic layer.

By assuming all the energy contributions, the domain wall energy (σ_{dw}) can be expressed by:

$$\sigma_{dw} = {2A^{'}}/{\Delta} + 2K_u^{'}\Delta - \pi D^{'} + \frac{\mu_0 M_s^{'2}}{2} 2\Delta\left(\frac{t^{'} - 2\Delta}{t^{'} + 2\Delta}\right) \quad \text{(S4)}$$

On the other hand, the domain wall energy in the effective medium approach can be determined from minimization of the total energy of the system relative to the domain structure periodicity (=$2W$):

$$\sigma_{dw} = \mu_0 M_s^{'2} \frac{(2W)^2}{t^{'}} \frac{1}{\pi^3} \sum_{n \geq 1, odd}^{\infty} \frac{1}{n^3}\left(1 - e^{-\pi n t^{'}/W} - \frac{\pi n t^{'}}{W} e^{-\pi n t^{'}/W}\right) \quad \text{(S5)}$$

Comparing equations (S4) and (S5) and implementing the domain wall width Δ, determined from equation (3), the parameter $D^{'}$ can be estimated. Then D can be calculated as $D = D^{'}/f$.

A critical threshold value D_{thr} above which a pure Néel type domain walls are formed is defined by the following equation [3].

$$D_{thr} = \frac{2 * \mu_0 M_S^{'2}}{\frac{\pi}{t^{'} + log2} + \pi\sqrt{\frac{K_u^{'} + \frac{\mu_0 * M_s^{'2}}{2}}{A^{'}}}} \quad \text{(S6)}$$

3. Details of the micromagnetic simulations

The influence of DMI energy on the magnetic domain pattern formation is illustrated by the results of micromagnetic calculations performed for the multilayered model, using the mumax3 software [6]. The domain structure ground state of the 60 nm thick W/Co/Pt layered film has been simulated by direct energy minimization. The initial state of magnetization is taken to be randomly oriented around +z direction (perpendicular to film plane). The simulation area with the periodic boundary conditions is set to 512 nm × 512 nm in the x-y plane. The cell size is taken to be equal to 2 nm × 2 nm in the x-y plane, and 0.6 nm in the z-direction, which is smaller than the exchange length of Co material. The simulated model assumes the layered structure

with 20 repetitions of the Co magnetic layers 0.6 nm thick, separated by the nonmagnetic 1.2 nm thick W and Pt layers.

The thickness of nonmagnetic slabs in simulations is slightly larger than the nominal value (1.0 nm for W and Pt) to allow us application of the 0.6 nm cell size along the thickness direction (z). The input material parameters used for the simulations are extracted from the experimentally obtained magnetization curves (parameters listed in the manuscript in Table 2). The equilibrium domain width W in the multilayers is simulated for D ranging from 0.5 mJ/m^2 to 6.0 mJ/m^2. A finite temperature effect and sample imperfections have not been considered in these simulations.

4. Details of the DFT calculations

The first-principles DFT calculations have been performed using the VASP [7] code based on the plane wave basis set and projector augmented wave method [8]. The calculation is fully relativistic by considering SOC. A plane-wave energy cut-off of 350 eV is applied. For the exchange-correlation functional, the GGA approximation [9] is adopted since it gives good results in the simulation of magnetic metallic multilayers [10,11].

To calculate the DMI vector, the same procedure and notation suggested by Yang *et al.*[12] are adopted. The supercells contain 4 in-plane translated unit cells (along the x direction). The calculations are performed using 3×12×1 k-points centred in Γ. It was shown earlier that DMI is mainly an interface-sensitive effect in these metallic multilayers. DMI does not change beyond 4 monolayers thickness of the ferromagnetic metal or heavy metal [13]. The conventional clockwise (CW) and anticlockwise (ACW) spin configurations are defined in agreement with Ref. [12]. The same strategy to calculate DMI at the interface [14], bulk frustrated [15] and insulating chiral-lattice-magnets [16] has been used commonly. The total DMI energy d^{tot} is calculated as

the difference in the total energies of the opposite chirality of spin configurations E_{CW} and E_{ACW} for a given value of cycloid wavelength:

$$d^{tot} = (E_{CW} - E_{ACW})/m \quad \text{(S7)}$$

where E_{CW} and E_{ACW} are the total energies of the CW and ACW spin configurations, respectively.

The number m depends on the wavelength of the cycloid. In the case considered here, the value of $m = 12\sqrt{3}$ is taken because the magnetic cycloid with wavelength 4 is simulated, as discussed by Yang *et al.* [12]. The global effect on the multilayer can be expressed in terms of the micromagnetic energy per volume unit [17,18]. The DMI coefficient D can be related to the total DMI energy d^{tot} by the following equation:

$$D = 3\sqrt{2}d^{tot}/(N_F a^2) \quad \text{(S8)}$$

where a is the in-plane lattice constant and N_F represents the number of ferromagnetic atomic layers.

The layer resolved DMI vector amplitude, presented in Figure 7b in the manuscript, is obtained from the corresponding DFT energies E_{CW}^{k} and E_{ACW}^{k} using equation (7).

References

1 M. A. Herman, W. Richter and H. Sitter, *Epitaxy*, Springer Berlin Heidelberg, Berlin, Heidelberg, 2004, vol. 62.

2 W. Legrand, J.-Y. Chauleau, D. Maccariello, N. Reyren, S. Collin, K. Bouzehouane, N. Jaouen, V. Cros and A. Fert, *Sci. Adv.*, 2018, **4**, eaat0415.

3 I. Lemesh, F. Büttner and G. S. D. Beach, *Phys. Rev. B*, 2017, **95**, 174423.

4 J. A. Osborn, *Phys. Rev.*, 1945, **67**, 351–357.

5 A. Mougin, M. Cormier, J. P. Adam, P. J. Metaxas and J. Ferré, *Europhys. Lett.*, 2007, **78**, 57007.

6 A. Vansteenkiste, J. Leliaert, M. Dvornik, M. Helsen, F. Garcia-Sanchez and B. Van Waeyenberge, *AIP Adv.*, 2014, **4**, 107133.

7 G. Kresse and J. Furthmüller, *Comput. Mater. Sci.*, 1996, **6**, 15–50.

8 G. Kresse and J. Furthmüller, *Phys. Rev. B*, 1996, **54**, 11169–11186.

9 J. P. Perdew, K. Burke and M. Ernzerhof, *Phys. Rev. Lett.*, 1996, **77**, 3865–3868.

10 C. Autieri and B. Sanyal, *J. Phys. Condens. Matter*, 2017, **29**, 465802.

11 C. Autieri, P. A. Kumar, D. Walecki, S. Webers, M. A. Gubbins, H. Wende and B. Sanyal, *Sci. Rep.*, 2016, **6**, 29307.

12 H. Yang, A. Thiaville, S. Rohart, A. Fert and M. Chshiev, *Phys. Rev. Lett.*, 2017, **118**, 219901.

13 A. Hajr, A. Hariri, G. Manchon, S. Ghosh and A. Manchon, *Phys. Rev. B*, 2020, **102**, 224427.

14 A. Soumyanarayanan, M. Raju, A. L. Gonzalez Oyarce, A. K. C. Tan, M.-Y. Im, A. P. Petrović, P. Ho, K. H. Khoo, M. Tran, C. K. Gan, F. Ernult and C. Panagopoulos, *Nat. Mater.*, 2017, **16**, 898–904.

15 H. J. Xiang, E. J. Kan, S.-H. Wei, M.-H. Whangbo and X. G. Gong, *Phys. Rev. B*, 2011, **84**, 224429.

16 J. H. Yang, Z. L. Li, X. Z. Lu, M.-H. Whangbo, S.-H. Wei, X. G. Gong and H. J. Xiang, *Phys. Rev. Lett.*, 2012, **109**, 107203.

17 J. Sampaio, V. Cros, S. Rohart, A. Thiaville and A. Fert, *Nat. Nanotechnol.*, 2013, **8**,

839–844.

18 A. Fert, V. Cros and J. Sampaio, *Nat. Nanotechnol.*, 2013, **8**, 152–156.