# Chirality-induced asymmetric magnetic nucleation in Pt/Co/AlOx ultrathin microstructures: Supplemental Materials

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### I. EXPERIMENTAL SETUP AND PROCEDURE

Measurements were carried out using a wide-field Magneto-Optical Kerr Effect microscope in polar geometry using a 630 nm LED. Figure 1(a) shows the variation of the nucleation field  $H_{z,n}$  at the edge of the sample, measured for two values of the applied  $H_z$  pulse length, 20 ms and 500 ms. For a fixed  $H_x$  field, we have (arbitrarily) defined as the *nucleation field* the  $H_z$  field for which 10-15 domains nucleate systematically at the edge. The experimental procedure was the following: with the  $H_x$  field switched on, a  $H_z$  pulse of fixed length was applied. The  $H_x$  field was then switched off, and an  $H_z$  field pulse was applied to allow the created domain to enlarge, via domain wall propagation, to a size larger than the spatial resolution of the Kerr microscope. In order to account for the unavoidable misalignment of the  $H_x$  electromagnet that can introduce a weak  $H_z$  component adding to the  $H_z$  pulse, the procedure was carried out twice, starting from positive and negative magnetic saturation, and the presented values are the average of the two measurements.

Note that a larger error bar is associated to  $H_{z,n}$  corresponding to the smallest  $H_x$  values. This is due to the fact that when the nucleation field for domains within the film becomes smaller than that at the edges, the observation of domain nucleation at the edge is hindered by the expansion of the domains within the film. The criterium which we have used to define  $H_{z,n}$  is more difficult to obtain, making the estimation of this field subject to larger errors. It should be noted that several samples with the same magnetic properties have been measured using the same experimental procedure. The minimum field for which the  $H_{z,n}$ field is obtained depends on the number and position of the defects within the film.

We have shown in the manuscript that the nucleation field depends on the length of the  $H_z$  pulse through the coefficient 1/p where  $p = \ln(\tau/\tau_0)$ . For the two  $H_z$  pulses of 20 ms and 500 ms, p is respectively 16.8 and 20 when assuming  $\tau_0 = 1$  ns, as usually found in the literature. Figure 1(b) shows the  $H_{z,n}$  values multiplied by the respective p values. The invariance of the results with respect to p, within the experimental error bars, is in agreement with the theoretical model.



FIG. 1. (a) nucleation fields  $H_{z,n}$  measured as a function of  $H_x$  for two values of the applied  $H_z$  pulse length, 20 ms and 500 ms. (b) the data in (a) have been multiplied by p, respectively 16.8 and 20.

### II. ZERO TEMPERATURE MODEL

We consider a semi-infinite 1D model to describe the magnetization profile in the vicinity of an edge of the sample. Let x be the coordinate normal to the edge, and z the easy magnetization axis. As the field is applied in the (x, z) plane, and as DMI forces a magnetization rotation in this plane too, the single unknown is the angle  $\theta(x)$  of the magnetization with the easy axis. The energy density reads then

$$E = A \left(\frac{d\theta}{dx}\right)^2 - D \frac{d\theta}{dx} + K_0 \sin^2 \theta - \mu_0 M_s \left(H_x \sin \theta + H_z \cos \theta\right)$$
(1)

where  $K_0 = K_u - \mu_0 M_s^2/2$  is the effective anisotropy constant including the shape anisotropy. Writing down the Lagrange-Euler equations for the minimization of the total energy and integrating back gives the following first integral

$$A\left(\frac{d\theta}{dx}\right)^2 = K_0 \sin^2 \theta - \mu_0 M_s \left(H_x \sin \theta + H_z \cos \theta\right) + C^{st}$$
(2)

Infinitely inside the sample, the angle is constant ( $\theta_0$  in the following) so that the energy reduces to a macrospin one:

$$K_0 e(\theta) = K_0 \sin^2 \theta - \mu_0 M_s (H_x \sin \theta + H_z \cos \theta)$$
(3)

The angle  $\theta_0$  minimizes this energy, which is solved by the standard Stoner-Wohlfarth problem under a general field [1, 2].

Thus, Eq. (2) can be re-expressed as

$$A\left(\frac{d\theta}{dx}\right)^2 = K_0 \left[e(\theta) - e(\theta_0)\right] \tag{4}$$

the right-hand side being indeed positive as  $\theta_0$  minimizes  $e(\theta)$ .

At the edge of the sample, the micromagnetic boundary condition with DMI reads [3]

$$2A\frac{d\theta}{dx} = D\tag{5}$$

Let us call  $\theta_1$  the magnetization angle at the edge. From Eq. (4) and Eq. (5) we have

$$e(\theta_1) - e(\theta_0) = \frac{D^2}{4AK_0} \equiv \left(\frac{2}{\pi}\frac{D}{D_{c0}}\right)^2 \tag{6}$$

This shows that the problem reduces to a two-macrospin problem. The first macrospin  $\theta_0$  is independently solved and an analytical solution can be found [1, 2]. Concerning the second macrospin (edge magnetization), Eq. (6) shows that its energy is larger than the previous one which lets expect a switching at smaller fields than for  $\theta_0$ , or in other words, an easier magnetization switching at the edges than inside the film.

Around the metastable solution for  $\theta_0$ , two solutions for  $\theta_1$  can be found which correspond to the two edges,  $\theta_1 - \theta_0$  being of the sign of D on the right edge (see Eq. 5). If the symmetry is broken (H arbitrarily oriented in space), the two solutions will disappear (which corresponds to a nucleation event) at different applied field magnitudes, revealing the chirality of edge nucleation process. Note that the easiest branches are iso-barrier curves of the Stoner-Wohlfarth problem. These have already been investigated, when studying the switching field of single-domain nanoparticles at non-zero temperatures, both experimentally through



FIG. 2. (a) Nucleation field  $H_z$  versus  $H_x$  at the edges of the sample, obtained for D = 0 and  $D = D_{c0}/2$  from Eq. 6. For  $D \neq 0$  two branches are obtained, depending on the considered edge orientation. The dots correspond to the micromagnetic simulation including complete dipolar coupling (it has been checked that, in the local demagnetizing field approximation, simulations are in perfect agreement with the model). (b) Sketch of the calculation using Eq. 6 for two different field magnitudes, and for a DMI energy scale indicated by the distance between the two dotted lines. The solutions for  $\theta_1$  are the intersection between the upper dotted line and the full curves (indicated by the arrows). For the lower field magnitude (in black), two solutions for  $\theta_1$  are found corresponding to both edges. For the higher one (in red), one solution disappears indicating that one edge magnetization is unstable and that nucleation occurs.

single particle measurements using the micro SQUID technique [4], and theoretically in the macrospin model [5]. These curves are simply shrunk astroids. The solutions for  $D/D_{c0} = 0$  and 0.5 are shown in Fig. 2.

This model neglects dipolar couplings (only the shape anisotropy is taken into account), as generally done for ultrathin films. However, the comparison to full micromagnetic simulations (2D mesh, modified OOMMF code) that include the demagnetizing field exactly, is also shown in Fig. 2. Small differences are seen, especially for small in-plane fields because of the magnetostatic charge that appears at the sample edge reduces the edge magnetization tilt [3].

### **III. MICROMAGNETIC SIMULATIONS**

We consider a magnetic ultrathin film grown on a substrate with a capping layer in a different material so that the inversion symmetry is broken along the vertical axis (z). The magnetization is oriented out-of-plane with a strong perpendicular anisotropy. In addition to the standard micromagnetic energy density which includes the exchange, the magnetocrystalline anisotropy, the Zeeman and the demagnetizing energy, we add the Dzyaloshinskii-Moriya contribution (DM) that reads in a continuous form [6]:

$$E_{DM} = D[m_z \frac{\partial m_z}{\partial x} - m_x \frac{\partial m_z}{\partial x} + id.(x \to y)]$$
(7)

Micromagnetic simulations are based on the Landau-Lifshitz-Gilbert equation integration:

$$\frac{\partial \mathbf{m}}{\partial t} = -\gamma_0 [\mathbf{m} \times \mathbf{H}_{eff}] + \alpha (\mathbf{m} \times \frac{\partial \mathbf{m}}{\partial \mathbf{t}})$$
(8)

3D micromagnetic simulations are performed using the homemade micromagnetic solver Micro3D [7]. The following material parameters have been used: exchange stiffness  $A = 1 \times 10^{-11}$  J/m, saturation magnetization  $M_s = 1.09 \times 10^6$  A/m, uniaxial magnetocrystalline anisotropy constant  $K = 1.25 \times 10^6$  J/m<sup>3</sup> (along the out-of-plane direction z), Dzyaloshinskii-Moriya constant D = 2 mJ/m<sup>2</sup>, Gilbert damping parameter  $\alpha = 0.5$ . They differ slightly from those considered in the main text (mainly by the lower A and higher anisotropy  $H_{K0} =$ 0.92 T), but correspond to a previous work [8].

The thermal fluctuations were neglected and the size of the sample was 100 nm × 1024 nm × 0.6 nm. Starting from positive out-of-plane remanent state, two types of situation were analyzed: *i*) simultaneous application of an in plane field  $H_x$  (parallel to the short edge of the sample) and an easy axis  $H_z$  field (perpendicular to the film plane). The Dzyaloshinskii-Moriya constant was first set to D = 0 J/m<sup>2</sup> and after to D = 2 mJ/m<sup>2</sup>; ii) application of one single in-plane  $H_x$  field with D = 2 mJ/m<sup>2</sup>.

Figure 3(a-b) summarizes the results of the simulations carried out applying  $\mu_0 H_x =$ +0.1 T and  $\mu_0 H_z = -0.7$  T for D = 0 J/m<sup>2</sup>. Starting from out of plane remanent state at t = 0 ps, the magnetization reverses by nucleation of a domain in the center of the sample, where the demagnetizing field is the largest. The magnetization reversal follows a symmetric mechanism. Figure 3(a) shows the time evolution of the in-plane  $m_x$  component of the magnetization across the short edge of the sample. Snapshots at different moments



FIG. 3. Micromagnetic simulations showing the modification of the micromagnetic structure of the sample under the effect of: (a)  $\mu_0 H_z = -0.7$  T and  $\mu_0 H_x = +0.1$  T for D = 0 J/m<sup>2</sup> - the reversal occurs starting from the center of the sample; (b)  $\mu_0 H_z = -0.5$  T and  $\mu_0 H_x = +0.1$  T for D = 2 mJ/m<sup>2</sup> - the reversal starts from the left edge of the sample.

of time are shown on Fig. 3(b): the color is related to the out-of-plane magnetization and the arrows give the in-plane magnetization vector projection. The magnetization reversal starts in the middle of the sample and propagates to the edges.

Figure 3(c-d) shows that the magnetization reversal changes drastically in the presence of DMI. Before the application of the field, the magnetization at the sides of the sample is tilted towards the center, in opposite directions on either side of the sample. The effect of the x-field is to tilt the magnetization in the direction of the applied field; with a positive  $H_x$ , the  $m_x$  component of the magnetization will have a maximum value on the left side of the sample. Magnetization reversal is then triggered by nucleation of reversed domains at this edge, where the  $m_x$  component of magnetization is initially parallel to the  $H_x$  field.

Figure 4 (a) shows the effect of an in-plane field  $\mu_0 H_x = +0.4$  T applied at time t = 0 ps, for the case where  $= 2 \text{ mJ/m}^2$ . For this value of the x-field, the magnetization simply tilts



FIG. 4. Micromagnetic simulations showing the modification of the micromagnetic structure of the sample under the effect of an in-plane  $H_x$  field applied at time t = 0 ps for  $D = 2 \text{ mJ/m}^2$ . (a) for  $\mu_0 H_x = +0.4 \text{ T}$  the magnetization tilts in the direction of the applied field; (b) for  $\mu_0 H_x = +0.5 \text{ T}$  a domain wall is formed at the left edge of the sample and stabilizes away from the edge due to magnetostatic effects; (c) snapshots of the magnetization illustrating the evolution towards equilibrium of the domain wall formed under  $\mu_0 H_x = +0.5 \text{ T}$ .

in the direction of the applied field. The situation is very different for a larger  $H_x$  field of +0.5 T (Fig. 4(b)). In this case, a domain wall is created starting from the left edge of the sample, and its position stabilizes around the center of the sample due to demagnetizing effects. The creation of this domain wall in the presence of an in-plane field agrees with the results of the model developed in the main text of this Letter, which shows that in the presence of a large DMI, the domain wall energy becomes negative beyond a threshold in-plane field, therefore favoring its formation. Figure 4(c) gives a 2D representation of the initial

tilt of the magnetization at one edge of the sample, the formation of the domain wall and its propagation away from the edge.

## IV. ESTIMATION OF THE MAGNETIC PARAMETERS OF THE SAMPLE

The various micromagnetic parameters of the sample are not exactly known: some of them are directly obtained from characterizations, but some others are more difficult to measure, so that one has to see whether or not, with reasonable values assumed for them, the experimental results can be reproduced. In the case at hand, the relevant parameters are: spontaneous magnetization  $M_s$ , exchange constant A, effective anisotropy  $K_0$  (the sum of the volumic, interfacial and shape anisotropies), thickness averaged interfacial DMI D, and p the number of thermal energy quanta  $k_BT$  involved in the Arrhenius process, given by  $\tau = \tau_0 e^p$  with  $\tau_0$  the attempt time and  $\tau$  the duration of field application for the domain nucleation. We assume for the calculations that  $\tau_0 = 1$  ns. The value 0.1 ns is often quoted but, as in the Brown theory the attempt time is linked to the damping constant  $\alpha$ , and because from the DW dynamics experiments [9] a value of  $\alpha$  about 10 times larger than for bulk cobalt was deduced, we took this larger value. Note also that taking  $\tau_0 = 0.1$  ns increases p by 2.3 only, for a typical value of 18 that corresponds to  $\tau = 66$  ms. Finally, the DW energy reduction at the defect in the center of the film is given by the factor  $\epsilon$ .

For  $M_s$ , we have taken the value 1.1 MA/m. The bulk Co value is 1.4 MA/m. SQUID measurements, with large error bars as the sample thickness is only 3 atomic layers (t = 0.6 nm), are consistent with a lower value of 1.1 MA/m, in agreement with literature.

The effective anisotropy field  $H_{K0} = 2K_0/(\mu_0 M_s)$  was directly measured by extraordinary Hall effect under in-plane field. We found  $\mu_0 H_{K0} = 0.7$  T.

The exchange constant was assumed to amount to 16 pJ/m. For bulk Co, the literature contains values ranging from 10 to 30 pJ/m. The value used here corresponds to an estimate based on the Curie temperature of the ultrathin films [9]. We will discuss below what changes if a lower value of 9 pJ/m is taken.

With these parameters, the primitive DW energy density (without DMI, and under no applied in-plane field) is  $\sigma_{00} = 4 \ (AK_0)^{1/2} = 9.92 \ \text{mJ/m}^2$ , thus the critical DMI for destabilization of the uniform state is  $D_{c0} = \sigma_{00}/\pi = 3.16 \ \text{mJ/m}^2$ . The DW width parameter is  $\Delta = (A/K_0)^{1/2} = 6.4 \ \text{nm}$ . From these values, we can evaluate the scaling z field of the



FIG. 5. Nucleation fields in the film and at the edge, normalized to  $H_{z,00}$  (see Eq. (9)), as a function of x field normalized to the effective anisotropy field  $H_{K0}$ , for different values of the normalized DMI, as computed by the semi-analytical model. The horizontal scale is always the same, but note the change of the vertical scale as DMI varies.

problem, given by

$$H_{z00} = \frac{\pi \sigma_{00}^2 t}{2\mu_0 M_s p k_B T} = \frac{4\pi t}{p k_B T} A H_{K0}$$
(9)

and find  $\mu_0 H_{z00} = 1172$  mT.

In order to estimate the value of the DMI parameter D, we have systematically varied the  $D/D_{c0}$  ratio and computed the nucleation fields in the film and at the edge, keeping  $\epsilon = 1$  for the moment so that the film nucleation field is very large. Results are shown in Fig. 5. From both the value of the in-plane field where the edge nucleation field becomes zero (about 250 mT i.e.  $H/H_{K0} = 0.36$ ), and the shape of the curve showing the nucleation field in the film, we find that  $D/D_{c0} = 0.7$  fits the best.

Finally, in order to reproduce the experimental value of the nucleation field in the film,

we need  $\epsilon = 0.42$ . From Eq. (9), we see that the value of the nucleation field in the film will be given by the product  $A\epsilon^2$ . Thus, with a lower value A = 9 pJ/m, a value  $\epsilon = 0.57$ would be required. This would however shift the value of the in-plane field where the two nucleation fields are equal, from 105 to 56 mT, whereas experimentally it is about 125 mT.

A word of caution is in order here about the physical meaning of this defect parameter  $\epsilon$ . It was originally introduced in order to model the effect of the defect on the nucleation field [10], in the case of a sole easy axis field, through a reduction of the DW energy. Here, when applying an in-plane field, the DW energy becomes orientation dependent so that the validity of an isotropic reduction of its value by the factor  $\epsilon$  should be investigated in more detail. We leave this as a subject of future work.

- E. C. Stoner and E. P. Wohlfarth, Phil. Trans. Roy. Soc. London A240, 599 (1948), reprinted in IEEE Trans. Magn., vol. 27, pp. 3475-3518 (1991).
- [2] A. Thiaville, J. Magn. Magn. Mater. **182**, 5 (1998).
- [3] S. Rohart and A. Thiaville, Phys. Rev. B 88, 184422 (2013).
- [4] M. Jamet, W. Wernsdorfer, C. Thirion, D. Mailly, V. Dupuis, P. Mélinon, and A. Pérez, Phys. Rev. Lett. 86, 4676 (2001).
- [5] C. Vouille, A. Thiaville, and J. Miltat, J. Magn. Magn. Mater. 272, E1237 (2004).
- [6] A. Thiaville, S. Rohart, E. Jué, V. Cros, and A. Fert, EPL 100, 57002 (2012).
- [7] L. D. Buda, I. L. Prejbeanu, U. Ebels, and K. Ounadjela, Comput. Mater. Sci. 24, 181 (2002).
- [8] O. Boulle, S. Rohart, L. D. Buda-Prejbeanu, E. Jué, I. M. Miron, S. Pizzini, J. Vogel,
   G. Gaudin, and A. Thiaville, Phys. Rev. Lett. 111, 217203 (2013).
- [9] P. J. Metaxas, J. P. Jamet, A. Mougin, M. Cormier, J. Ferré, V. Baltz, B. Rodmacq, B. Dieny, and R. L. Stamps, Phys. Rev. Lett. 99, 217208 (2007).
- [10] J. Moritz, B. Dieny, J. Nozières, Y. Pennec, J. Camarero, and S. Pizzini, Phys. Rev. B 71, 100402 (2005).