Room temperature chiral magnetic skyrmion in ultrathin magnetic nanostructures

Olivier Boule,1,2,3 Jan Vogel,4,5 Hongxin Yang,1,2,3 Stefania Pizzini,4,5 Dayane de Souza Chaves,4,5 Andrea Locatelli,6 Tevfik Onur Mentes,6 Alessandro Sala,6 Liliana D. Buda-Prebeann,1,2,3 Olivier Klein,1,2,3 Mohamed Belmeguenai,7 Yves Roussigné,7 Andrey Stashkevich,7 Salim Mourad Chérief,7 Lucia Aballe,8 Michael Foerster,8 Maïrbek Chshiev,1,2,3 Stéphane Auffret,1,2,3 Ioan Mihaï Miron,1,2,3 and Gilles Gaudin1,2,3

1Univ. Grenoble Alpes, SPINTEC, F-38000 Grenoble, France
2CNRS, SPINTEC, F-38000 Grenoble, France
3CEA, INAC-SPINTEC, F-38000 Grenoble, France
4CNRS, Institut Néel, 25 avenue des Martyrs, B.P. 166, 38042 Grenoble Cedex 9, France
5Univ. Grenoble Alpes, Institut Néel, 25 avenue des Martyrs, B.P. 166, 38042 Grenoble Cedex 9, France
6Elettra-Sincrotrone, S.C.p.A, S.S 14 - km 163.5 in AREA Science Park 34149 Basovizza, Trieste, Italy
7LSPM (CNRS-UPR 3407), Université Paris 13, Sorbonne Paris Cité, 99 avenue Jean-Baptiste Clément, 93430 Villetaneuse, France
8ALBA Synchrotron Light Facility, Carretera BP 1413, Km. 3.3, Cerdanyola del Valls, Barcelona 08290, Spain

Magnetic skyrmions are chiral spin structures with a swirling configuration. Their topological properties, nanometer size and the fact that they can be moved by small current densities have opened a new paradigm for the manipulation of magnetisation at the nanoscale\cite{1,7}. A key feature of such magnetic nano-objects is their chiral and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$.

The recent discovery of nanometer size whirling magnetic structures named magnetic skyrmions has opened a new path to manipulate magnetisation at the nanoscale\cite{1,7}. A key feature of such magnetic nano-objects is their chiral and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologically non-trivial spin structure, i.e their magnetisation texture cannot be continuously transformed into the uniform magnetic state without causing a singularity $S$. For a surface and topologi...
class of materials combines several features which makes their use appealing for the study of skyrmion structures and their applications: i/ The magnetic parameters controlling the skyrmion stability and size, i.e. the anisotropy, the DMI and the exchange [30, 34], can be easily tuned by playing with the nature and thickness of the materials composing the multilayers. ii/ They are characterized by a large DMI [35–38, 41–44] which leads to chiral Néel domain walls (DWs) [45, 46]. iii/ Large current induced spin orbit torques are present [47, 48] which results in fast current induced DW motion [49, 50]. iv/ The deposition by sputtering is fast and spatially homogeneous and is compatible with standard spintronics devices such as magnetic tunnel junctions, which makes the industrial integration straightforward. Whereas several recent experimental works have studied magnetic bubbles in such materials [14, 18, 51, 52], and demonstrated their current induced motion [14, 52], the direct evidence of their chiral internal structure is still lacking. Here we report on the observation of stable chiral skyrmions in sputtered ultrathin Pt/Co(1 nm)/MgO nanostructures at room temperature and zero applied magnetic field. We used photoemission electron microscopy combined with X-ray magnetic circular dichroism (XMCD-PEEM) to demonstrate their chiral Néel internal structure. The XMCD-PEEM combines several advantages for the observation of magnetic nanostructures, such as skyrmions: firstly, a high lateral spatial resolution (down to 25 nm); secondly, the magnetic contrast is proportional to the projection of the local magnetisation along the X-ray beam direction. In our experiment, the X-ray beam impinges at a grazing angle of 16 degrees on the sample surface plane so that the contrast is approximately three times larger for the in-plane component of the magnetisation than for the out-of-plane one. This important feature allows the direct imaging of the internal in-plane spin structure of DWs or skyrmions.

FIG. 1: Imaging of the chiral Néel structure of domain walls using XMCD-PEEM magnetic microscopy. (a) Magnetic image of a multidomain state in the continuous Pt/Co/MgO film. For DWs lying perpendicular to the X-ray beam direction, thin white and black lines can be seen, corresponding to the magnetisation being aligned antiparallel and parallel to the photon beam respectively. This demonstrates their chiral Néel structure. (b) Linescan of the magnetic contrast corresponding to the dotted white line in (a). To reduce the noise, the contrast has been averaged perpendicularly to the linescan over 60 nm. The red line is a fit assuming a chiral Néel DW structure convoluted by a Gaussian function to take into account the finite spatial resolution [53].

Observation of chiral Néel domain walls using XMCD-PEEM

All images shown here were acquired at room temperature and, unless otherwise stated, no external magnetic field was applied during the experiments. The PEEM observations were done in a virgin demagnetized state obtained after the sample nanofabrication and annealing. Complementary magnetisation measurements on unpatterned thin films show that the sample is magnetised perpendicularly to the film in the magnetic domains, which is due to a large interfacial uniaxial anisotropy. Figure 1(a) shows an XMCD-PEEM magnetic image of a multidomain state in the continuous film. Dark and bright grey regions correspond respectively to the magnetisation being aligned antiparallel and parallel to the X-ray beam direction. This demonstrates their chiral Néel structure. A peak in
the contrast is observed at the up/down DW position while a dip is observed at the down/up DW position. Thus, the magnetisation in the up/down DW is aligned antiparallel to the in-plane direction of the X-ray beam whereas the magnetisation in the down/up DW is aligned parallel. We conclude that the DW magnetisation is perpendicular to the DW surface with an opposite magnetisation direction for the two DWs. This demonstrates that DWs in this material are chiral Néel DWs with a left-handed chirality. Note that for Bloch DWs, the magnetisation would be always perpendicular to the beam direction so that no peak or dip in the magnetic contrast should be observed. The linescan is well fitted assuming a chiral Néel DW structure, the finite resolution of the instrument being modeled by a Gaussian convolution (red curve, Fig. 1(b)). The fit leads to a DW width of 29.5 ± 4 nm (π√A/K_{eff}) [52].

FIG. 2: Brillouin Light Scattering spinwave spectroscopy. (a) Principle of the measurement. At a given time \( t \), when moving along \( x \), the magnetisation rotates clockwise (counterclockwise) around the \( D \) vector for spin waves propagating along \( x \) (-\( x \)), which leads to different DM energy. (b) BLS spectra for an in-plane magnetic field \( H=0.7 \) T and \( k_x=4.1\mu m^{-1} \). The red squares are experimental data whereas the red lines are Lorentzian fits. The blue line is a Lorentzian fit of the experimental data inverted with respect to \( f = 0 \). (c) \( \Delta f = f_S - f_{AS} \) as a function \( k_x \) for \( H=0.7 \) T, where \( f_S \) and \( f_{AS} \) are respectively the Stokes and Anti-Stokes resonance frequency. Inset: \( f_S \) and \( f_{AS} \) as a function \( k_x \).

Spin wave spectroscopy experiments and \textit{ab-initio} calculations

The driving force of the DW and skyrmion chiral structure is the DMI. To further quantify its amplitude in our films, we measured the frequency shift of oppositely propagating spin waves using spin wave spectroscopy experiments [38–43]. The idea of the measurement is the following: When the magnetisation is pulled in the plane by an external magnetic field \( H_y \), the \( D \) vector is oriented along \( y \) for spin waves propagating along the \( x \) axis (see Fig. 2(a)). Thus, at a given time \( t \), when moving along the \( x \) axis, the magnetisation rotates anticlockwise around the \( D \) vector for spin waves with \( k_x < 0 \) and clockwise for \( k_x > 0 \). This leads to an energy shift for spin waves with opposite \( k_x \) vector due to the DMI and the corresponding frequency shift writes \( \Delta f(k_x) = f(k_x) - f(-k_x) = 2\gamma k_x D/(\pi M_s) \). To measure \( \Delta f \), we have carried out spin wave spectroscopy using the Brillouin Light Scattering technique in a backscattering geometry [43]. A spin wave spectrum is shown on Fig. 2(b) (red dots) for an in-plane magnetic field of 0.7 T and \( k_x=4.1\mu m^{-1} \). The Stokes (S) and Anti-Stokes (AS) peaks are observed, corresponding to ±\( k_x \). The blue line is a Lorentzian fit of the experimental data inverted with respect to \( f = 0 \), which shows that the Stokes peak has a frequency different to the Anti-Stokes peak, as is expected in the presence of DMI. The shift in frequency \( \Delta f = f_S - f_{AS} \) scales linearly with \( k_x \) (Fig. 2(c)), which allows to extract a DM parameter \( D = 2.05\pm0.3 \) mJ/m². Note that the sign of \( \Delta f \) is consistent with the sign of \( D \) and the left handedness of the Néel DW we observe experimentally. As \( D \) is expected to be inversely proportional to the film thickness \( t \) [43], one can define a related interfacial DM parameter \( D_s \) such that \( D = D_s/t \) and we find a value \( D_s = 2.17\pm0.14 \) pJ/m. To our knowledge, this value is the highest reported so far for a sputtered magnetic ultrathin film. To better understand this large value, we carried out \textit{ab-initio} calculations of the DMI in Pt/Co(n ML)/vacuum and Pt/Co(n ML)/MgO multilayers [44, 53]. For \( n=5 \) ML of Co, equivalent to a total Co thickness of 1 nm, the \textit{ab-initio} calculations predict \( D = 2.3 \) mJ/m² in relatively good agreement with experiments. Note that a lower value \( D = 1.5 \) mJ/m² is predicted for a Pt/Co[8N]/vacuum structure,
which underlines the role played by the Co/oxide interface and in particular the large electric field due to the charge transfer between O and Co atoms.

Room-temperature skyrmion in a magnetic nanostructure

In continuous thin films, isolated magnetic bubbles [54] or chiral skyrmions [6] have been observed so far in the presence of a perpendicular magnetic field, which breaks the stripe domains or helical structure driven by the magnetostatic or DM energy. However, it is known that single magnetic bubbles can be stabilized without external magnetic fields using geometrical confinement in patterned nanostructures [55, 56]. Here we patterned different structures with various sizes and shapes (circular, square) in our Pt/Co/MgO thin films. Fig. 3(a) shows a circular magnetic domain stabilized in the middle of a 420 nm wide square dot, imaged at room temperature and no applied magnetic field. As observed in the multidomain structure, a sharp black/white contrast is observed at the DW position at the bottom/top of the central domain. This leads to a dip/peak in the dichroic contrast when doing a linescan along the domain diameter in the beam direction (see Fig. 3(b)). This indicates that the in-plane DW magnetisation is aligned parallel/antiparallel to the X-ray beam at the bottom/top of the reversed domain, i.e. the DW surrounding the circular domain is a chiral Néel DW. This chiral border leads to a skyrmion number \( |S| = 1 \) for this structure. This demonstrates that the observed circular domain is a Néel like magnetic skyrmion. To extract the size of the skyrmion from the image, we assume that the magnetisation profile can be described by a 360° DW profile [29, 57, 58]:

\[
m(r) = \sin(\theta(r))u_r + \cos(\theta(r))u_z,
\]

with

\[
\theta(r) = \theta_{DW}(r - d/2) + \theta_{DW}(r + d/2),
\]

where \( \theta_{DW} = 2 \arctan(\exp(r/\Delta)) \); \( d \) is the skyrmion diameter and \( \Delta \) is the DW width. The unit vector \( u_r \) is the polar unit vector. The blue curve (Fig. 3(b)) shows a fit of the experimental linescan assuming a Gaussian convoluted 360° DW and a good agreement is obtained with experimental data. From the fit, a skyrmion diameter \( d = 130 \pm 2.5 \) nm is extracted.

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**FIG. 3**: Magnetic skyrmion observed at room temperature and zero applied external magnetic field (a) XMCD-PEEM image of a 420 nm square dot (indicated by the dotted line) and (b) linescan along the dotted black line (black line). The linescan has been averaged perpendicularly to the linescan over 30 nm. The blue line is a fit to the data using a Gaussian convoluted 360° DW profile [53]. The orange line is the contrast predicted by the micromagnetic simulations.

To better understand these experimental results, we carried out micromagnetic simulations using experimental values for the magnetocrystalline anisotropy constant \( K \), the magnetic moment per surface area and the DMI interaction amplitude \( D \). The exchange constant was used as a free parameter and the best agreement between experiments and micromagnetic simulations is obtained for \( A = 27.5 \) pJ/m, a value in line with previous measurements of \( A \) in ultrathin magnetic multilayers [59]. Using these parameters, micromagnetic simulations predict a stable left-handed chiral skyrmion structure at zero external magnetic field (see Figure 4(a)) with a diameter of 128 nm and a DW width of 37 nm. From this magnetisation pattern, an experimental magnetic image can be simulated and a good agreement is obtained with the experimental results (see Fig. 4(b) and Fig. 3(b), orange curve for the simulated linescan). We show on Fig. 4(d) the same reconstructed experimental image assuming a Bloch DW structure. The image is rotated 90° with respect to the chiral Néel bubble structure and it is in clear disagreement with our experimental data. Finally, the simulations allow us to reconstruct the structure of the observed skyrmion. We show in Fig. 4(c) a linescan of the...
in-plane ($m_x$) and out-of-plane ($m_z$) component of the magnetisation along the skyrmion diameter, as predicted by the micromagnetic simulation. The skyrmion diameter ($\sim 130$ nm) being large compared to the DW width (37 nm), the magnetisation profile is close to two independent chiral Néel DWs, as can be seen on the $m_z$ profile.

FIG. 4: Micromagnetic simulations. (a) Magnetisation distribution and (b) simulated magnetic contrast of the magnetic bubble in a 420 nm dot. (c) Magnetisation component $m_x$ and $m_z$ along the dotted white line in (a). (d) XMCD-PEEM contrast assuming a Bloch bubble structure. For the simulated magnetic contrast image, a spatial lateral resolution of 28 nm is assumed as obtained from the fit of the topological image of Fig. 3.

Numerical calculations

The observation of stable skyrmions at zero external magnetic field raises the question of the physical mechanisms that govern the skyrmion stability and size in our experiments. To address this point, we consider a simple model where the magnetisation in the dot $\theta(r)$ is described by a circular 360° Néel DW profile. The free energy $E$ in the circular dot-shaped nanostructure can be written as the sum of two terms [1, 30, 34, 60]: (1) the skyrmion energy $E_\sigma[\theta(r)]$ due to the exchange, anisotropy and internal DW stray field energies and (2) the energy due to the magnetostatic interactions between the domains $E_{mag}$. Assuming a radial symmetry, $E_\sigma[\theta(r)]$ can be written as [1, 30, 34, 60]:

$$E_\sigma[\theta(r)] = 2\pi t \int_0^R \left\{ A \left[ \left( \frac{d\theta}{dr} \right)^2 + \frac{\sin^2 \theta}{r^2} \right] - D \left[ \frac{d\theta}{dr} + \frac{\cos \theta \sin \theta}{r} \right] + (K_{eff} + E_{DW}) \sin^2 \theta \right\} r dr$$

(1)

where $K_{eff} = K - \mu_0 M_s^2 (1 - N_{DW})/2$ is the effective anisotropy [52, 61] ($K$ is the magneto-crystalline anisotropy constant), $t$ is the film thickness, $R$ the dot radius. The demagnetizing energy due to the magnetic charges within the DWs $E_{DW}$ is described by a constant demagnetizing factor $N_{DW}$ such that $E_{DW} = N_{DW} \mu_0 M_s^2/2$. The energies $E_\sigma$ and $E_{mag}$ [62] can be evaluated as a function of the skyrmion diameter $d$ assuming a 360° DW profile and a 420 nm diameter circular dot (the magnetic parameters correspond to the experiment of Fig. 3, see methods). More physical insight is obtained from the effective forces $F_\sigma(d) = -\frac{\partial E_\sigma}{\partial d}$ and $F_{mag}(d) = -\frac{\partial E_{mag}}{\partial d}$ which are plotted in Fig. 5. A first interesting feature is that $F_\sigma(d)$ cancels out for $d \sim 20$ nm, which thus would be an equilibrium size for the skyrmion in the absence of the domain magnetostatic energy. This equilibrium is the result of a balance between the DW energy cost which is proportional to $d$ and tends to decrease the skyrmion diameter and the curvature energy
cost due the exchange energy which scales as $1/d$ [34]. However, the magnetostatic force $F_{\text{mag}}$ is large enough at low diameter to destabilize this balance and the final equilibrium position is obtained for a larger value of $d \sim 90$ nm, where the two forces are equal. This underlines that the magnetostatic energy plays an important role in the stability and size of the skyrmion at zero external magnetic field. We also carried out micromagnetic simulations for square nanostructures with larger lateral dimensions. We observed that for sides larger than 1.2 $\mu$m, the skyrmion structure is not stable and a stripe domains structure appears [53]. This may explain why we did not observe any skyrmions but stripe domains for larger structures with 1 $\mu$m sides [53]. The confinement is thus an additional important feature for the skyrmion stability.

![Graph](image)

**FIG. 5:** **Forces acting on the DW as a function of the skyrmion diameter.** $F_\sigma$ is the force due to the exchange and effective anisotropy (black curve), $F_{\text{mag}}$ is the force due to the magnetostatic interaction between the domains (red curve).

It may seem surprising that the domain magnetostatic energy plays such an important role in the skyrmion stability given the very small thickness of the layer. Indeed, magnetostatic effects are expected to favor multidomain states only when the film thickness $t$ is of the order or larger than the characteristic length $l_w = \sigma/(\mu_0 M_s^2)$, where $\sigma$ is the DW surface energy. This criterion expresses the competition between the cost in the DW energy and the gain in the magnetostatic energy when creating a domain. The energy $\sigma$ scales as $\sigma \sim 4\sqrt{AK_{\text{eff}}} - \pi D$ so that it is decreased in the presence of DMI. Thus, the criterion $l_w \sim t$ can be fulfilled even for very low thickness. In our experiments, we find that $l_w$ is indeed of the order of the film thickness, $l_w \sim 1$ nm. This leads to the counterintuitive conclusion that the higher the DMI, the larger the role played by the magnetostatic interaction in the skyrmion stability. Note that this analysis holds for $\sigma > 0$, i.e $D < D_c$ where $D_c = 4\sqrt{AK_{\text{eff}}}/\pi$, which is the case in our sample where $D/D_c = 0.8$.

To conclude, we have observed stable magnetic skyrmions at room temperature in ultrathin Pt/Co/MgO films in the absence of applied external magnetic field. Using the high resolution XMCD-PEEM magnetic microscopy technique, we were able to demonstrate its internal left-handed chiral Néel structure, which can be explained by the large Dzyaloshinskii-Moriya interaction measured in this material. Micromagnetic simulations are in agreement with our experimental results and we show that the equilibrium skyrmion structure is the result of a balance between the DW energy modulated by the DMI and the magnetostatic interaction in the patterned structure. This balance is very sensitive to the different magnetic parameters that are $D$, $K$, $A$ and the total magnetic moment. The lateral confinement also plays an important role since it leads to a modulation of the magnetostatic energy. Hence, the size and stability of magnetic skyrmions can be finely tuned by playing with these different parameters. This will be a key for the design of future devices based on the manipulation of magnetic skyrmions.

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**Methods**

Sample preparation and magnetic microscopy experiment The Ta(3)/Pt(3)/Co(0.5-1)/MgOₓ/Ta(1) (thickness in nm) film was deposited by magnetron sputtering on a 100 nm high resistivity Si wafer, then annealed for 1.5 h at 250°C under vacuum and under an in-plane magnetic field of μ₀H = 240 mT. The Co layer was deposited as a wedge using a rotating cover during the deposition. The nominal thickness at the position of observation is t = 0.98 nm. The samples were patterned in different shapes (circle, square) and sizes using standard nanofabrication techniques. The XMCD-PEEM magnetic microscopy experiments were carried out with the SPELEEM III microscope (Elmitec GmbH) at the Nanospectroscopy beamline 64 at the Elettra synchrotron in Basovizza, Trieste, Italy and at the CIRCE beamline with the collaboration of ALBA staff 65. To fit the linescan of Fig. 1(b), the standard deviation of the spatial Gaussian convolution σ is used as a free parameter 53 leading to 2σ = 40 nm . In Fig 1(b), σ is deduced from a fit with an error function of a linescan of the topological image of the dot , which leads to 2σ = 28 nm.
Brillouin light scattering experiment The Brillouin light scattering experiments setup and conditions are the same as described in Ref. [43]. A backscattering geometry has been used. The investigated spin wave vector lies in the plane of incidence and its length is \( k_x = 4\pi \sin(\theta_{\text{inc}})/\lambda \) (with \( \theta_{\text{inc}} \) the angle of incidence and \( \lambda = 532 \) nm the wavelength of the illuminating laser). The external magnetic field was applied perpendicular to the incidence plane, which allows spin waves propagating along the in-plane direction perpendicular to the applied field to be probed (Damon-Eshbach geometry).

Micromagnetic simulations The micromagnetic simulations were carried out using different micromagnetic codes: a homemade code [66], the Mumax3 code [67], the OOMMF code [68]. The following parameters were used [53]: \( K = 1.45 \times 10^6 \) J/m\(^3\), \( M_s = 1.4 \times 10^6 \) A/m, \( A = 27.5 \) pJ/m, \( D = 2.05 \) mJ/m\(^2\) and a film thickness \( t = 1.06 \) nm. The lateral size of the elementary cells was typically between 1 and 3 nm. The results presented in this paper were obtained for a lateral cell size of 1 nm. In Fig. 4(b), a spatial Gaussian convolution with standard deviation \( \sigma = 14 \) nm was used to simulate the finite lateral spatial resolution of the microscope.

Numerical calculations of the skyrmion energy \( E_\sigma(d) \) was calculated numerically by minimizing \( E_\sigma \) with respect to \( \Delta \) at fixed \( d \). \( E_{\text{mag}}(d) \) was then evaluated from the total magnetostatic energy \( E_{\text{mag}0} \) of a Bloch DW in a dot using Ref. [62].

\[
E_{\text{mag}}(d) = E_{\text{mag}0} - t(1 - N_{\text{DW}}) \frac{\mu_0 M_s^2}{2} \int_0^R \cos^2 \theta \ 2\pi r dr \ \\
E_{\text{mag}0} = \frac{4\pi t}{R} \int_0^\infty [1 - \exp(-\beta x)] I^2(x) dx \ \\
I(x) = \int_0^1 dr' r' J_0(xr') \cos \theta(r')
\]

where \( r' = r/R \) and \( J_0 \) is the Bessel function of order zero. \( R \) is the dot radius. \( N_{\text{DW}} = 0.0188 \) was deduced from the width of the DW predicted by the micromagnetic simulations.

Competing Interests The authors declare that they have no competing financial interests.

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Author Contributions O.B. conceived and designed the experiments. O.B., J.V., S.P., D.S.C., A.L, T.O.M., A.S., L.A., M.F. participated in the XMCD-PEEM experiments. O.B and J.V. analyzed the microscopy data. H.Y. and M.C. carried out the \textit{ab-initio} calculations. O.B., L.B-D. and O.K. carried out the micromagnetic simulations, O.B. carried out the numerical calculations, S.A. deposited the magnetic multilayers, M.B., Y.R., A.S. carried out the BLS experiments. O.B. wrote the manuscript. All authors discussed the results and commented on the manuscript.

Correspondence Correspondence and requests for materials should be addressed to O.B. (email: olivier.boulle@cea.fr).