

Supplementary Information

Spatial magnetic imaging of non-axially symmetric vortex domains in cylindrical nanowire by Transmission X-ray Microscopy

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1. Micromagnetic modelling of monovortex and trivortex states.

We have modelled magnetic states of a bisegmented CoNi-Ni (Co₈₅Ni₁₅) nanowires with periodic boundary conditions for the magnetostatic fields in the axial direction to mimic multi-segmented nature and long-range interactions in long nanowires. Micromagnetic parameters of CoNi and Ni are listed in Table S1. We have varied the direction of the easy axis of the magnetocrystalline anisotropy with respect to the nanowire axis between 60° and 88° and rotated it in 360° around the nanowire axis. Single vortex and trivortex magnetic states have been set as an initial condition in the CoNi segment, and a uniform magnetic domain in the Ni segment. Their energy was further minimized with the aim to achieve stable magnetisation states. We used mumax3 micromagnetic program [1] with finite-difference discretization of 1.25 nm.

Compound	$\mu_0 M_s$ (T)	A_{ex} (pJ/m)	Crystal Symmetry	K_1 (kJm ⁻³)	Magnetization Easy Axis (E.A.)
Ni (111) [2]	0.61	3.4	Cubic	-4.8	[111] crystal lattice direction parallel to the nanowire axis
Co ₈₅ Ni ₁₅ [3]	1.60	26.0	Uniaxial	350	at 60 - 88° with nanowire axis.

Table S1. Material parameters and crystal structures of Ni and CoNi used in the micromagnetic modelling. M_s is the saturation magnetization, A_{ex} is the exchange stiffness, K_1 is the magnetocrystalline anisotropy constant and E.A. is the easy axis of the anisotropy.

1.1 Dependence of the vortex core shape on anisotropy value and direction.

The variations of the uniaxial anisotropy constant K_1 in a monovortex state with an easy axis at $\theta=88^\circ$ with respect to the nanowire axial direction evince a gradual transition between an axial monodomain state ($K_1 = 0 \text{ J/m}^3$) and a vortex with a large ellipsoidal core size ($K_1 = 150 \text{ J/m}^3$). In brief, we have checked that a monovortex can be stabilized with at least a magnetocrystalline anisotropy of 50 J/m^3 with easy axis perpendicular to the nanowire axis. However, the trivortex state is stabilized for a magnetocrystalline anisotropy of at least 100 J/m^3 . For higher values of K_1 , the vortex

core reduces its size in the direction perpendicular to the easy axis (See Fig. S1). As a result, this increases the core eccentricity.

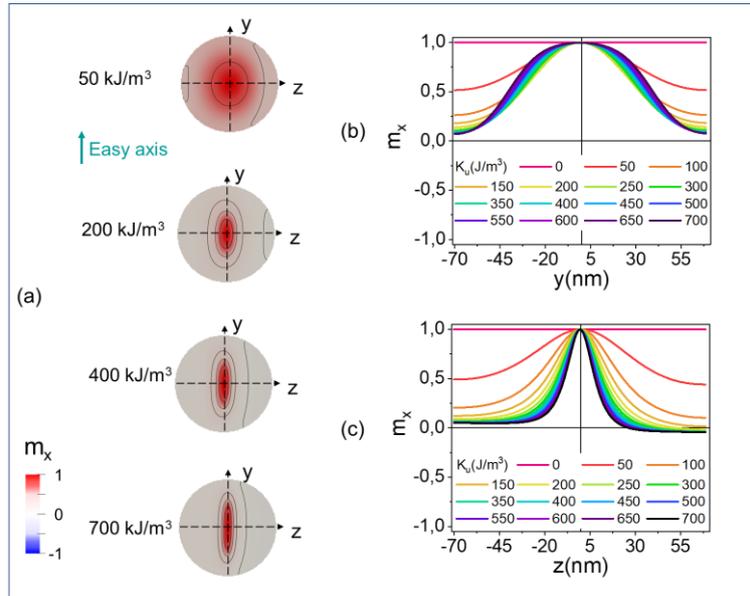


Figure S1. Shape and size of the vortex core structure in the CoNi-segment as a function of the magnetocrystalline anisotropy. (a) The x-component of the magnetization at the cross section of the CoNi segment for different anisotropy values. (b,c) Profiles of the vortex along the major and minor directions for different values of the anisotropy constant. The easy axis direction was set to $\theta=88^\circ$

In regard to the variations of the monovortex and trivortex state as a function of the magnetocrystalline anisotropy easy axis direction, we have checked that a monovortex state is stable for the easy axis forming at least 40° with respect to the axis of the nanowire, while at least 50° are required for a trivortex state.

1.2 Structure of the domain wall between two vortex domains with opposite chiralities.

Figures S2(a-b) show two representative cases of the domain wall formed between two vortex states with opposite chiralities with $\theta=88^\circ$, and $\theta=60^\circ$ anisotropy axis directions respectively. Specifically, the axial component of the easy axis of magnetocrystalline anisotropy competes with the effective anisotropy induced by curvature, resulting in a trajectory of the zig-zagging or sinusoidal lines in the vortex-type domains in the trivortex case. This sinusoidal trajectory is identical in all vortex domains and responds to the canting of the plane that contains the rotation in each domain towards the direction of the easy axis of magnetocrystalline anisotropy as indicated in Figure S3(a) for the 60° axis. A better visualization of streamlines for $\theta=88^\circ$ is Fig. S3(b)

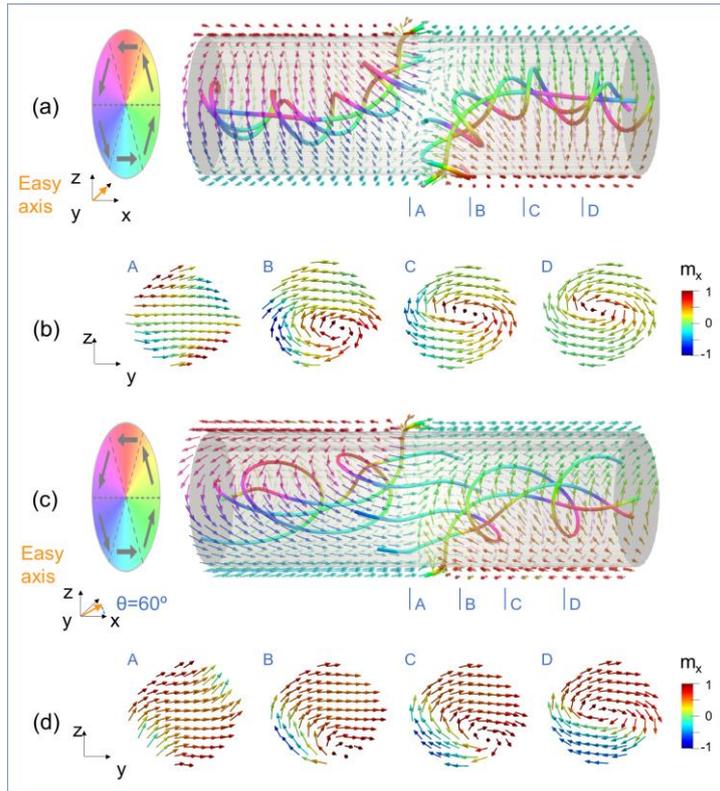


Figure S2. Domain wall formed at the encounter between two vortices with opposite chiralities for two directions of the easy axis: (a, c) Arrows show the direction of the magnetization on the surface of the nanowire (in grey colour). The selected streamlines (tubes) of the magnetization inside the volume follow the core and the curling direction of the vortices. Arrows and tubes are coloured by the yz-angle of the magnetization field. (b, d) Magnetization at cross sections at positions labelled by A-D. Arrows are coloured by the x-component of the magnetization.

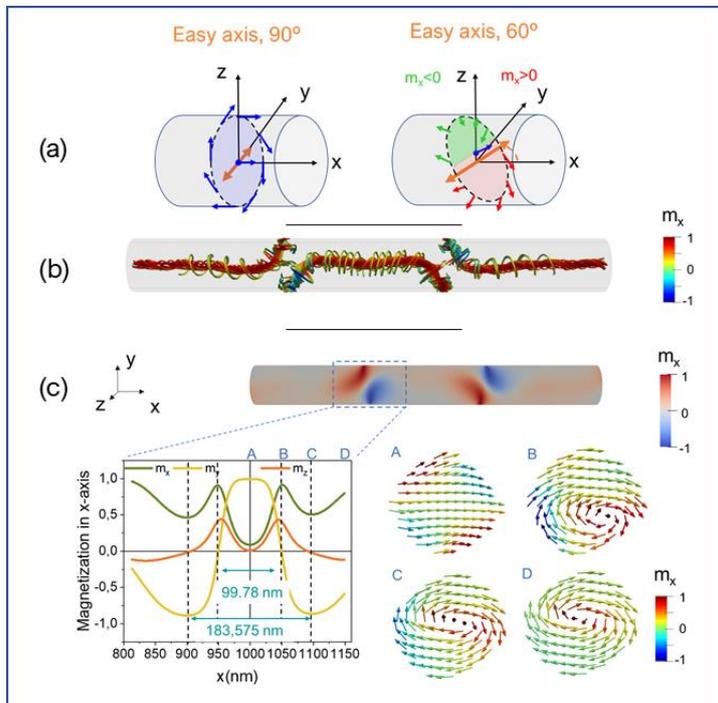


Figure S3. (a) Schematic representation of the modification of vortex structure with easy anisotropy axis direction. (b) Domain wall inner structure indicated with streamlines of magnetization calculated with points along the x-axis for $\theta=88^\circ$. These streamlines indicate that the direction followed by the cores of the vortices.

(c) On the top, the magnetization at the surface of the nanowire coloured by the x-component for a prototypical domain wall. At the bottom left, the three components of the magnetization in the section of axis enclosed by the dashed box. At the bottom right, the magnetization at cross sections at positions A-D coloured by the x-component.

In the case of the easy axis at $\theta=88^\circ$, for which the vortices are better defined and the cores of the vortex domains are coaxial with the axis of the wire, it is natural to measure the distances at which each vortex begins to move, that is, define a size of the prototypical wall structure that facilitates comparison with experimental measurements. Figure S3(c) represents the three components of magnetization on the axis of the wire (x-axis) and it is observed that the displacement of the vortices begins gradually at a distance of about 90 nm from the center of the wall, until the vortices touch the surface at about 50 nm.

The characterization of the magnetization of the domain wall on the surface is more complex. We observed four topologically non-trivial structures located in diametrically opposite positions two by two at the ends of a cross-arms as depicted in Figure S4: The first pair consists of a vortex with outward magnetization and inward magnetization for the positions in which the inner cores of the vortices reach the surface.

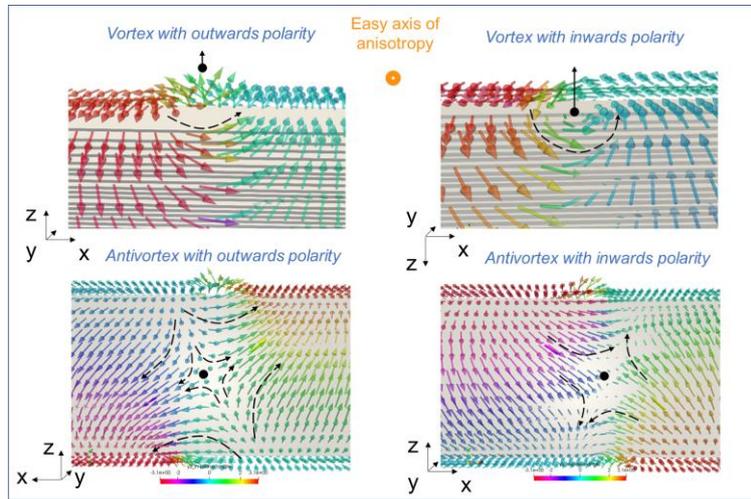


Figure S4. Examples of pairs of topological defects formed on the surface of the nanowire at the encounter of two vortex domains. $\theta=88^\circ$.

The direction of magnetization of the surface vortices (pointing outwards / inwards from the surface) and their direction of rotation are inherited from the internal vortices. Furthermore, here we observed that the vortices have moved in the direction normal to the axis of anisotropy. Besides, a pair of anti-vortices have been formed with inward and outward magnetizations at their center in the direction perpendicular to that of the displacement of the vortices on the surface of the wire.

2. Variation of the Transmission X-ray Microscopy (TXM) contrast with the orientation of the easy axis.

2.1 Dependence of the contrast on the nanowire orientation with an easy axis nearly perpendicular to the nanowire

Figure S5 presents a simulated TXM contrast calculated from the micromagnetic model in a Ni/CoNi nanowire with an anisotropy axis almost perpendicular to nanowire in CoNi. The rotation of the magnetocrystalline easy axis (or the nanowire around its axis) in Figure S5(a) modifies the TXM contrast so that the contrast gradient becomes sharper or less sharp, as displayed by the simulated contrasts in Figure S5(b) and the different curves in Fig S5(c).

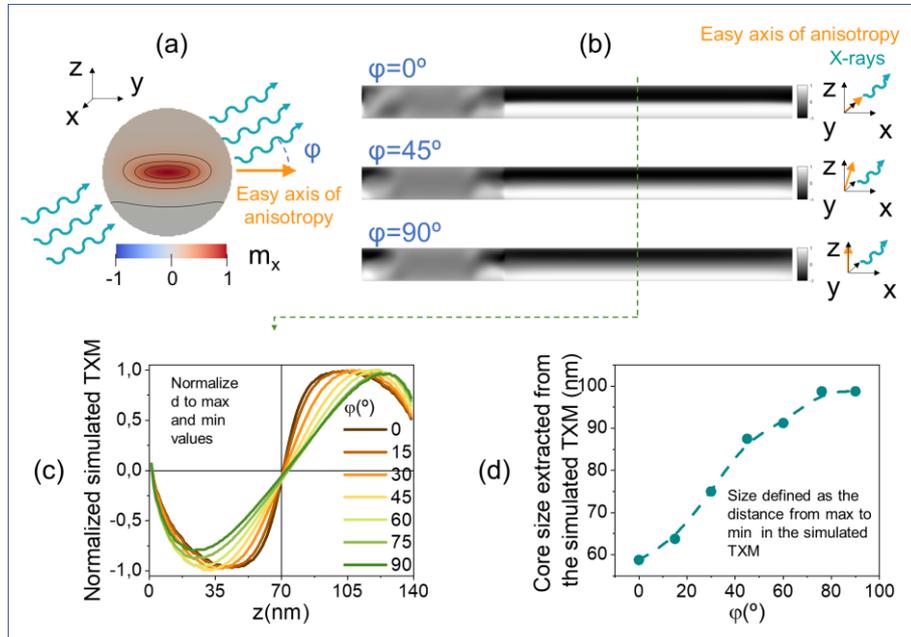


Figure S5. Simulated TXM contrast as a function of the angle between the X-ray beam and the magnetocrystalline easy axis of the CoNi: (a) The X-ray beam crossed the cross-section of a nanowire with a vortex structure with asymmetric core. (b) The TXM contrast calculated for different easy axis orientations. The signal has been normalized to +1 and -1. (c) Contrast extracted in the direction of the dashed lines figures in b). (d) Vortex core size in the direction of the x-ray beam defined as the distance between the maximum and minimum of contrast. The anisotropy easy axis is set to 88°.

Similar results are presented in Tables S2 and S3 for a monovortex and a trivortex states, respectively. The angle between the incident X-rays and the magnetocrystalline easy axis modifies the amount of black and white pixels, evincing the ellipticity of the cores of the vortices. Importantly, these results indicate that the direction of the anisotropy axis can be extracted from angular measurements of TXM on a single wire.

φ (°)	Ni	CoNi	$K_u=3.5e5$ J/m ³ , $\theta=88^\circ$
0			
15			
30			
45			
60			
75			
90			

Table S2. Simulated TXM for a magnetocrystalline easy axis perpendicular to the nanowire in the Ni/CoNi segment that rotates an angle φ around the nanowire. Here there is a monovortex state in the CoNi and nearly axial in Ni.

Importantly, with an easy axis nearly perpendicular to the nanowire the displacement of the core of the vortex and the centre of the TXM contrast due to the vortex shape elongation on Fig. S5(c) is minimum and practically negligible in contrast to what is observed experimentally. This confirms the obliquity of easy axis with respect to the easy axis of the nanowire at an angle different from 90° . The distances between the maximum and minimum of the TXM contrast define approximately a vortex core in the direction of X-rays.

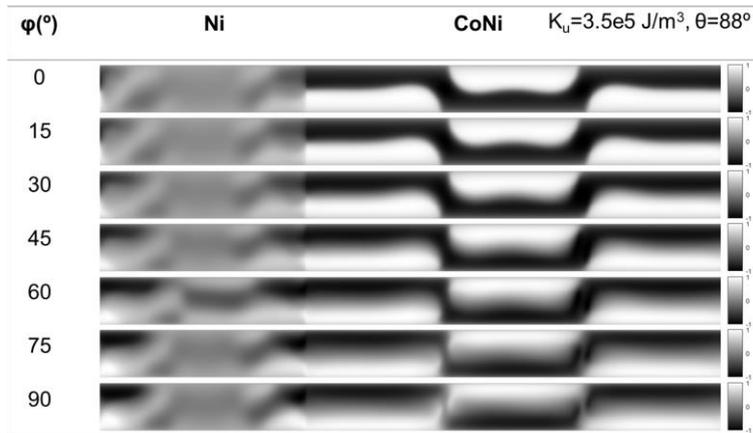


Table S3. Simulated TXM for a magnetocrystalline easy axis perpendicular to the nanowire in the Ni/CoNi segment that rotates an angle φ around the nanowire. Here there is a trivortex state in the CoNi and nearly axial in Ni.

2.2 Dependence of the contrast on the vortex chirality and polarity, and the orientation of the easy axis

Figure S6 illustrates TXM contrast for vortices with different polarities (i.e. the direction of the magnetization in its core) and chiralities (the circulation of the vortex) as a function of the orientation between the magnetocrystalline easy axis and the incident X-rays. An example of polarity and chirality is depicted in Fig. S6(a). The magnetocrystalline easy axis forms 60° with the easy axis of the nanowire in the way that is indicated by the orange arrow in Figure S6(b) in each table.

The left column of each table corresponds to the standard HSL colour encoding characteristic of mumax3 indicating the axial magnetization pointing rightward/leftwards by the red/cyan colour. The right column corresponds to the TXM calculated from the micromagnetic states for each state in the left column.

Importantly, a switch in the chirality switches the characteristic contrast of a vortex from bright-dark to dark-bright. A switch in the polarity switches the contrasts of the domain walls between vortices in the same way. These domain walls are oriented with respect to the easy axis as does the TXM contrast.

The yellow-boxed figures correspond with the TXM contrast compatible with the experimental TXM contrast. Both contrasts correspond to a different orientation of the cores (polarity) of the vortices. A measurement of the axial component allowed us to determine the polarity of the vortex and thus the orientation of the easy axis in the experiment in the three dimensions of space.

This study permitted us the determination of challenging 3D structures such as trivortex states and domain walls between vortices from TXM contrasts. This is, so far,

the first study of such kind on nanowires and the first time that a non-trivial domain wall between vortices is directly measured by one technique.

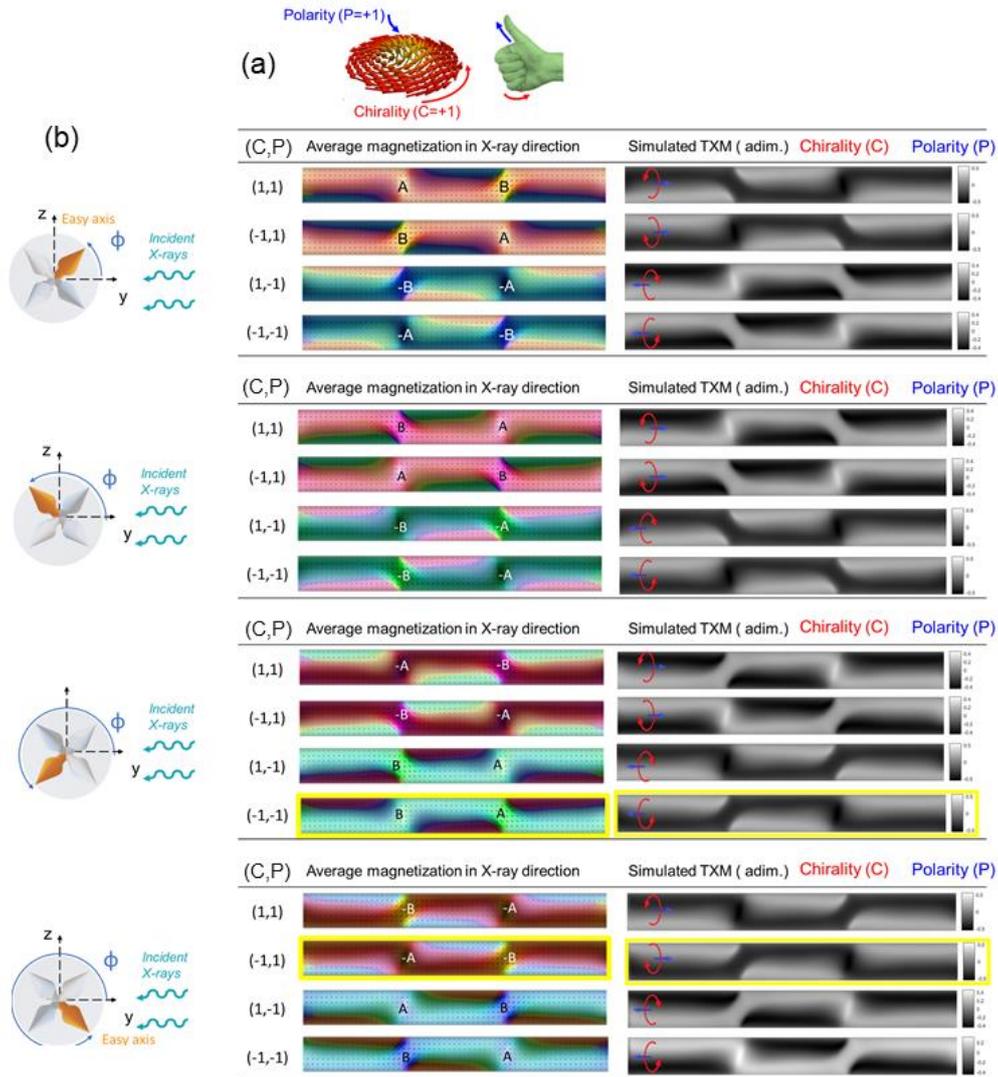


Figure S6. Tables summarizing the simulated TXM contrast for different magnetic structures as a function of the polarity(P), and chirality (C) of the vortices, and the direction of the magnetocrystalline easy axis in the nanowire with respect to the x-ray beam. This direction is indicated by the orange arrow on the left side of each table. The left column of each table shows the magnetization averaged in the direction of the x-ray beam coloured by the standard HSL scale in mumax3. The right column depicts the corresponding simulated TXM-contrast. The labels A and B indicate the positions of the wall, while -A and -B stand for the same domain wall under a reversal symmetry. The red and blue arrows in the right column indicate the polarity of the vortex and the chirality in order to facilitate the interpretation of the black /white contrast.

References:

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2. Ivanov, Y.P.; Vázquez, M.; Chubykalo-Fesenko, O. Magnetic reversal modes in cylindrical nanowires. *J. Phys. D. Appl. Phys.* 2013, 46, 485001, doi:10.1088/0022-3727/46/48/485001.
3. Bran, C.; Fernandez-Roldan, J.A.; Palmero, E.M.; Berganza, E.; Guzman, J.; Del Real, R.P.; Asenjo, A.; Fraile Rodríguez, A.; Foerster, M.; Aballe, L.; et al. Direct observation of transverse and vortex metastable magnetic domains in cylindrical nanowires. *Phys. Rev. B* 2017, 96, 125415, doi:10.1103/PhysRevB.96.125415.