Magnetic Field Magnitudes needed for Skyrmion Generation in a General Perpendicularly Magnetized Film.

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1. Determination of the perpendicular magnetic anisotropy energy strength

The effective PMA energy is defined as $K_{\text{eff}} = \mu_0 H_k M_S / 2$, where $\mu_0 H_k$ and $M_S$ are the anisotropy field and saturation magnetization, respectively; $\mu_0 H_k$ is the effective field holding the magnetization in the perpendicular direction. Because the perpendicular magnetization is tilted when $\mu_0 H_x$ and $\mu_0 H_z$ are applied, the degree of tilt presents the information of PMA energy strength. When $\mu_0 H_x$ and $\mu_0 H_z$ are applied to the PMA sample, the tilted magnetization vector, applied magnetic fields, and effective PMA field are represented as shown in Figure S1a, where the normalized magnetization vector is $\vec{m} = \frac{M_z}{|M_s|}$. The tangent ratio for the magnetization and field is given by the following equations,

$$\tan \theta = \frac{(1 - m_z^2)}{m_z} \quad (S1)$$

$$\tan \theta = \frac{\mu_0 H_x}{\mu_0 H_x m_z + \mu_0 H_z} \quad (S2)$$

Thus, the relation between $m_z$, $\mu_0 H_k$, $\mu_0 H_x$ and $\mu_0 H_z$ is given by

$$\mu_0 H_x = \frac{\sqrt{(1 - m_z^2)}}{m_z} \times (\mu_0 H_k m_z + \mu_0 H_z). \quad (S3)$$

To evaluate the PMA energy strength, we measured the variation in the perpendicular magnetic moment $m_z$ using the polar magneto-optic Kerr effect (MOKE) microscope.
intensity under the application of an external in-plane magnetic field ($\mu_0 H_x$). Before the $\mu_0 H_x$ was applied, a constant $\mu_0 H_z$ (here, $\mu_0 H_z = 3.55$ mT) was applied to form a saturated magnetic state. The measured $m_z$ data according to $\mu_0 H_x$ and $\mu_0 H_z$ (Figure S1b, c) can be fitted by Equation (S3). Figure S1d shows representative experimental data of the sample having a $\mu_0 H_k$ value of 68.0 mT.

**Figure S1.** (a) The normalized magnetization vector and effective PMA field tilted by external magnetic fields. (b, c) The measured $m_z$ data according to the external magnetic field. (d) the experimental data of sample having a $\mu_0 H_k$ value of 68.0 mT, where the error bar is the standard deviation from four measurements about the results of $\pm \mu_0 H_z$ and $\pm \mu_0 H_x$. 
2. Skyrmion density control using a perpendicular magnetic field

Figure S2. Skyrmion MOKE images under various perpendicular magnetic fields

Figure S2a is a MOKE image under zero magnetic field after applying an appropriate magnetic field pulse to generate a bunch of skyrmions. Figure S2(b-f) represent MOKE images under various $z$-axis magnetic fields ($H_z$). As shown in Figure S2, the skyrmion density and radius can be controlled by applying $H_z$ as shown in Figure 5c in the main text.
3. Detailed analysis of the presence of the skyrmion Hall effect

In this work, the motion of magnetic bubble domains is in the creep regime, where pinning dominantly affects the dynamics. Therefore, we need to do statistical analysis using a significant number of data to observe the skyrmion Hall effect.

Figure S3-1. Configuration for measurement of the skyrmion Hall effect.

As Figure S3-1 presents, we created a bunch of magnetic bubble domains using the presented method in the main text, followed by applying a constant 4.6 Oe of $H_z$ to control magnetic bubble domain density. Then, after applying a 0.948 mA current pulse for 500 ms, we obtain the longitudinal velocity ($v_x$) and the transverse velocity ($v_y$). Most magnetic bubble domains do not move, as shown in Figure 1l, which indicates that this dynamics is in the creep
regime. Magnetic bubble domains sometimes move. From more than 4,000 events analysis, we can see the magnetic bubble domain motion has a 9° angle with the current direction.

\[ \text{Figure S3-2. Configuration for measurement of the skyrmion Hall effect.} \]

To obtain clearer images, we investigated magnetic bubble domains under zero magnetic fields, where the magnetic bubble domains were created by using the presented method in the main text. Because zero magnetic field induces the stripe domain state as the ground state, applying a current pulse sometimes elongates the magnetic bubble domains as shown in Figure S3-2. In this state, we can reduce the pinning effect by injecting a current pulse with higher amplitude. It is worth noting that the elongated stripe domain also presents the skyrmion Hall effect due to the presence of a half-skyrmion at the end of the stripe domain.\textsuperscript{1,2} The skyrmion
Hall angle inversion by the core polarity is clearly observed, indicating the magnetic bubble domains investigated in the main text are magnetic skyrmions with a finite topological number.
4. The high $O_{hv}$ region at the boundary between DS and SkD

In our previous work, $^3$ we investigated how the stripe domain changes under in-plane magnetic fields. First, under in-plane magnetic fields, the stripe domain width decreases, and the initial disordered stripe domain (DS) is aligned along the field direction under sufficient in-plane magnetic fields, as shown in the below figure. Here, the sufficient in-plane magnetic fields mean that $H_x > H_{DMI}$.

**Figure S4-1.** Stripe domain MOKE images as a function of $H_k$ and $H_x$

A detailed explanation for the reasons and models are presented in the paper.
Here, the in-plane magnetic fields' role in creating magnetic skyrmions is to reduce the stripe domain width. There is a critical stripe domain width where magnetic skyrmions can be spontaneously created only by a perpendicular magnetic field, as shown in Figure 4(b) in the main text. Therefore, in Figure 1(i), the region just to the left of the SkD region is the result of a slightly small amplitude of the $H_x$ to reach a critical stripe domain width. However, the $H_x$ is still larger than $H_{\text{DMI}}$ (= 163 Oe), which induces the aligned stripe domain. Therefore, we can observe highly aligned stripe domain at the left region of the SkD region.

**Figure S4-2.** MOKE images at each position in the graph of Figure 1(i).
5. Validity of the ultra-thin Ta thickness control

Our samples in the main text are deposited by a wedge technique. Therefore, the samples with various Ta thicknesses represent different positions of one sample, where the Ta thickness is extracted from the deposition rate of the two ends of the 12x12 mm² substrate.

The island growth of the inserted Ta layer is evident because the Ta layer thickness is less than the interatomic radius of Ta atoms, which corresponds to a perfect Ta monolayer thickness. However, now, we should consider the magnetic exchange length; within the magnetic exchange length, spins are uniformly aligned and act like one single spin. Therefore, considering the magnetic exchange length ($l_{ex}$), a ferromagnetic thin-film (< several nm) consists of many individual magnetic moments on a 2-dimensional plane, where the individual spins have averaged magnetic property for each area corresponding to the square of the magnetic exchange length ($l_{ex}^2$), as shown in Figure S5-1.
Figure S5-1. A schematic of the meaning of the magnetic exchange length

Thus, to obtain the magnetic exchange length, we use the well-known equation as follows: \(^4\)

$$l_{ex} = \frac{A}{\sqrt{\left(\mu_0 M_S^2/2\right)}}$$

, where \(A\) is the exchange stiffness \([\text{J/m}]\), \(\mu_0\) is the magnetic permeability \([\text{J/(A}^2\text{ m})]\) and \(M_S\) is the saturation magnetization \([\text{A/m}]\). To obtain the saturation magnetization of local positions of the wedged sample, we utilized the previously reported method to determine \(M_S\) from a hysteresis loop using a MOKE microscopy.\(^5\)
Figure S5-2. An example hysteresis loop for measurement of the saturation magnetization.

From the normalized hysteresis loop in Figure S5-2, we extracted the saturation magnetization of each wedged position as a function of the stripe width, which is around $0.6 \times 10^6 \text{A/m}$, as shown in Figure S5-3.
**Figure S5-3.** Measurement of the saturation magnetization as a function of the stripe domain width

Then, using the previously reported exchange length of a one nm-thick CoFeB layer (20 pJ/m)⁶ and the measured magnetization (0.6 × 10⁶ A/m), the magnetic exchange length \( l_{ex} \) is calculated as 9.40 nm.

Therefore, now, we should consider how many islands of Ta atoms exist in the area of the \( l_{ex}^2 \). As an example, let’s say one Fe-O bonding induces 1 of \( K_u \). Then, if a perfect Ta monolayer covers the entire area of the \( l_{ex}^2 \), the interfacial perpendicular magnetic anisotropy (PMA) from Fe-O bondings will disappear (a of Figure S5-4). If there are no Ta atoms in the area of the \( l_{ex}^2 \), the interfacial PMA energy has the maximum value (for example, 5 in b of Figure S5-4). If several Ta atoms exist, the PMA energy has an intermediate value by distributing the reduced PMA energy to all the Fe atoms in the area of the \( l_{ex}^2 \). Then, we can define the effective Ta thickness as the averaged thickness over the area of the \( l_{ex}^2 \) as shown in Figure S5-4.
Figure S5-4. A schematics for the distribution of perpendicular magnetic anisotropy energy and the concept of the effective thickness.

Furthermore, we also calculate the specific material parameters from the experimental result to clarify the above explanation. The below figure is a plot of $H_k$ (A/m) as a function of the effective Ta thickness (nm), which is just a re-scaled graph from Fig. 2b.
**Figure S5-5.** A plot of the effective perpendicular magnetic anisotropy field as a function of Ta thickness.

The linear fit in Figure S5-5 is \( -3.7 \times 10^7 x + 4.04 \times 10^6 = y \). Using the PMA energy density equation of \( K_{\text{eff}} = \frac{1}{2} \mu_0 M_s H_k = K_u - \frac{1}{2} \mu_0 M_s^2 \), when \( K_u \) is zero, \( H_k \) equals to \( -M_s \).

From the linear fit, the Ta thickness corresponding to \( H_k = -M_s \) is 0.125 nm, which is similar to the Ta interatomic radius. Therefore, we can conclude that an almost monolayer Ta growth on the CoFeB layer completely suppresses the interfacial PMA by blocking the Fe-O bondings.

Now, from the calculated \( l_{\text{ex}} \) (9.40 nm), we can think that there are about 1,000 Fe atoms in the area of \( l_{\text{ex}}^2 \). It means that one Ta atom in the area of \( l_{\text{ex}}^2 \) represents 0.000125 nm (0.125/1000 nm) of the effective Ta thickness. From the slope of the above graph, one Ta atom changes 4,625 (A/m) or 5.8 (mT) of \( H_k \), which has a similar scale to our experimental results.

In addition, large thin-film roughness induced by the Volmer-Weber growth mechanism can act as defects and show spatial variation of magnetic properties. Considering the above
magnetic exchange length explanation, if the Ta island growth is uniform over the scale of the magnetic exchange length, the Ta island does not act as a defect. However, unfortunately, at this stage, we cannot directly investigate the size of as-deposited Ta islands because we do not have access to an in-situ sputtering and scanning probe microscopy. Instead, we examined the spatial distribution of stripe domain width over a large area, as below.

**Figure S5-6.** Stripe domain MOKE images as a function of Ta thickness.
Figure S5-6 shows MOKE images with various stripe domain widths at each position. The spatially continuous stripe width change is well observed, at least in the scale of the \( \mu \text{m} \) scale stripe domain width.

In addition, the stripe domain width change is more dramatic with a tiny change of \( \text{Ta} \) thickness. As explained in the main text, stripe domain width is exponentially proportional to PMA energy density per volume. Therefore, in this experiment, to observe the \( \mu \text{m} \) scale stripe domain, we controlled the \( K_{\text{eff}} = K_u - \frac{1}{2} \mu_0 M_S^2 \) as very small value. However, what the Ta insertion changes is indeed \( K_u \). Therefore, the \( K_{\text{eff}} \) is relatively more changed by the change in \( K_u \). For example, let’s say that we have \( K_{\text{eff}} \) of 5 J/m\(^3\), \( K_u \) of 100 J/m\(^3\) and \( \frac{1}{2} \mu_0 M_S^2 \) of 95 J/m\(^3\). Then, if we reduce the \( K_u \) by only 1 J/m\(^3\) (to 99 J/m\(^3\)), it results in \( K_{\text{eff}} \) of 4 J/m\(^3\), which is 20 % change of \( K_{\text{eff}} \). Therefore, the stripe domain width, which is exponentially proportional to \( K_{\text{eff}} \), is extremely sensitive to the change in \( K_u \) or the Ta insertion thickness.

Based on the above explanation, we believe that the Ta thickness control in pm scale is valid.
6. The role of DMI

For the well-known materials or structures including Pt/Co, Ta/CoFeB, W/CoFeB, \( H_{\text{DMI}} \) is usually smaller than a critical in-plane magnetic field \( (H_{\text{crit}}) \) to reach the critical stripe domain width. Therefore, in this case, applying the \( H_{\text{crit}} \) aligns all the magnetizations in domain walls along the in-plane field direction, not the DMI effective field direction. That is, DMI negligibly affects initial creation of magnetic bubbles without chirality. However, when turning off the magnetic field, the re-alignment of magnetizations in the domain wall is determined the presence of DMI, as shown in the below figure.

![Schematic of stripe domain cutting into skyrmion](image)

**Figure S6.** Schematics of a stripe domain cutting into a skyrmion.\(^7\)

In case that \( H_{\text{crit}} \) is smaller than \( H_{\text{DMI}} \), the disordered stripe domain is not aligned along the in-plane magnetic field direction, and the Zeeman energy term in Eq. (3) disappears. Therefore, although there would be a slight offset change in the tendency of stripe domain width change
under in-plane magnetic fields, the disordered stripe domain width decreases with keeping the disordered stripe domain form. When the stripe domain width reaches the critical stripe domain width, the stripe domains are cut into skyrmions with chirality under a certain perpendicular magnetic field.

For the above two cases, the only difference is whether the stripe domains have chirality or not when they are cut into magnetic bubbles. It can make a difference in the critical stripe domain width because, in the case of the chiral stripe domain, the barrier to cut a stripe domain is lifted as topological barrier energy. However, a more dominant factor for cutting stripe domains into magnetic bubbles is the probability of collision of two adjacent domain walls, which is dominantly determined by the stripe domain width itself. Therefore, we think that DMI only plays a role in determining the chirality of the remaining magnetic bubbles.
7. **Confirmation of the SkD images created by the magnetic pulses of several points**

To check the SkD images of different points in Figure 5a, we obtained five different MOKE images for the samples with $\mu_0H_k = 68.0$ and 83.0 mT. The selected points of the magnetic field pulses are shown in Figure S2a, where the calculated critical magnetic fields are marked as empty stars. After application of the magnetic field pulses, SkD was stabilized. (Figure S2b, c) These results show that the SkD domains created by magnetic field pulses of five different points inside the boundary were almost identical.

**Figure S7.** (a) Various skyrmion generations for the samples with $\mu_0H_k$ values of 68.0 and 83.0 mT. (b, c) The SkD images created by the magnetic pulses of the corresponding points (A–E in Figure S7a) for the samples with $\mu_0H_k$ values of 68.0 and 83.0 mT.
8. Precise evaluation of skyrmion size.

**Figure S8-1.** Schematics of a skyrmion with nine pixels.

Let us consider that a skyrmion exists within nine pixels as shown in Fig. S8-1. The pixel size is 0.357 μm, and one pixel can have an intensity value between 0-255 (8-bits), where 0 indicates the most bright color and 255 is the darkest color. From the brightness of the MOKE signal, we can distinguish the up and down magnetization states. Before observing the skyrmion, a uniform up state and a uniform down state can be created using a sufficient perpendicular magnetic field, and the brightness of each state is normalized to have values of 0 and 255. Then, we can expect that the center pixel has the highest intensity value, and the other pixels have intermediate values proportional to the included skyrmion area. Therefore, we can determine the size of skyrmions with a higher resolution by considering the intensity value of each pixel.
Figure S8-2. (a) MOKE image of a skyrmion. (b) line profile of pixel intensity along the black line in (a).

For example, Fig. S8-2(a) presents a MOKE image of a skyrmion. Fig. S8-2(b) shows the line profile of pixel intensity along the black line in Fig. S8-2(a). Here, we define the intensity value of 255 as the effective one pixel, as described in Fig. S8-1.

Then, to obtain the effective pixel number, we sum the intensity values of all the pixels and divide the result by 255.

\[
\text{Effective pixel number} = \frac{\sum I_i}{255}
\]

One pixel area is \(0.357 \times 0.357 \, \mu m^2\) (\(= 0.127 \, \mu m^2\)). Then, the effective area of a skyrmion is

\[
\text{Effective skyrmion area} = \text{Effective pixel number} \times \text{one pixel area} = \frac{\sum I_i}{255} \times 0.127 \, \mu m^2.
\]

Finally, we obtain the skyrmion radius by the relation; the effective skyrmion area \(= \pi r^2\).

More than 500 skyrmions are investigated for more accurate data analysis to get one skyrmion diameter data point. MOKE images of skyrmions with various diameters are shown in Fig. S8-3.

Figure S8-3. Skyrmion MOKE images under various perpendicular magnetic fields.
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