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Controlled transformation of skyrmions and antiskyrmions in a non-centrosymmetric magnet

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Supplementary Information

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Magnetic property characterization

Supplementary Fig. 1a shows M - T curves of the bulky crystal of $\text{Mn}_{1.4}\text{Pt}_{0.9}\text{Pd}_{0.1}\text{Sn}$ (MnPtPdSn) measured in the zero-field cooling (ZFC) and FC at 0.01 T, indicating the higher Curie temperature (T_C) of ~ 375 K. Supplementary Fig. 1b represents M - H curves without magnetic hysteresis in the bulky MnPtPdSn at 100 K and 300 K.

Numerical simulations for various magnetic textures and their energies

We have performed numerical simulations for various magnetic textures (antiskyrmion, NT-bubble, skyrmion) and their energies with increasing the in-plane magnetic field in a dipole-interaction-dominating 2D system with or without anisotropic DMI, respectively (Supplementary Fig. 2). The Hamiltonian is described as the Eq. 2 in the Method. A set of parameters $\{J = 1.0, K = 0.6, I_{\text{dip}} = 0.09, h_z = 0.023\}$ and an open boundary condition are used for the simulation, where the normalized magnetic moment $\mathbf{n}_r = (n_{r,x}, n_{r,y}, n_{r,z})$ is defined at the $\mathbf{r}$ on the 2D square-lattice sites spanned by the basis set $\{\mathbf{e}_x, \mathbf{e}_y\}$ but $\mathbf{r}$ for $\mathbf{n}_r$ is in a disk inscribed in the 150×150 system. The energy $E_{\text{FM}} (= -J \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} -K \sum_r + I_{\text{dip}} \sum_{\mathbf{r} \neq \mathbf{r}'} |\mathbf{r} - \mathbf{r}'|^{-3} - h_z \sum_r)$ of the ferromagnetic (FM) state ($\mathbf{n}_r = (0, 0, +1)$ for all $\mathbf{r}$ on the 2D square-lattice sites) is taken as the zero-energy level, where the number of the bonds $\sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} 1 = 35028$ and the number of the sites (magnetic moments) $\sum_r 1 = 17663$. In the present dipole-interaction dominating system, the energies of antiskyrmion, NT-bubble, and skyrmion are almost degenerate for both cases of $D = 0$ and $D = 0.01$ in comparison with the energy of the FM state E_{FM} , and are sensitive to the in-plane field.

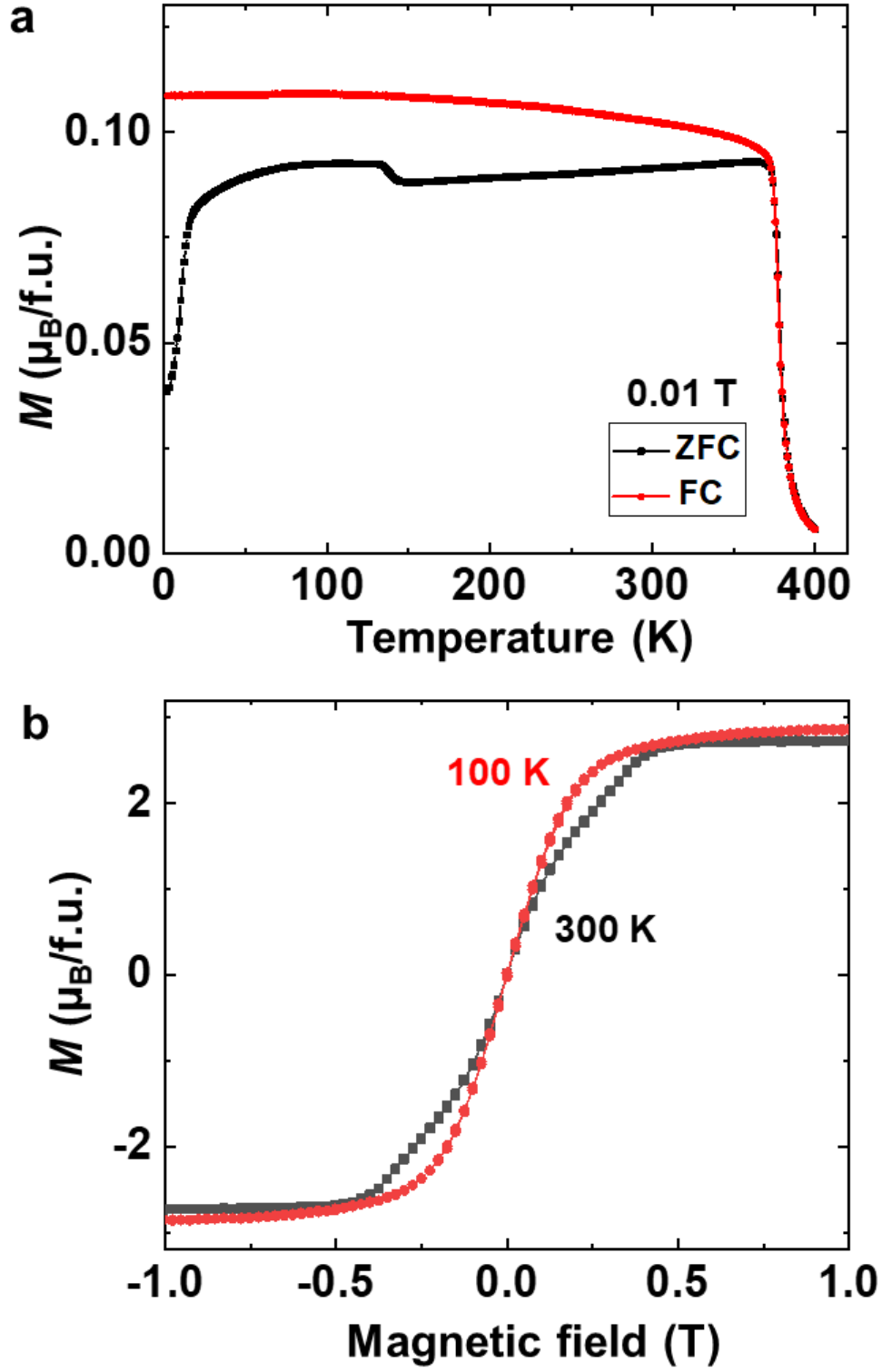
Supplementary Videos 1-2 demonstrate directly the dynamics of skyrmion-to-NT-bubble transformation in a 2D system with the application of the in-plane magnetic fields $h_x = 0.018$ and 0.03 , respectively, which is revealed by changes of the total energy, topological charge, magnetic texture, and the spatial distribution of emergent field b_z ($b_z = \frac{1}{2} \mathbf{n}_r \cdot (\frac{\partial \mathbf{n}_r}{\partial x} \times \frac{\partial \mathbf{n}_r}{\partial y})$) with in-plane field.

Supplementary Fig. 3a shows the time evolution of the total energy of spin configurations at $h_x = 0.016, 0.018$, and 0.03 , respectively. The saturated energy at $h_x = 0.03$ is taken as the zero. At the

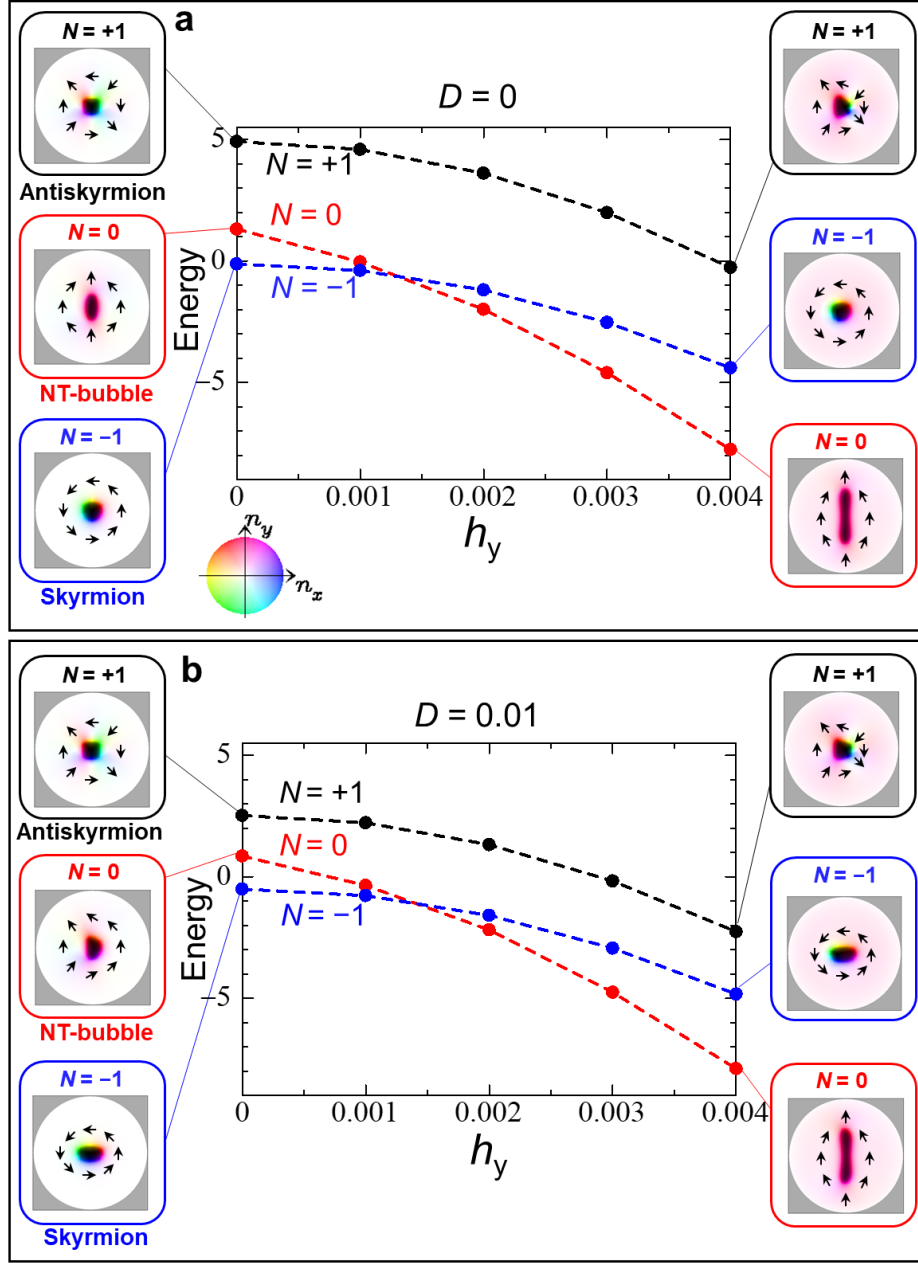
transition point (indicated by a black arrow), the energy decreases rapidly and saturates immediately. The energy barrier between the skyrmion and NT-bubble is $\sim 10 J$ at $h_x = 0.018$. Supplementary Fig. 3b shows the schematic of the free-energy landscape of skyrmion ($N = -1$) and NT-bubble ($N = 0$) at $h_x = 0, 0.016, 0.018$, and 0.03 , respectively, indicating the in-plane field-induced changes in the energy barrier between the skyrmion and the NT-bubble, and hence the fluctuation between such two magnetic textures: When $h_x = 0$, the energy level of skyrmion is lower than that of NT-bubble, indicating that the skyrmion is more stable than the NT-bubble; With increasing h_x up to 0.016 , both energies of skyrmion and NT-bubble decrease while the energy of skyrmion becomes higher than that of NT-bubble at $h_x = 0.016$ and 0.018 , energetically revealing the reasonable transformation of skyrmion into NT-bubble; When the field is increased further to 0.03 , energies of both magnetic textures decrease dramatically and approach the zero. Note that the energy barrier is of the order of $10J$ and is not that large compared with what is expected from the large number of spins involved in the textures. This means that the local singular configurations leading to the change in the topological number determine the energy barrier. These local singularities during the transient transformation process, however, are hard to capture by using L-TEM.

Supplementary Fig. 4 represents various magnetic textures with $N = -1$ in a dipole-interaction-dominating 2D system with and without the anisotropic DMI, respectively. The shape of the skyrmion shows an isotropic circle (the left panel in Supplementary Fig. 4a) in a dipole-interaction system without DMI, whereas it changes to elliptical (the right panel in Supplementary Fig. 4a) with adding $D = 0.01$. The formation of such an elliptically-deformed skyrmion arises reasonably from the competition between the magnetic dipole interaction and the anisotropic DMI. The quantitative calculation of DMI strength is difficult, but it is possible to be roughly estimated from the elliptic form of observed skyrmions. Note that the length scale η of the skyrmion is determined by the ratio of the exchange interaction J and the effective interaction energy D_{eff} of either magnetic dipole interaction or the DMI as $\eta = \frac{J}{D_{eff}}a$ (a , lattice constant). In the presence of both magnetic dipole interaction and

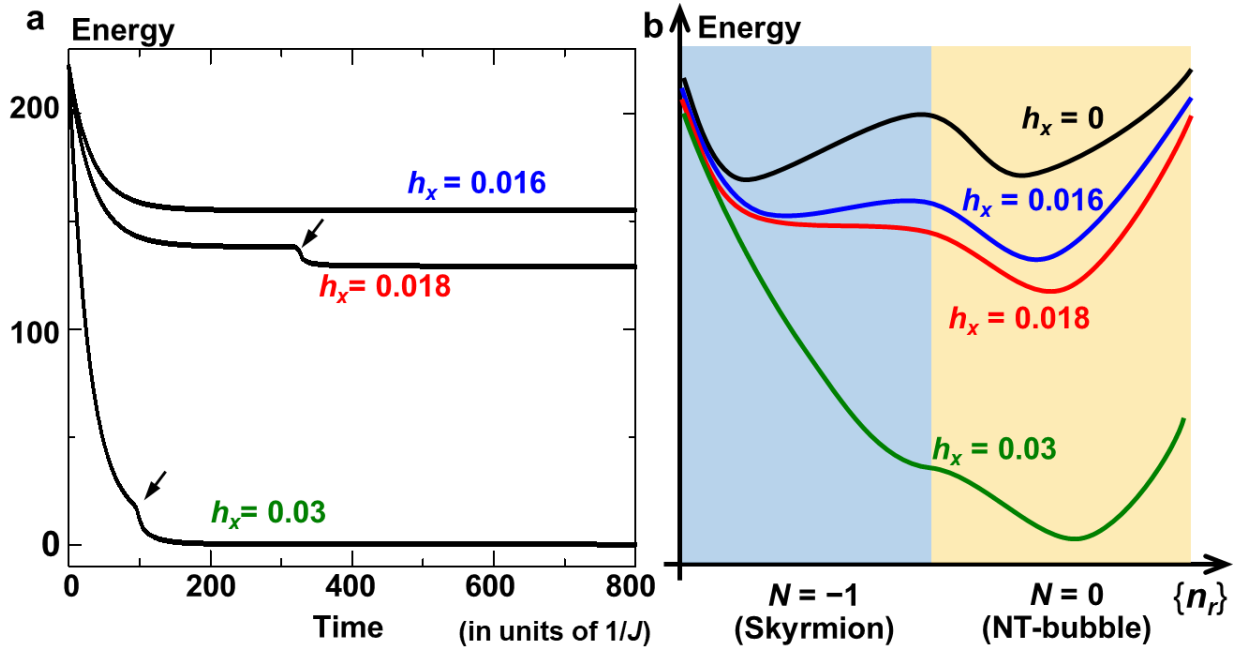
DMI, η depends on the direction because D_{eff} is given by $D_{dip} + D$ and $D_{dip} - D$, respectively, with D being the DMI. The ellipticity of the deformed skyrmion observed here (Supplementary Fig. 4b) is $\frac{\eta_{long}}{\eta_{short}} = 1.57$, which is estimated from a L-TEM image (Supplementary Fig. 4b). Here, the η_{long} and η_{short} are the orthogonal axis lengths of the elliptic skyrmion, respectively. Therefore, the ratio of D_{dip} to D can be roughly estimated from the equation $\frac{D_{dip}+D}{D_{dip}-D} = \frac{D_{short}}{D_{long}} = \frac{\eta_{long}}{\eta_{short}} = 1.57$. (D_{long} and D_{short} are the *effective* interaction energies along x and y axes, respectively). According to this estimation, D_{dip} is 4.5 times as large as D in the present magnet.



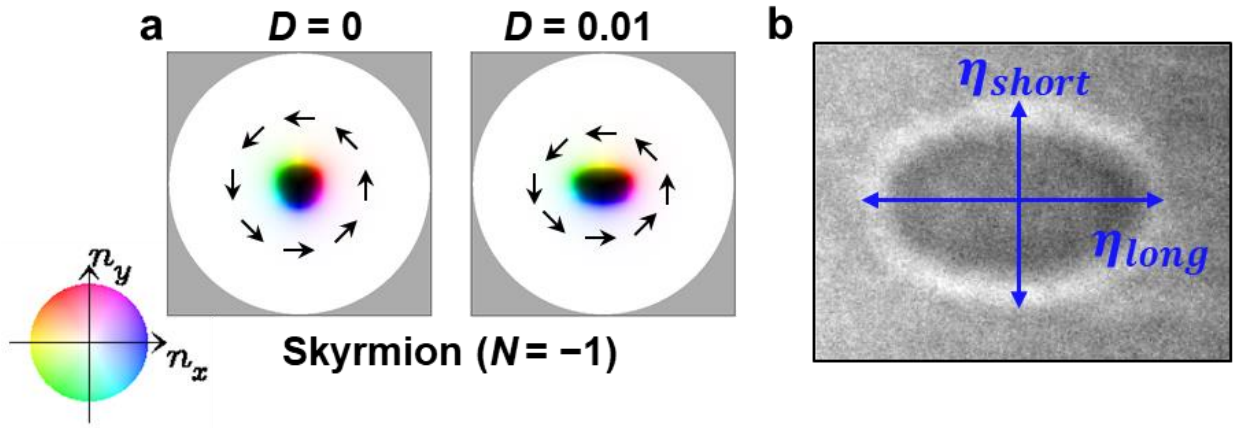
Supplementary Fig. 1 | Magnetic property characterization. **a**, M - T curves under ZFC and 0.01T-FC processes. **b**, M - H curves respectively measured at 100 K and 300 K.



Supplementary Fig. 2 | Energies of antiskyrmion (black), NT-bubble (red), and skyrmion (blue) with increasing the in-plane magnetic field h_y under the conditions of (a) DMI strength $D = 0$ and (b) $D = 0.01$, respectively. A set of parameters $\{J = 1.0, K = 0.6, I_{dip} = 0.09, \text{ and } h_z = 0.023\}$ and an open boundary condition are used here. The energy of the ferromagnetic state $\mathbf{n}_r = (0, 0, +1)$ is taken as the zero. Left and right panels in (a-b) show the magnetic textures of antiskyrmion, NT-bubble, and skyrmion at $h_y = 0$ and $h_y = 0.004$, respectively. The colour wheel in (a) and the arrows surrounding the magnetic textures in (a-b) represent the directions of in-plane magnetic moments. The darkness represents the out-of-plane components, i.e., black (white) represents $n_{r,z} = -1$ ($n_{r,z} = +1$).



Supplementary Fig. 3 | **a**, Numerical simulations showing the time evolution of the total energy of magnetic textures under the in-plane magnetic fields $h_x = 0.016, 0.018$, and 0.03 , respectively. The saturated energy at $h_x = 0.03$ is taken as the zero. **b**, Schematic of the free-energy landscape based on the numerical simulations in **a** with the change of magnetic moments $\{n_r\}$ from skyrmion ($N = -1$) to NT-bubble ($N = 0$) at $h_x = 0, 0.016, 0.018$, and 0.03 , respectively. The short dark arrows denote the transition points.



Supplementary Fig. 4 | **a**, Simulated skyrmions showing circle-shape and elliptically-deformed with the anisotropic DMI strength $D = 0$ and $D = 0.01$, respectively, in a magnetic dipole-interaction dominating system. **b**, Experimentally-observed elliptically-deformed skyrmion.