

Supplementary Materials

***In situ* visualization reveals the magnetization reversal behavior of shape-anisotropic nanostructures**

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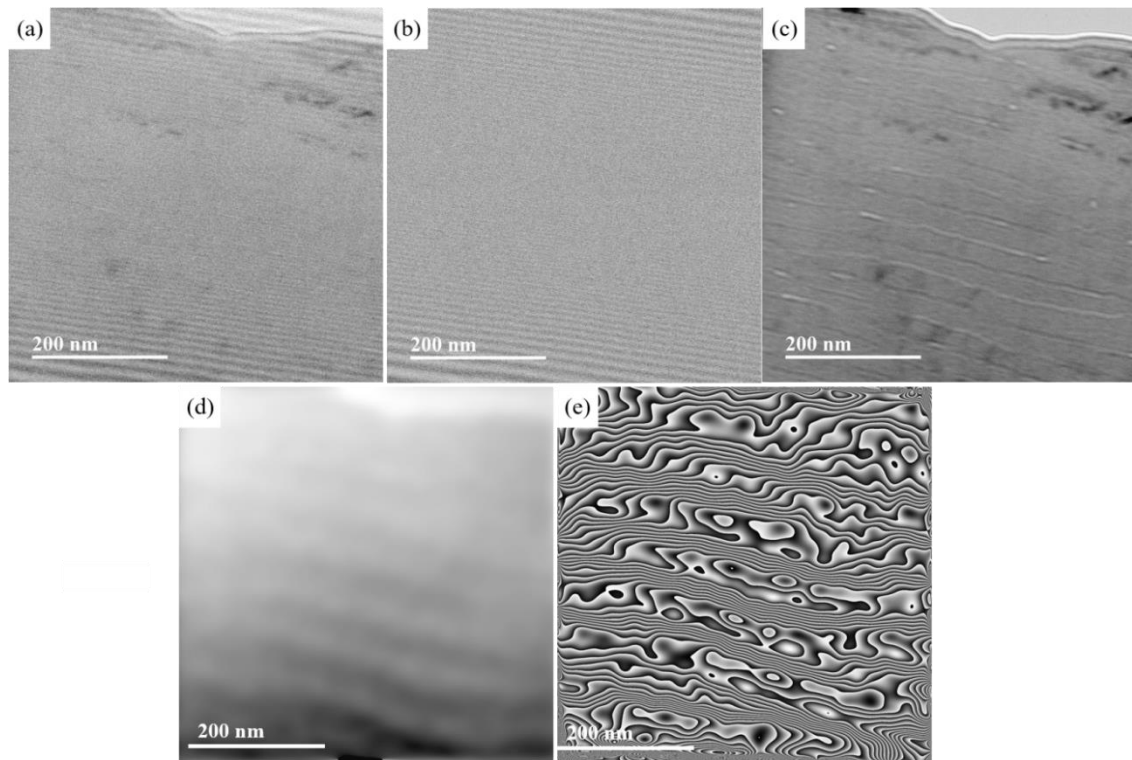
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Electron holography experiments were conducted on a JEOL 2100F LTEM system with an acceleration voltage of 200 kV, an electrostatic biprism voltage of 45 V, and an exposure time of 3s per hologram. The data acquisition and reconstruction workflow is described as follows: first, an electron hologram of the target sample region was collected [Supplementary Figure 1A]; the sample was then retracted, and a reference hologram of the vacuum region [Supplementary Figure 1B] was recorded under identical parameters to subtract systematic phase offset. Raw phase shift images [Supplementary Figure 1D] were extracted from the sample and reference holograms by Fourier transform using Digital Micrograph (DM) software, and further processed by cosine amplification to reconstruct the final magnetic phase map [Supplementary Figure 1E]. The phase shift in the phase map is proportional to the line integral of magnetic induction along the electron beam propagation direction. Combined with the calibrated sample thickness, the saturation magnetic induction of the α_1 and α_2 phases

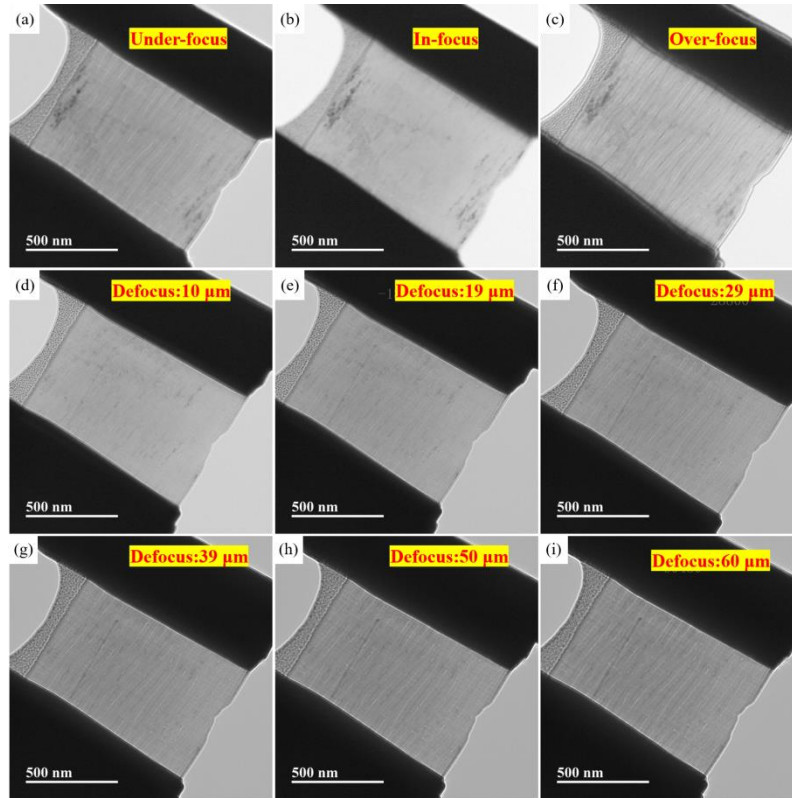
can be quantitatively derived.



Supplementary Figure 1. (A) electron hologram of Alnico; (B) electron hologram of vacuum; (C) Fresnel image of the corresponding region; (D) the phase shift image extracted from (A) and (B), (E) equal-phase map obtained via $\cos(\text{phase})$ transformation.

To differentiate magnetic domain wall contrast from α_1/α_2 phase boundary contrast in Fresnel imaging and validate the reliability of in-situ magnetization reversal observations, we recorded Lorentz transmission electron microscopy (LTEM) images of the same field of view under varying focus conditions. At in-focus condition, magnetic contrast from domains is nearly absent, and only morphological contrast from phase interfaces remains. By contrast, domain walls show characteristic black-and-white contrast reversal under under-focus and over-focus conditions (as shown in Supplementary Figure 2A-C), which effectively rules out interference from phase boundary morphological contrast and confirms the magnetic nature of the observed features. A series of Fresnel images acquired at different defocus values further verifies the magnetic origin of the contrast: as defocus increases stepwise from 10 μm to 60 μm , the contrast intensity of domain walls rises steadily while domain wall positions remain unchanged, fully consistent with the defocus-dependent

behavior of magnetic contrast (as shown in Supplementary Figure 2D-I). These results establish the reliability of dynamic observations of the in-situ magnetization reversal process.



Supplementary Figure 2. Under-focus (A), in-focus (B), and over-focus (C) images obtained by Lorentz transmission electron microscopy, along with Fresnel images at different defocus (D) 10 μm ; (E) 19 μm ; (f) 29 μm ; (G)39 μm ; (H) 50 μm ; (I) 60 μm .

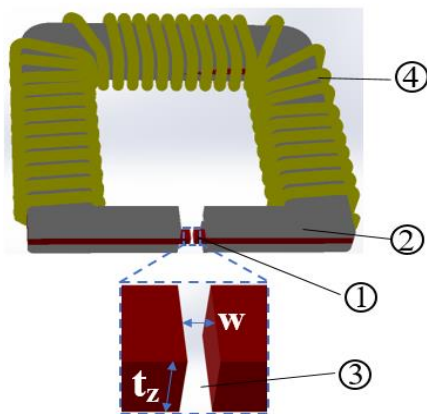
The following details the procedure for measuring the magnetic field strength of the micro-electromagnet. Supplementary Figure 3 shows a schematic diagram of a micro electromagnet, with a magnetic field range of approximately $6 \times 20 \mu\text{m}$. It is impossible to directly measure the magnetic field within such a small range. We first used finite element simulation to calculate the distribution of the magnetic field within the gap, then measured the electron beam deflection angle by measuring the distance of the electron diffraction spot movement, and then calculated the relationship between the magnetic field and the coil current. Supplementary Figure 4 shows the magnetic field distribution obtained by finite element simulation at a current of 150 mA. It can be seen that except for a decrease in magnetic field in the edge region, the internal magnetic field is uniform. During the experiment, we placed the sample in a

uniform magnetic field area.

The deflection angle of the electron beam can be calculated using formula (1):

$$\theta = \frac{\lambda e}{h} B_a \int f(z, I_a) dz \quad (1)$$

where λ , e , h is the electron wavelength, electron charge, and Planck constant, respectively. B_a is the maximum value of the magnetic field under different excitation currents, $\int f(z, I_a) dz$ is the integration along the electron beam path of magnetic field distribution $f(z, I_a)$. The value of the magnetic field can be obtained by measuring the deflection angle θ . Although the finite element analysis can be used to calculate B_a by inputting the excitation current and the soft magnetic properties of the magnetic core, the method of obtaining the magnetic field strength by measuring θ can obtain more accurate experimental values.

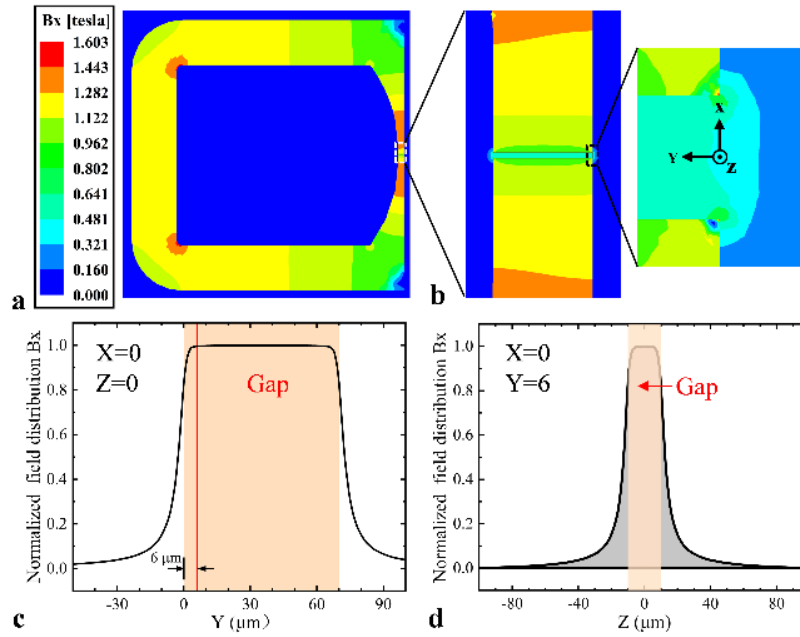


Supplementary Figure 3. Structure diagram of micro-magnetic field generating device. ①-Soft magnetic core; ②-Non-magnetic titanium alloy sheet; ③-Gap; ④-Coil.

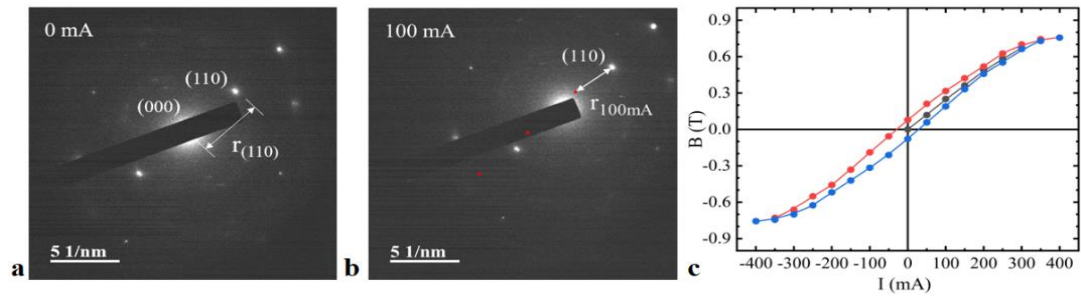
Supplementary Figure 5A shows the center point and (110) diffraction point of alnico when the excitation current is 0 in the demagnetization state of the core of the magnetic field generator. $r_{(110)}$ is the distance between the center point and (110) diffraction point. According to Bragg's law, the Bragg angle of the (110) crystal face is

$$\theta_B = \frac{r_{(110)}}{L} = \frac{\lambda_{200KV}}{2d_{(110)}} = \frac{0.00251 \text{ nm}}{2 \times 0.2028 \text{ nm}} = 6.19 \text{ mrad} \quad (2)$$

where L is the camera length, λ_{200KV} is wavelength of electrons, and $d_{(110)}$ is the crystal plane spacing of (110) crystal face. Supplementary Figure 5B shows the diffraction spots when the excitation current is 100 mA. The spots when the current is 0 are represented by red dots. The displacement of the diffraction spot is $r_{100mA} = 0.68r_{(110)}$, so $\theta = r_{100mA}/L = 4.21 \text{ mrad}$. Substitute $\int f(z, I_a) dz = 27.46 \text{ T} \cdot \mu\text{m}$ into Formula (2), we can get $B_a = \theta h / (\lambda e \int f(z, I_a) dz) = 0.2529 \text{ T}$. In this way, we calculate the magnetic induction intensity corresponding to the current from 0 to +400 mA and from +400 mA to -400 mA. The relationship between the current and the magnetic induction intensity is shown in Supplementary Figure 3C. When the current is 400 mA, the maximum magnetic field value is 0.75 T, and the electron deflection angle is 12.5 mrad.



Supplementary Figure 4. (A) The magnetic induction intensity distribution of the central plane ($Z = 0$) of the core thickness at 150 mA; (B) amplification diagram and coordinate origin at the air gap region; (C) the normalized magnetic field distribution along the Y axis ($X = Z = 0$); (D) the normalized magnetic field distribution along the Z axis ($X = 0, Y = 6$).



Supplementary Figure 5. (A) The electron diffraction spots of alnico when the excitation current is 0 mA; (B) The electron diffraction spots of alnico when the excitation current is 100 mA; (C) The corresponding relationship between excitation current and magnetic field.