

1 Switching the magnetization of a ferromagnetic layer with a current induced spin-orbit torques
2 requires the breaking symmetry, either via an in-plane magnetic field, or in the case of field-free
3 switching via a device asymmetry. Here, Liang et al carefully control the Burgers vector of crystal
4 dislocations to break the in-plane symmetry and allow for field-free switching of magnetization in a
5 Pt/Co heterostructure.

Field-free Spin-orbit Switching of Perpendicular Magnetization Enabled by Dislocation-induced In-plane Symmetry Breaking

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25 Abstract

26 Current induced spin-orbit torque (SOT) holds great **promise** for next generation magnetic-
27 memory technology. Field-free SOT switching of perpendicular magnetization requires the breaking
28 of in-plane symmetry, which can be artificially introduced by external magnetic field, exchange
29 coupling **or** device asymmetry. **Recent it has been** shown that the exploitation of inherent crystal
30 symmetry offers a simple and **potentially** efficient route towards field-free switching. However,
31 **applying** this approach to the benchmark SOT materials such as ferromagnets and heavy metals is
32 **challenging**. Here, we **present** a strategy to break the in-plane symmetry of Pt/Co heterostructures
33 by designing the orientation of Burgers vectors of dislocations. We show that the lattice of Pt/Co is
34 tilted by about 1.2° when the Burgers vector has an out-of-plane component. Consequently, a tilted
35 magnetic easy axis is induced and can be tuned from nearly in-plane to out-of-plane, enabling the
36 field-free SOT switching of perpendicular magnetization components at room temperature with a
37 relatively low current density ($\sim 10^{11}$ A/m²) and excellent stability ($> 10^4$ cycles). This strategy is
38 expected to be applicable to engineer a wide range of symmetry-related functionalities for future
39 electronic and magnetic devices.

41 Main Text

42 Modern magnetic-memory technology requires all-electric control of magnetization to realize
43 devices with low energy consumption, high storage density and good thermal stability¹. Recent
44 advances have revealed that current-induced SOT provides an efficient means to electrically
45 manipulate magnetic states²⁻⁴. In typical heavy metal (HM)/ferromagnetic metal (FM)
46 heterostructures, a charge-current flowing in the plane gives rise to a spin-current along the out-of-
47 plane direction according to spin-Hall scenario⁵⁻⁷. This spin-current is injected into FM layer,
48 exerting a damping-like torque $\tau_{DL} \propto \mathbf{M} \times \boldsymbol{\sigma} \times \mathbf{M}$ and a field-like torque $\tau_{DL} \propto \boldsymbol{\sigma} \times \mathbf{M}$ on the
49 magnetization $\mathbf{M}$, with $\boldsymbol{\sigma}$ being the spin polarization⁷. Considering the symmetry of heterostructure,
50 the $\boldsymbol{\sigma}$ is in-plane and therefore a deterministic switching of perpendicular magnetization requires an
51 additional in-plane symmetry breaking⁸. Previously, the in-plane symmetry breaking was artificially
52 introduced by external magnetic fields³⁻⁷, exchange coupling⁹⁻¹³, and device asymmetry¹⁴⁻¹⁹.
53 Recently, the exploitation of inherent crystal structure has been shown as a simple and efficient route

54 to realize field-free SOT switching of perpendicular magnetization. However, so far, demonstration
55 has been limited to only a few unconventional materials with low crystal symmetry²⁰⁻²³. It is thus
56 highly desirable to extend this symmetry-design approach to the benchmark SOT materials of
57 HM/FM (such as Co, Pt, etc.), which remains a grand challenge since these materials usually show
58 high crystal symmetry.

59 Here, we develop a strategy that uses dislocations to induce in-plane symmetry breaking in
60 magnetic heterostructures. Dislocations, as one-dimensional defects prevailing in crystals, were
61 commonly regarded as problematic for functional materials²⁵. It has recently been shown that a
62 proper engineering of dislocations could improve materials properties²⁶⁻²⁸. Dislocations disturb the
63 perfect alignment of a crystal lattice, which potentially provides a microstructural tool to control the
64 symmetry. Through rationally designing the orientation of Burgers vectors $\mathbf{B}$ of dislocations in
65 model system of Pt/Co heterostructures on oxide templates, we realize the tilting of crystal lattice
66 of Pt/Co by about 1.2° with respect to the underlying oxide templates. This lowering of crystal
67 symmetry leads to a tilted magnetic easy axis that can be tuned from nearly in-plane to out-of-plane.
68 Furthermore, a field-free SOT switching at room temperature is successfully demonstrated with a
69 relatively low current density ($\sim 10^{11}$ A/m²) and excellent stability ($> 10^4$ cycles). Given that
70 dislocations are prevailing in different classes of materials, this strategy should be generally
71 applicable to manipulate the symmetry-related functionalities for high-performance devices.

72 **Design of dislocations for crystal tilting**

73 In epitaxial heterostructures, the lattice mismatch between epitaxial layers and underlying
74 substrates leads to the formation of dislocations near the interfaces in order to reduce the elastic
75 energy. Dislocations introduce structural irregularities in perfect crystals, such as an extra half
76 atomic plane introduced by an edge dislocation. This translational discontinuity, which is described
77 by the Burgers vectors $\mathbf{B}$, depends on both the crystal structure of epitaxial layer and interface
78 condition. In our model systems, Pt and Co in the bilayer heterostructure have face-centered-cubic
79 (FCC) crystal structure^{29, 30}, and therefore their Burgers vectors $\mathbf{B}$ are along $\frac{1}{2}\langle 110 \rangle$ directions³¹.
80 We consider three main crystalline orientations of the heterostructures, e.g. (100)-, (110)- and (111)-
81 orientations. As shown in Fig. 1a, for the (100)-orientation, there are two equivalent $\frac{1}{2}\langle 110 \rangle$

directions in the film plane. Therefore, the lattice mismatch between metal layers and underlying substrates can be fully accommodated by dislocations with in-plane Burgers vectors **B** (see Fig. 1b). As a result, the extra half atomic plane is normal to the underlying substrates and the crystal lattice is not expected to tilt as shown in Fig. 1c. Similar arguments are also valid for the (111)-orientation with three equivalent $\frac{1}{2}\langle 110 \rangle$ directions in the film plane (see Supplementary Fig. 1). By contrast, for the (110)-orientation (see Fig. 1d), there is only one $\frac{1}{2}\langle 110 \rangle$ direction within the film plane. Therefore, in order to fully accommodate the lattice mismatch across the interface, dislocations with Burgers vector that has an out-of-plane component are indispensable (see Fig. 1e). As schematically illustrated in Fig. 1f, this type of dislocation is expected to induce the tilting of the crystal lattice^{32, 33}, which can be quantified by the angle θ_e . As a result, the system becomes asymmetric relative to a 180° rotation along the normal direction of underlying substrates, leading to the breaking of in-plane symmetry.

To verify the above designing strategy, we fabricated textured Pt/Co bilayer heterostructures on NiO/MgO templates with the aforementioned three crystalline orientations (see Methods and Supplementary Fig. 2). Both MgO and NiO have the rock-salt structure with lattice constants (~4.2 Å) larger than that of textured Pt/Co films^{29, 34}, which would lead to the formation of dislocations at the interfaces. The high-angle annular dark-field scanning transmission electron microscope (HAADF-STEM) is employed to characterize the atomic structure of the heterostructures. For the (100)-oriented heterostructure, dislocations with only in-plane Burgers vector are observed (Fig. 2a), revealed by drawing a Burgers circuit around the dislocation (white dash line). The tilting angle θ_e , as defined in Fig. 1f, is found to be close to 0° by performing statistical analysis of the HAADF-STEM results (Fig. 2b and Supplementary Fig. 3). By contrast, for the (110)-oriented heterostructure, dislocations with Burgers vectors that have both in-plane and out-of-plane components are observed (Fig. 2c). The crystal lattice of Co/Pt shows a clear tilting with respect to the oxide templates, with θ_e around 1.2° via statistical analysis (Fig. 2d and Supplementary Fig. 4) and arrays of dislocations at the interface (see Supplementary Fig. 5). We note that the magnitude of θ_e and dislocation density are in good agreement with our theoretical calculations based on the low-angle boundary model (see Supplementary Note 1 and Supplementary Fig. 6). In addition, the crystal lattice tilting is further detected by synchrotron-based X-ray diffraction, as shown in

111 Supplementary Fig. 7.

112

113 **Emergence of titled magnetic easy axis**

114 Owing to the tilting of crystal lattice in the (110)-oriented heterostructures, the original mirror
115 symmetry with respect to the (001)-plane is broken, and thereby a tilted magnetic easy axis with an
116 angle θ_M is expected as shown in Fig. 3a. Fig. 3b shows the magnetic hysteresis results for the
117 Pt/Co/NiO heterostructure with the (110)-orientation, measured along different crystallographic
118 directions. The results exhibit a magnetic easy (hard) axis along the [001] ($\bar{1}\bar{1}0$) direction within
119 the film plane. In addition, the finite remnant magnetization (M_r) along the out-of-plane ($\bar{1}10$)
120 direction reveals the tilting of the magnetic easy axis, showing the angle θ_M around 16° by using θ_M
121 $= \arcsin(M_r/M_s)$ (M_s is the saturation magnetization). We also observe that the magnitude of θ_M
122 decreases by increasing the Co layer thickness (t_{Co}) in the heterostructures (see Supplementary Fig.
123 8 and Fig. 9), indicating the interfacial origin of the perpendicular magnetization component.

124 To further verify the tilted anisotropy, we measure polar magneto-optic Kerr effect (MOKE)
125 and anomalous Hall effect (AHE) signals, both of which mainly correspond to the perpendicular
126 magnetization component. Through in-situ monitoring the change of polar MOKE signals as
127 sweeping magnetic field along the [001] direction, we find that the perpendicular magnetization is
128 reversibly switched by reversing the in-plane magnetic field (Fig. 3c, Supplementary Figs. 10b and
129 11), which strongly supports the presence of the tilted magnetic easy axis in the (110)-oriented
130 heterostructures (see Supplementary Fig. 10a). This is further supported by the shift of polar MOKE
131 hysteresis loop as sweeping out-of-plane field, when small in-plane bias field is applied³⁵ (see
132 Supplementary Fig. 12 and Fig. 13). Furthermore, we also measure the polar angular (γ) dependence
133 of Hall resistance (R_{xy}) by rotating the external magnetic field ($H = 100$ mT) within the ($\bar{1}10$) plane,
134 as shown in Fig. 3d. The results reveal that the perpendicular magnetization switches when the angle
135 γ is around 160° and 340° , which are the characteristics of the tilted magnetic easy axis³⁶. By
136 comparing experimental data in Fig. 3d to our simulation results, the magnitude of θ_M is estimated
137 to be around 20° , consistent with the hysteresis results in Fig. 3b (see Supplementary Note 2 and
138 Supplementary Fig. 14). By contrast, this tilted magnetic easy axis is not observed in

139 heterostructures with (100)- and (111)-orientations, see Supplementary Fig. 15.

140 The observation of tilted magnetic easy axis in the (110)-orientated heterostructures implies its
141 strong correlation to the tilting of crystal lattice. Below we discuss the underlying mechanisms by
142 considering different contributions to the magnetic anisotropy of the Pt/Co heterostructures,
143 including shape anisotropy, magneto-crystalline anisotropy, magneto-elastic anisotropy and
144 interfacial anisotropy. The shape anisotropy of ultrathin films favors the magnetic easy axis within
145 the film plane. As revealed by previous reports, the presence of easy (hard) axis along [001] ($\bar{1}\bar{1}0$)
146 in-plane direction is mainly determined by the magneto-elastic effect^{37, 38}. Moreover, the interfacial
147 contribution leads to an easy axis along the film normal direction^{30, 39}. By considering the energy
148 competition using a phenomenological model, we can determine the angular dependence of the
149 anisotropy energy and thereby the direction of magnetic easy axis (see Supplementary Note 3 and
150 Supplementary Fig. 16). When the crystal lattice of (110)-oriented Co/Pt does not show any titling
151 ($\theta_e = 0^\circ$), our simulation results show that the magnetic easy axis would be along either the out-of-
152 plane [110] ($\theta_M = 90^\circ$) or in-plane [001] ($\theta_M = 0^\circ$) axis. Moreover, we find that the presence of
153 crystal lattice tilting ($\theta_e > 0^\circ$) affects the angular dependence of anisotropy, leading to the tilting of
154 magnetic easy axis within the ($\bar{1}\bar{1}0$) plane. Our simulation results further reveal that the magnitude
155 of θ_M depends on both the tilting angle of crystal lattice (θ_e) and the relative strength of anisotropy
156 energies. As shown in Fig. 3e, we find that the θ_M can be continuously tuned from 16° to 84° by
157 inserting a thin Pt layer with controlled thicknesses ($t_{\text{in-Pt}}$) between Co and NiO (see details in
158 Supplementary Fig. 17), which is most likely due to the modulation of interfacial perpendicular
159 magnetic anisotropy energy³⁸ and agrees well with our simulation results.

160

161 **Field-free SOT switching of perpendicular magnetization**

162 We finally study the field-free SOT switching in the (110)-orientated heterostructures. Two
163 representative types of heterostructures are used for demonstration, with different magnitudes of θ_M
164 as schematically shown in Fig. 4a ($\theta_M = 16^\circ$) and 4b ($\theta_M = 84^\circ$). Current pulses (I_{pulse}) are injected
165 along the Hall bar channels (see Supplementary Fig. 18), which are along both in-plane $[\bar{1}\bar{1}0]$ and
166 [001] directions. Perpendicular magnetization is probed by measuring the anomalous Hall resistance

R_{xy} . Fig. 4c and 4d show the changes of Hall resistance (ΔR_{xy}) by sweeping I_{pulse} without magnetic field for the two heterostructures respectively. When I_{pulse} is along $[\bar{1}10]$, the deterministic switching of perpendicular magnetization is observed for both heterostructures. Compared with remnant Hall resistance at $\mu_0 H = 0$ mT, the field-free SOT switching ratio is estimated to be about 70% for the $\theta_M = 16^\circ$ sample, and about 60% for the $\theta_M = 84^\circ$ sample (see Supplementary Figs. 19). By contrast, no obvious switching events occur when I_{pulse} is along $[001]$ direction in both heterostructures, see Fig. 4c and 4d. The SOT efficiencies as current applied along both in-plane directions are also estimated by second Harmonic Hall resistance^{14,40,41}, showing values close to similar heterostructures⁴² (see Supplementary Note 4 and Supplementary Fig. 20). Further measurements by varying the magnitude and pulse-width of currents indicate that the Joule heating effect is likely to assist the switching of perpendicular magnetization in our samples (see Supplementary Note 5, Supplementary Fig. 21 and Fig. 22). Nevertheless, the distinct switching behaviors as current pulses applied along two in-plane directions in (110)-oriented heterostructures, along with the absence of field-free SOT switching in (100)- and (111)-oriented heterostructures (see Supplementary Fig.23), demonstrate the essential role of tilted anisotropy induced by the tilting of crystal lattice.

These results above can be explained by considering the symmetry between spin polarization (σ) induced by the spin-Hall effect in Pt and the tilted magnetic easy axis. As schematically shown in Fig. 4e, by applying current along $[\bar{1}10]$ axis, the σ provided by Pt exerts a damping-like torque τ_{DL} on the magnetization of Co and forces the magnetization to align parallel to σ during I_{pulse} ^{7,10,18}. After removing the I_{pulse} , the magnetization will relax to downward direction due to the presence of the tilted magnetic easy axis. Similarly, inverting current direction switches the magnetization from downward to upward, leading to the deterministic switching behaviors. This scenario is further supported by the field-dependent SOT switching when external magnetic fields are applied parallel to the current pulse, see Supplementary Note 6, Supplementary Figs. 24 and 25. The critical switching current density J_c of our device is about 3×10^{11} A/m² (normalized by thickness and width of Hall bar), which is comparable or better than the other typical field-free SOT systems at room temperature using the conventional ferromagnetic and heavy metals, such as CoFeB/Ta/CoFeB with interlayer coupling¹⁰ ($\sim 10^{12}$ A/m²) and Pt/Co/NiO with exchange bias¹³ ($\sim 10^{12}$ A/m²). Moreover, we also test the stability and endurance of field-free switching events by reversibly applying current

196 pulses along $[\bar{1}10]$ direction for more than 10^4 cycles. As shown in Fig. 4f and Supplementary Fig.
197 26, our results reveal that the field-free switching is highly reversible without clear fatigue.

198 As the in-plane symmetry breaking is enabled by the design of dislocations, our strategy to
199 realize the field-free SOT switching should be potentially extended to other materials. Indeed, we
200 also successfully realize the field-free SOT switching of perpendicular magnetization in Pd-based
201 heterostructures on (110)-oriented oxide templates, see Supplementary Fig. 27. Moreover,
202 compared with other approaches to realize tilted magnetic anisotropy^{17,43,44}, our strategy does not
203 rely on complex fabrication process or delicate interlayer coupling, and exhibits excellent
204 performances (see Supplementary Note 7 and Supplementary Table 1). Given that oxides with
205 different orientations can be deposited on silicon⁴⁵, we believe this strategy to break in-plane crystal
206 symmetry by designing the orientation of Burgers vectors **B** provides a simple and applicable path
207 approaching the field-free SOT devices.

208 In conclusion, we have demonstrated that the in-plane symmetry in magnetic heterostructures
209 of benchmark SOT materials can be broken by properly designing the orientation of Burgers vector
210 **B** of dislocations. Such an in-plane symmetry breaking leads to a tilted magnetic easy axis that can
211 be tuned from being nearly in-plane to nearly out-of-plane, and enables a reliable field-free SOT
212 switching of perpendicular magnetization at room temperature with relatively low current density
213 and excellent stability. This revelation has a broader implication for designing the symmetry of
214 functional materials, which include but not limit to magnetoelectronics, ferroelectrics, topotronics,
215 antiferromagnetic spintronics, etc.

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218 **References**

- 219 1. Kent, A. & Worledge, D. A new spin on magnetic memories. *Nat. Nanotech.* **10**, 187-191 (2015).
- 220 2. Manchon, A. et al. Current-induced spin-orbit torques in ferromagnetic and antiferromagnetic
221 systems. *Rev. Mod. Phys.* **91**, 035004 (2019).
- 222 3. Khang, N.H.D., Ueda, Y. & Hai, P.N. A conductive topological insulator with large spin Hall effect
223 for ultralow power spin-orbit torque switching. *Nat. Mater.* **17**, 808-813 (2018).

- 224 4. Baumgartner, M. et al. Spatially and time-resolved magnetization dynamics driven by spin-orbit
225 torques. *Nat. Nanotech.* **12**, 980-986 (2017).
- 226 5. Liu, L. et al. Spin-torque switching with the giant spin Hall effect of tantalum. *Science* **336**, 555-558
227 (2012).
- 228 6. Miron, I. M. et al. Perpendicular switching of a single ferromagnetic layer induced by in-plane
229 current injection. *Nature* **476**, 189-193 (2011).
- 230 7. Liu, L. et al. Current-Induced Switching of Perpendicularly Magnetized Magnetic Layers Using Spin
231 Torque from the Spin Hall Effect. *Phys. Rev. Lett.* **109**, 096602 (2012).
- 232 8. Garello, K. et al. Symmetry and magnitude of spin-orbit torques in ferromagnetic heterostructures.
233 *Nat. Nanotech.* **8**, 587-593 (2013).
- 234 9. Lau, Y.-C. et al. Spin-orbit torque switching without an external field using interlayer exchange
235 coupling. *Nat. Nanotech.* **11**, 758-762 (2016).
- 236 10. Kong, W. J. et al. Spin-orbit torque switching in a T-type magnetic configuration with current
237 orthogonal to easy axes. *Nat. Commun.* **10**, 1-7 (2019).
- 238 11. Luo, Z. et al. Chirally coupled nanomagnets. *Science* **363**, 1435-1439 (2019).
- 239 12. Fukami, S. et al. Magnetization switching by spin-orbit torque in an antiferromagnet-ferromagnet
240 bilayer system. *Nat. Mater.* **15**, 535-541 (2016).
- 241 13. Grochot, K. et al. Current-induced magnetization switching of exchange-biased NiO
242 heterostructures characterized by spin-orbit torque. *Phys. Rev. Applied* **15**, 014017 (2021).
- 243 14. Z. Zheng. et al. Field-free spin-orbit torque-induced switching of perpendicular magnetization in a
244 ferrimagnetic layer with a vertical composition gradient. *Nat. Commun.* **12**, 1-9 (2021).
- 245 15. Wu, H. et al. Chiral symmetry breaking for deterministic switching of perpendicular magnetization
246 by spin-orbit torque. *Nano Lett.* **21**, 515-521 (2020).
- 247 16. Yu, G. et al. Switching of perpendicular magnetization by spin-orbit torques in the absence of
248 external magnetic fields. *Nat. Nanotech.* **9**, 548-554 (2014).

- 249 17. You, L. et al. Switching of perpendicularly polarized nanomagnets with spin orbit torque without an
250 external magnetic field by engineering a tilted anisotropy. *Proc. Natl. Acad. Sci. U. S. A.* **112**, 10310-
251 10315 (2015).
- 252 18. Liu, L. et al. Current-induced magnetization switching in all-oxide heterostructures. *Nat. Nanotech.*
253 **14**, 939-944 (2019).
- 254 19. Kang, Min-Gu. et al. Electric-field control of field-free spin-orbit torque switching via laterally
255 modulated Rashba effect in Pt/Co/AlO_x structures. *Nat. Commun.* **12**, 7111 (2021).
- 256 20. Liu, L. et al. Symmetry-dependent field-free switching of perpendicular magnetization. *Nat.*
257 *Nanotech.* **16**, 277-282 (2021).
- 258 21. Kao, I. et al. Deterministic switching of a perpendicularly polarized magnet using unconventional
259 spin-orbit torques in WTe₂. *Nat. Mater.* **21**, 1029–1034 (2022).
- 260 22. MacNeill, D. et al. Control of spin-orbit torques through crystal symmetry in WTe₂/ferromagnet
261 bilayers. *Nat. Phys.* **13**, 300-305 (2017).
- 262 23. Lee, S., Ryu, J. & Park, BG. Magnetization switching through symmetry. *Nat. Nanotech.* **16**, 227–
263 228 (2021).
- 264 24. Park, D.-S. et al. Induced giant piezoelectricity in centrosymmetric oxides. *Science* **375**, 653-657
265 (2022).
- 266 25. Chu, M.-W. et al. Impact of misfit dislocations on the polarization instability of epitaxial
267 nanostructured ferroelectric perovskites. *Nat. Mater.* **3**, 87-90 (2004).
- 268 26. Hameed, S. et al. Enhanced superconductivity and ferroelectric quantum criticality in plastically
269 deformed strontium titanate. *Nat. Mater.* **21**, 54-61 (2022).
- 270 27. Sugiyama, I. et al. Ferromagnetic dislocations in antiferromagnetic NiO. *Nat. Nanotech.* **8**, 266–270
271 (2013).
- 272 28. Höfling, M. et al. Control of polarization in bulk ferroelectrics by mechanical dislocation imprint.
273 *Science* **372**, 961-964 (2021).
- 274 29. Sun, Y. et al. Electric-field modulation of interface magnetic anisotropy and spin reorientation

275 transition in (Co/Pt)₃/PMN–PT heterostructure. *ACS Appl. Mater. Inter.* **9**, 10855-10864 (2017).

276 30. Nakajima, N. et al. Perpendicular magnetic anisotropy caused by interfacial hybridization via
277 enhanced orbital moment in Co/Pt multilayers: Magnetic circular x-ray dichroism study. *Phys. Rev.*
278 *Lett.* **81**, 5229-5232 (1998).

279 31. Hull, D. & Bacon, D. J. *Introduction to dislocations* (Butterworth-Heinemann, 2001).

280 32. Ayers, J. E., Ghandhi, S. K. & Schowalter, L. J. Crystallographic tilting of heteroepitaxial layers. *J.*
281 *Cryst. Growth.* **113**, 430-440 (1991).

282 33. Jia, C. L. et al. Effect of a single dislocation in a heterostructure layer on the local polarization of a
283 ferroelectric layer. *Phys. Rev. Lett.* **102**, 117601 (2009).

284 34. Baltz, V. et al. Antiferromagnetic spintronics. *Rev. Mod. Phys.* **90**, 015005 (2018).

285 35. Han, D. S. et al. Long-range chiral exchange interaction in synthetic antiferromagnets. *Nat. Mater.*
286 **18**, 703–708 (2019).

287 36. Kan, D. et al. Tuning magnetic anisotropy by interfacially engineering the oxygen coordination
288 environment in a transition metal oxide. *Nat. Mater.* **15**, 432-437 (2016).

289 37. Hillebrands, B. & Dutcher, J.R. Origin of very large in-plane anisotropies in (110)-oriented Co/Pd
290 and Co/Pt coherent superlattices. *Phys. Rev. B* **47**, 6126-6129 (1993).

291 38. Lin, C.-J. et al. Magnetic and structural properties of Co/Pt multilayers. *J. Magn. Magn. Mater.* **93**,
292 194-206 (1991).

293 39. Johnson, M.T. et al. Magnetic anisotropy in metallic multilayers. *Rep. Prog. Phys.* **59**, 1409 (1996).

294 40. Wu, H. et al. Room-temperature spin-orbit torque from topological surface states. *Phys. Rev. Lett.*
295 **123**, 207205 (2019).

296 41. Fan, W. et al. Asymmetric spin-orbit-torque-induced magnetization switching with a noncollinear
297 in-plane assisting magnetic field. *Phys. Rev. Appl.* **11**, 034018 (2019).

298 42. Pai, C. F. et al. Determination of spin torque efficiencies in heterostructures with perpendicular
299 magnetic anisotropy. *Phys. Rev. B* **93**, 144409 (2016).

300 43. Li, Z. et al. Field-free magnetization switching induced by bulk spin-orbit torque in a (111)-oriented
301 CoPt single layer with in-plane remanent magnetization. *ACS Appl. Electron. Mater.* **4**, 4033-4041
302 (2022).

303 44. Kim, H. J. et al. Field-free switching of magnetization by tilting the perpendicular magnetic
304 anisotropy of Gd/Co multilayers. *Adv. Funct. Mater.* **32**, 2112561 (2022).

305 45. Chen, X. Y. et al. Selective growth of (100)-, (110)-, and (111)-oriented MgO films on Si (100) by
306 pulsed laser deposition. *J. Appl. Phys.* **91**, 5728-5734 (2002).

307

308 **Methods**

309 **Sample fabrication**

310 Conventional pulse layer deposition (PLD) method was utilized to fabricate an epitaxial NiO layer,
311 using a KrF excimer pulsed laser with a wavelength of 248 nm, repetition rate of 5 Hz and energy
312 density as 1.4 J/cm². The deposition temperature and oxygen pressure were controlled at 650 °C and
313 6.7 Pa, respectively. The distance between target and substrate was set as 8 cm, to obtain a stable
314 deposition rate of 0.8 nm/min. The miscut angle of MgO substrates is less than 0.2°. After deposition,
315 the NiO film was immediately transferred to a magnetron sputtering chamber with a background
316 vacuum around 2×10⁻⁸ Torr, heated to 150 °C for 15 minutes, to avoid possible gas absorbed on the
317 surface. Then dc sputtering was employed to deposit Co and Pt layers at room temperature. During
318 the deposition, the substrate was rotated to ensure spatial homogeneity. The Co layer was deposited
319 with power as 15 W under 3 mTorr, and the rate was controlled as 0.6 nm/min. The Pt layer was
320 deposited with power as 30 W under 2 mTorr, and the rate was controlled as 2 nm/min. The samples
321 were patterned into Hall bar devices with a width of 20 μm by photolithography and Ar ion etching
322 for further transport measurement and current-switching experiments. Electrodes and contact pads
323 were formed by using photolithography and a lift-off process.

324

325 **HAADF-STEM experiments and the determination of tilting of crystal lattice**

Cross sectional STEM samples were prepared using focused ion beam (FIB, Zeiss Auriga) with Ga^+ ions. Samples were thinned to less than 100 nm at an accelerating voltage of 30 kV with a decreasing current from the maximum of 600 pA, followed by fine polishing at an accelerating voltage of 5 kV with a small current of 20 pA, and then further milled with Ar ions to remove the surface damage. The STEM experiments were performed on an electron microscope equipped with double aberration correctors (Titan Cubed Themis G2 300). The atomic-resolved HAADF images were collected with a convergence angle of 25 mrad and a collection angle of 64 – 200 mrad. To determine the tilting of crystal lattice on the metal/oxide interface, we determined the directions of adjacent atoms arrows of metal layer and NiO near metal/oxide interface, respectively, and the angle between two directions is defined as tilting angle θ_c .

Characterization

The epitaxy relationship to the substrate was characterized by Empyrean high-resolution X-ray diffractometer produced by Company Panalytical with $\text{Cu K}_{\alpha 1}$. A Superconducting quantum interfere device (SQUID) magnetometer was utilized to characterize basic magnetic properties of samples by vibrating magnetometer mode. The polar MOKE microscopy produced by Evico (German) was employed to measure the change of polar MOKE signal with the application of in-plane magnetic field. [The synchrotron XRD data were obtained at 1W1A Diffuse X-ray Scattering Station, Beijing Synchrotron Radiation Facility \(BSRF-1W1A\).](#) The transport property was measured in Physical Property Measurement System (PPMS). In current-induced SOT switching of perpendicular magnetization experiments, a pulsed current with a duration of 2 ms was applied for each data point. After 6 s, a small a.c. excitation current (0.5 mA) is utilized to measure the Hall resistance.

Data availability

The data supporting the findings of this study are available within the paper and its Supplementary Information and Supplementary Data, and also from the corresponding author upon reasonable request.

354

355 **Acknowledgments** We thank Prof. Nian-Xiang Sun and Dr. Liang Wu for fruitful discussions. We
356 thank Dr. Hao Bai, Dr. Yiqing Dong for SOT experiments supporting. [A portion of this work is based](#)
357 [on the data obtained at BSRF-1W1A. The authors gratefully acknowledge the cooperation of the](#)
358 [beamline scientists at BSRF-1W1A beamline.](#) Y.-H. L and R. Y. were supported by the Basic Science
359 Center Project of the National Natural Science Foundation of China (NSFC) (grant numbers
360 51788104). W.-J. J thank National Natural Science Foundation of China (Grant Nos. 52271181,
361 51831005), the National Key R&D Program of China (Grant No. 2022YFA1405100), the NSF
362 Distinguished Young Scholars Grant (No.12225409), Beijing Natural Science Foundation (Grant
363 No. Z190009), the Beijing Advanced Innovation Center for Future Chip (ICFC). D. Y. was
364 supported by National Natural Science Foundation of China (52002204, 92163113). T. N. was
365 supported by National Natural Science Foundation of China (52073158, 52161135103) and
366 National Key R&D Program of China (2021YFA0716503). Y. Z. was supported by Natural Science
367 Foundation of China (Grant No. 52002370). J.-M. H. acknowledges support from the NSF award
368 CBET-2006028. J. S. is grateful for the partial financial support by JSPS KAKENHI Grant Number
369 19H00864.

370

371 **Author contributions** Y.-H.L., Y.L., D.Y. and T.N. conceived this study. Y.L., D.Y. and T.N.
372 performed this study with the supervision of Y.-H. L.. J.-M.H., Y.L., H.C. and M.D. performed the
373 phenomenological simulations. Y.L., D.Y. and T.N. fabricated the samples and carried out the
374 transport and magnetic measurement. R.Y. and S.L. performed the STEM characterizations. W.-J.J.,
375 Y.L. and T.X. conducted the SOT measurement. W.-J.J., Y.L. and L.Z. conducted the polar MOKE
376 characterization. J.S., Y.L. and Y.-H.L. discussed and analysis the results. Y.L., D.Y., T.N., W.-J.J.,
377 R.Y. and Y.-H.L. wrote the manuscript. All authors discussed the results and revised the manuscript.

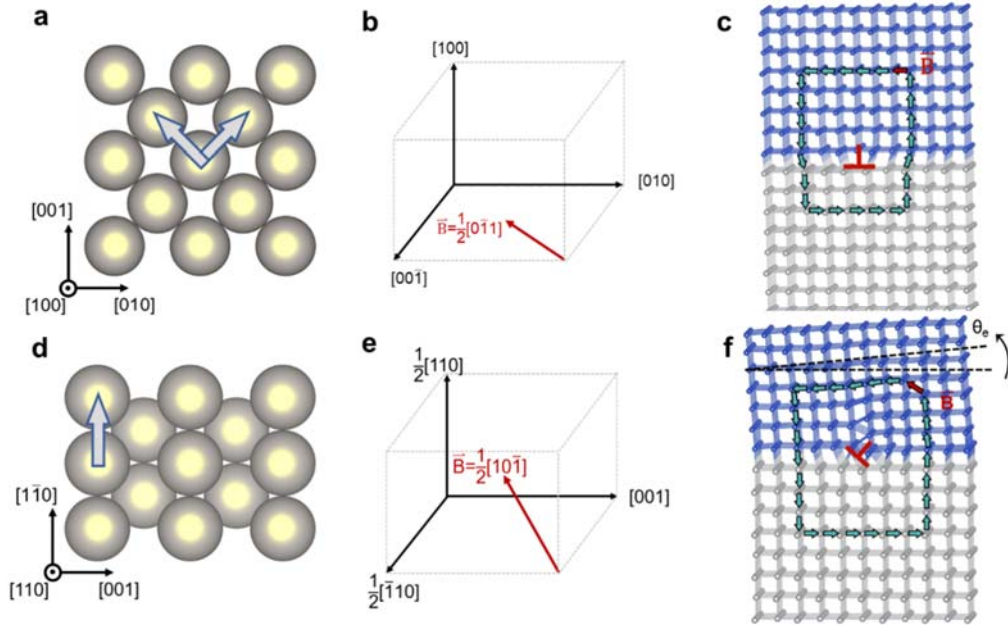
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379 **Competing interests** The authors declare no competing interests.

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381 **Additional information** Supplementary Information is available for this paper.

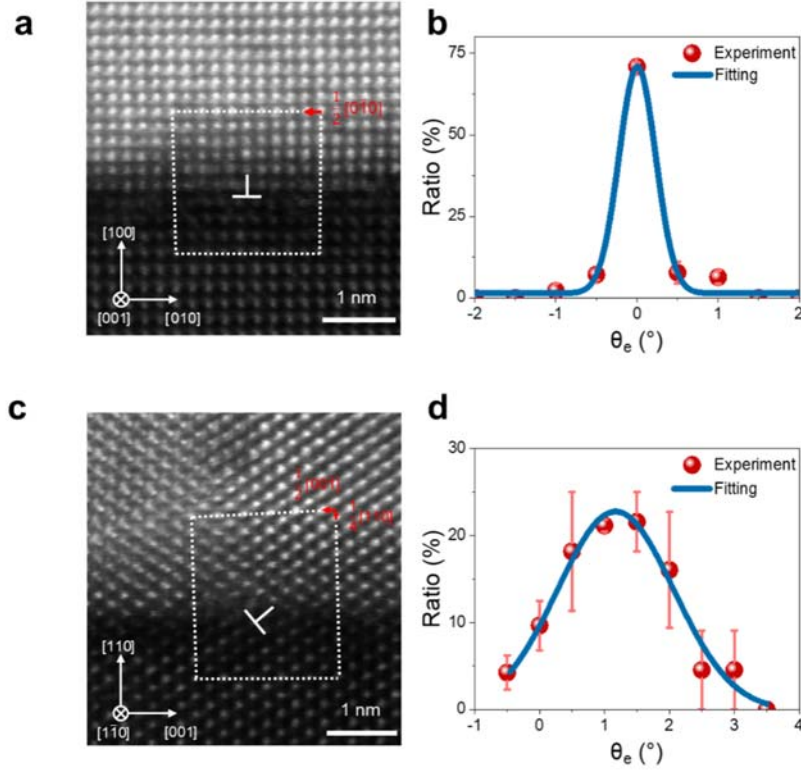
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384 **Fig. 1. The schematic of in-plane crystal symmetry breaking by dislocations.** **a**, For the (100)-
 385 orientation, there are two $\frac{1}{2}\langle 110 \rangle$ lattice vectors in the film plane. The gold balls indicate the atoms
 386 constructing FCC lattice. Representative Burgers vector **B** is shown in **b**, resulting in **c**, dislocation
 387 with in-plane Burgers vector and no tilting of crystal lattice. **d**, For (110)-orientation, there is only
 388 one $\frac{1}{2}\langle 110 \rangle$ lattice vector in the film plane. Therefore, Burgers vector **B** with out-of-plane
 389 component are required. Representative Burgers vector **B** is shown in **e**, results in **f**, dislocation with
 390 an out-of-plane Burgers vector component and tilting of crystal lattice that is quantified by the angle
 391 θ_e . The gray arrows indicate $\frac{1}{2}\langle 110 \rangle$ lattice vectors. Gray and blue balls indicate two distinct crystal
 392 lattices, and the Burgers circuits and Burgers vectors are indicated by green and red arrows,
 393 respectively. It is noted that FCC structure is not shown in Figs. **c** and **f** for better visualization.

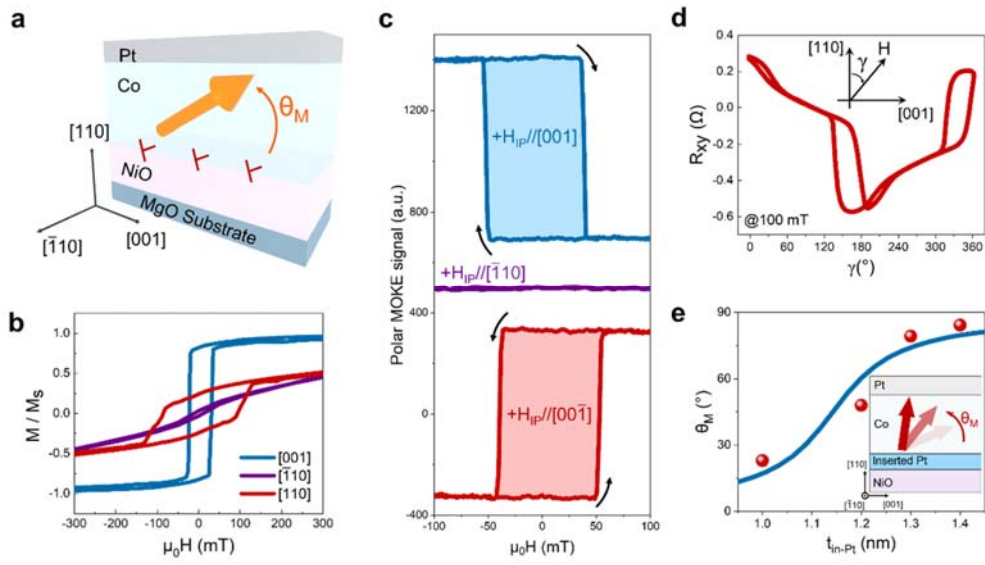
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396 **Fig. 2. The observation of in-plane crystal symmetry breaking by dislocations.** **a**, HAADF-
 397 STEM image of the (100)-oriented Pt(5)/Co(1.5)/NiO(20)/MgO heterostructure. Values in
 398 parentheses are thickness in nm. The statistical results of θ_e are shown in **b**. **c**, HAADF-STEM
 399 image of the (110)-oriented Pt(5)/Co(1.5)/NiO(20)/MgO heterostructure. The statistical results of
 400 θ_e are shown in **d**. The white dash boxes show the Burgers circuits, and the red arrows show the
 401 projected Burgers vectors of dislocations. The symbol $\perp$ indicates the dislocations. The blue line is
 402 the gaussian fitting of the data. The error bars represent the standard deviation of θ_e .

403



404

405 **Fig. 3. The controllable tilted magnetic easy axis induced by in-plane symmetry breaking.** **a**,
 406 the schematic diagram for the tilted magnetic easy axis induced by dislocations in (110)-oriented
 407 Pt/Co/NiO/MgO heterostructures. **b**, the magnetic hysteresis loops along the [001]-, $[\bar{1}10]$ -, [110]-
 408 direction of (110)-oriented Pt(5)/Co(1.2)/NiO(20)/MgO. **c**, field dependence of polar MOKE signals
 409 by sweeping the magnetic field along in-plane [001] and $[\bar{1}10]$ directions. Note that polar MOKE
 410 signals mainly correspond to the perpendicular magnetization. **d**, the polar angular (γ) dependent Hall resistance (R_{xy}) by rotating external magnetic
 411 field H in the $(\bar{1}10)$ plane. The inset is the geometry set for the measurement. **e**, the modulation of
 412 θ_M by increasing the thickness of inserting Pt layer (t_{in-Pt}), with blue line as guideline to the eye. The
 413 inset is the schematic diagram of the sample for the controllable tilted magnetic easy axis by
 414 inserting Pt layer between the Co and NiO layers.

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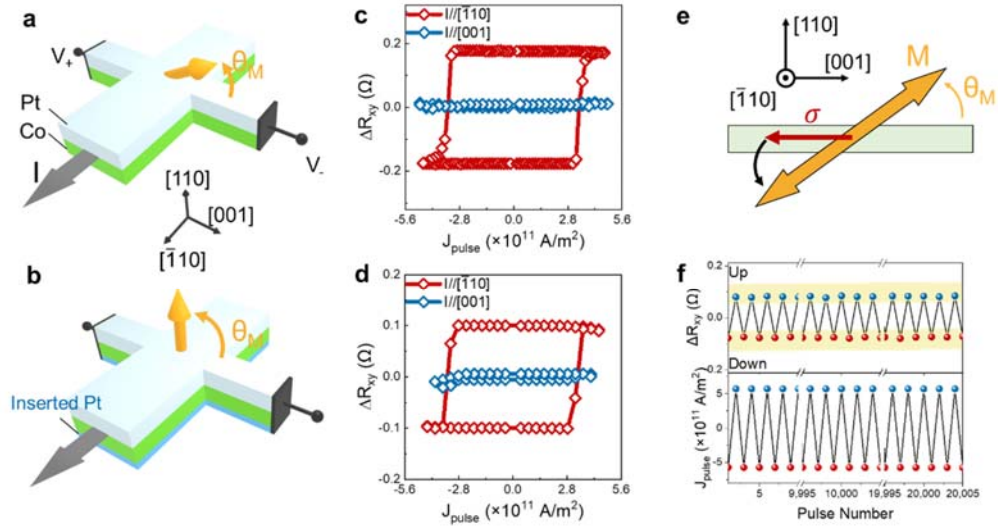
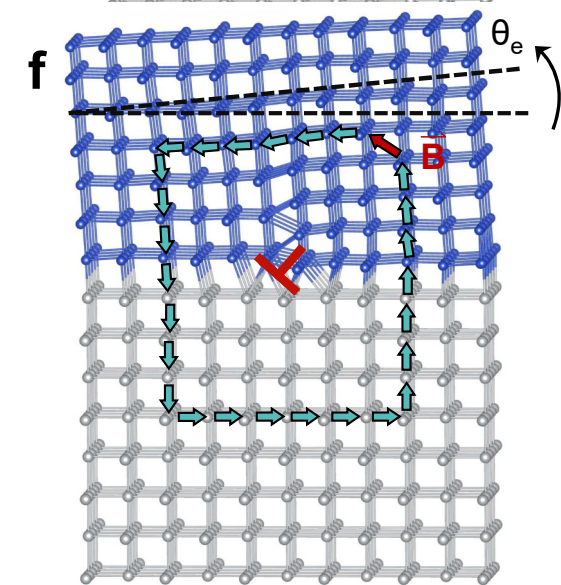
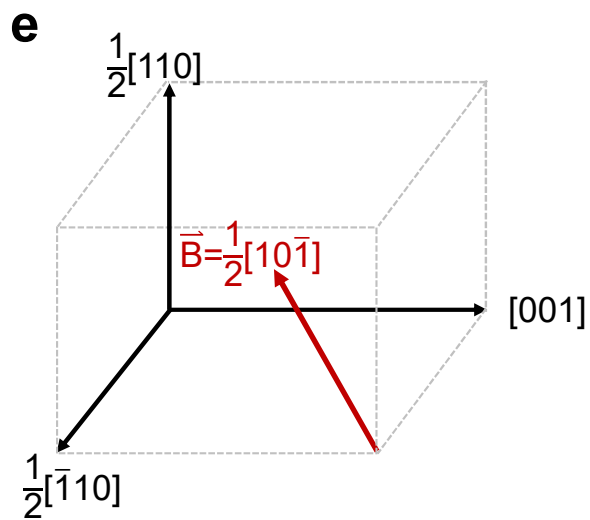
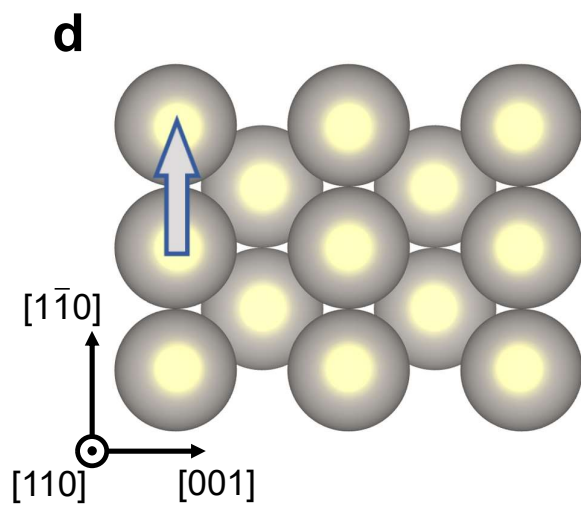
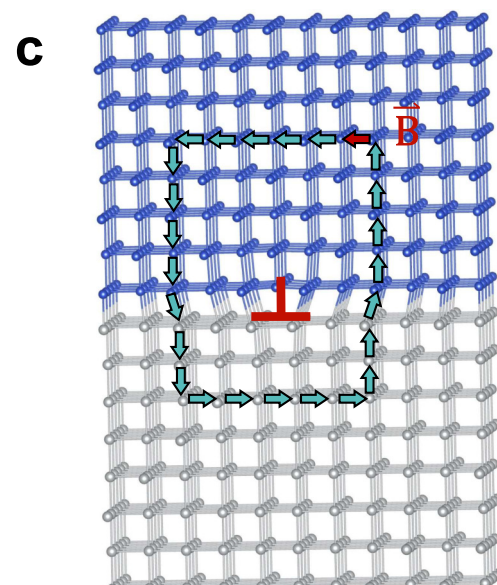
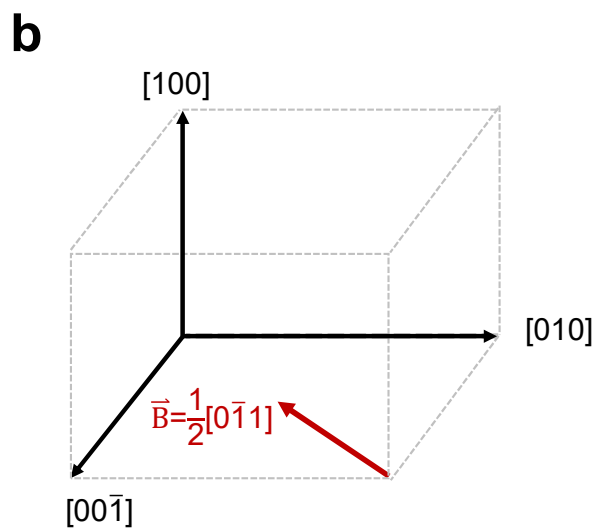
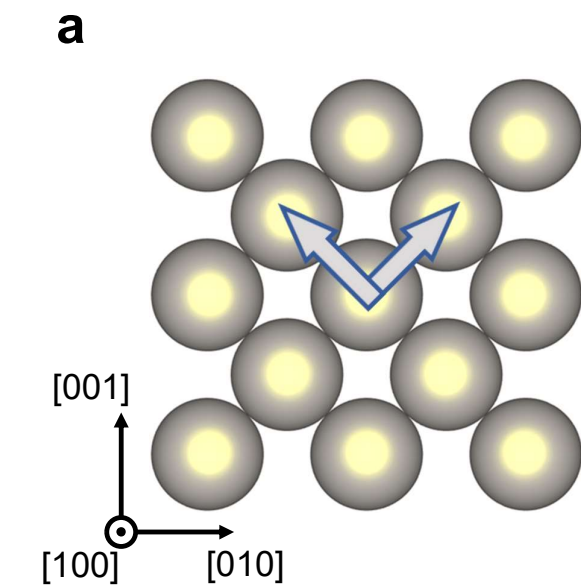
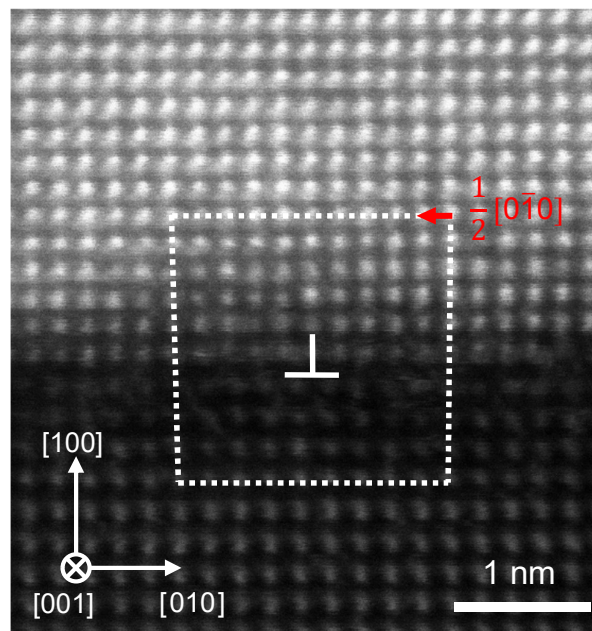
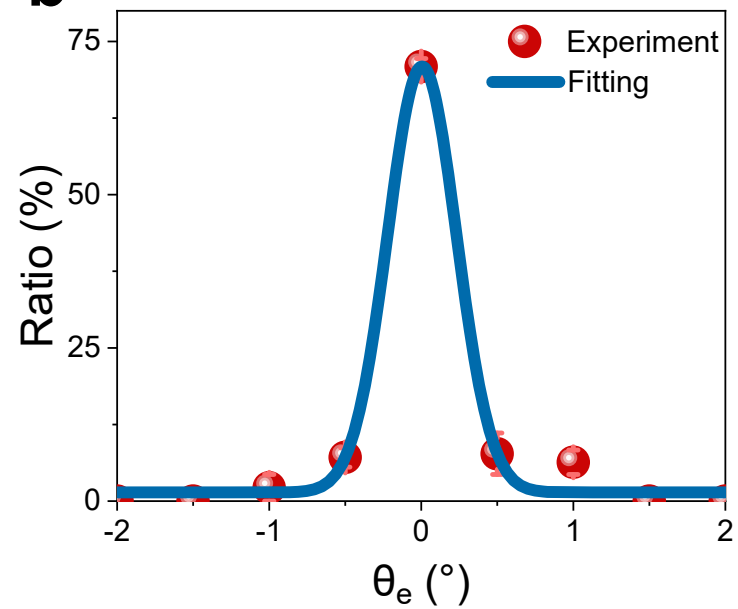
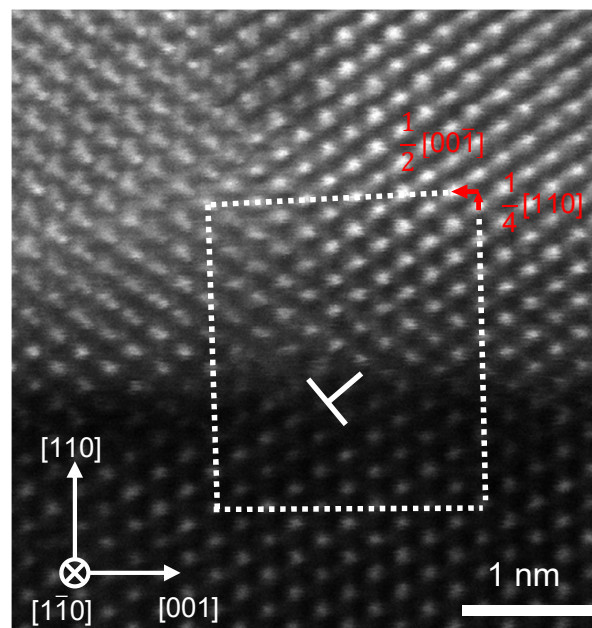


Fig. 4. Field-free current-induced switching of magnetization with perpendicular components. The schematic diagram of the set-up for SOT switching of perpendicular magnetization is shown in **a**, with small θ_M and **b**, with large θ_M . The yellow arrows indicate the ferromagnetic magnetization. The black arrows indicate the I_{pulse} . The results of field-free SOT switching with I_{pulse} (pulse width of 2 ms) applied along $[110]$ and $[001]$ are shown in **c**, for Pt(5)/Co(1.2)/NiO(20)/MgO heterostructure with $\theta_M = 16^\circ$ and **d**, for Pt(5)/Co(0.6)/Pt(1.4)/NiO(20)/MgO heterostructure with $\theta_M = 84^\circ$, respectively. Field-free SOT switching is only observed as I_{pulse} is applied along $[110]$. **e**, the schematic diagrams illustrate the switching of magnetization by SOT. The yellow arrow indicates the magnetization. The red arrow indicates the spin polarization σ provided by Pt as I_{pulse} is applied along $[110]$ direction. The black curve arrow indicates the direction of magnetization relaxation. **f**, reversible field-free SOT switching with $\pm J_{pulse}$ about 5.7×10^{11} A/m 2 and pulse width of 300 μs along $[110]$ in (110)-oriented Pt(5)/Co(0.6)/Pt(1.4)/NiO(20)/MgO heterostructure for more than 10^4 cycles. Values in parentheses are thickness in nm.



a**b****c****d**