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Supplementary Materials for

Large Dzyaloshinskii-Moriya interaction induced by chemisorbed oxygen on a ferromagnet surface

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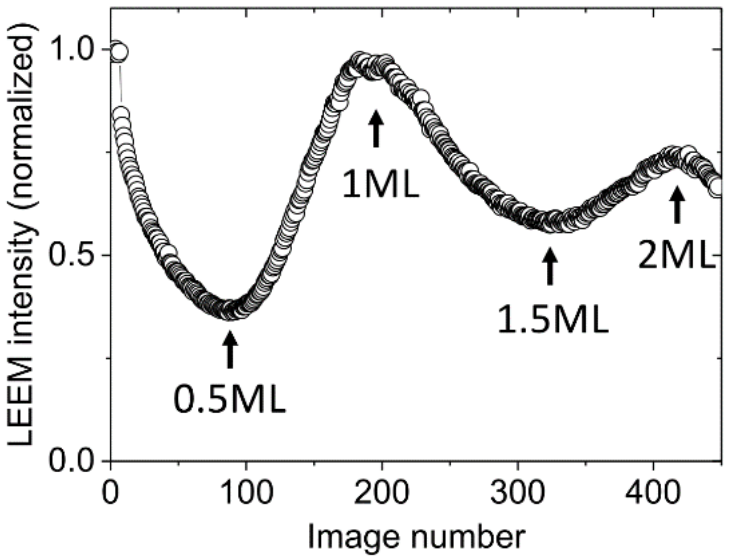

Fig. S1. Calibration of the Pd film thickness. Electron reflection intensity measured from low-energy electron microscopy images during Pd film growth, where the reflectivity oscillations associated with layer-by-layer growth allow precise control of the film thickness.

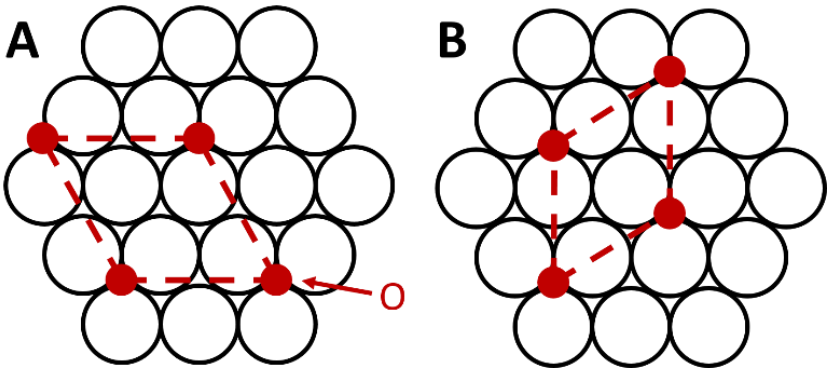

Fig. S2. Sketches of oxygen adsorption sites on Ni(111). (**A**) $p(2 \times 2)$ structure. Red solid circles are oxygen adatoms, black open circles are Ni(111) surface atoms. (**B**) $(\sqrt{3} \times \sqrt{3})R30^{\circ}$ structure. Oxygen atoms occupy threefold-hollow sites on Ni(111) for both cases.

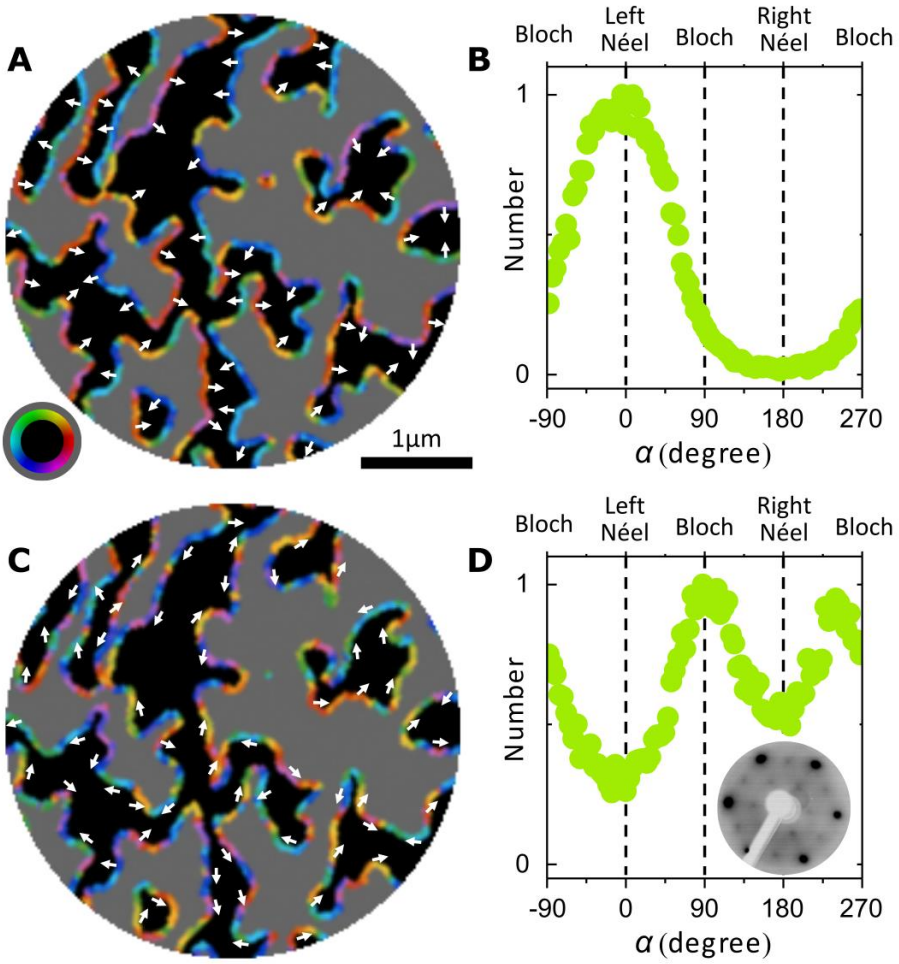

Fig. S3. Chemisorbed oxygen induced domain wall evolution in the $[Co/Ni]_3/Cu(111)$ system. (**A**) compound SPLEEM image of a $[Co_{1ML}/Ni_{2ML}]_3/Cu(111)$ multilayer. Scale bar is 1 μm. White arrows highlight the domain walls' in-plane magnetization directions. Color wheel indicates the magnetization orientation of the in-plane components within domain walls. Grey/black regions show out-of-plane components of the magnetization pointing down (-z) or up (+z). (**B**) Histogram of angle α derived from panel A, where the peak at 0° indicates a left-handed chiral Néel wall system. (**C**) compound SPLEEM image of a $[Co_{1ML}/Ni_{2ML}]_3/Cu(111)$ multilayer covered by 0.21 ML of chemisorbed oxygen. (**D**) Histogram of angle α derived from panel C, where double peaks near 90° and 270° indicate an achiral Bloch wall system. Insert: *p*(2x2) LEED pattern of the sample in panel C, which is comparable with the Ni surface. The energy of incident electrons is 95 eV.

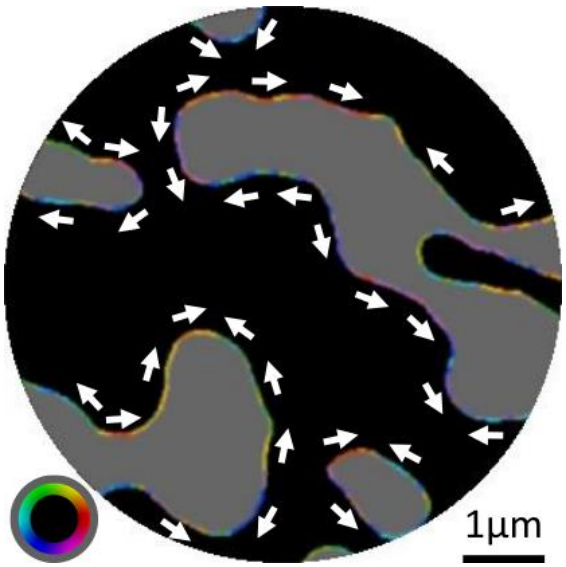

Fig. S4. SPLEEM image of [3Ni/1Co]$_4$/3Ni/2Co/3.46Pd/W(110). The in-plane magnetization of DWs is highlighted by the color wheel and white arrows, showing an achiral Bloch DW configuration. This indicates that the uniaxial magnetic anisotropy, and its effect on DWs, is suppressed.

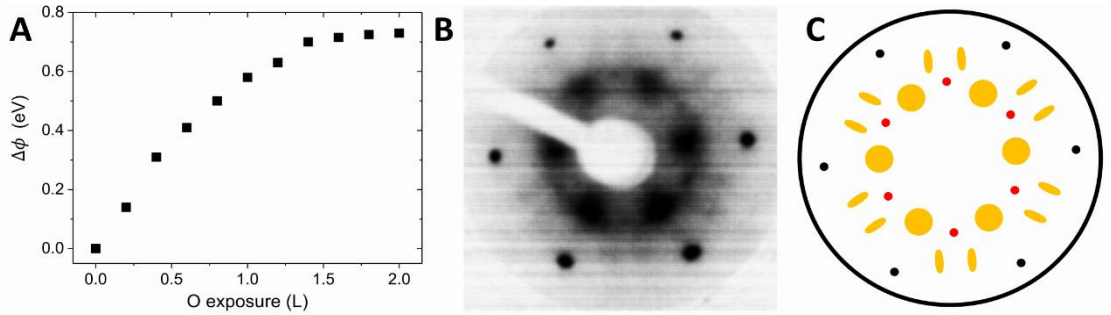

Fig. S5. Measurements of the oxygen coverage. (**A**) Chemisorbed oxygen induced work function change $\Delta\phi$ on the surface of 1ML Ni/3ML Co/2.5ML Pd/W(110), with the oxygen pressure kept at 1×10^{-8} torr. The initial work function of the system without oxygen is around 5.05 eV. (**B**) Low-energy electron diffraction pattern of chemisorbed oxygen at a total dose of 2 Langmuir's on the surface of 1ML Ni/3ML Co/2.5ML Pd/W(110), at an electron energy of 80 eV. The pattern is consistent with that of 0.29 ML oxygen on Ni(111) reported in ref. 36 in the main text. (**C**) Colorized patterns highlight the contribution from different phases: black dots – original (111) surface; orange patterns – *p*(2x2)-like complex phase at d_O=0.27-0.29 ML, red dots – $\left(\sqrt{3} \times \sqrt{3}\right)R30°$ phase.

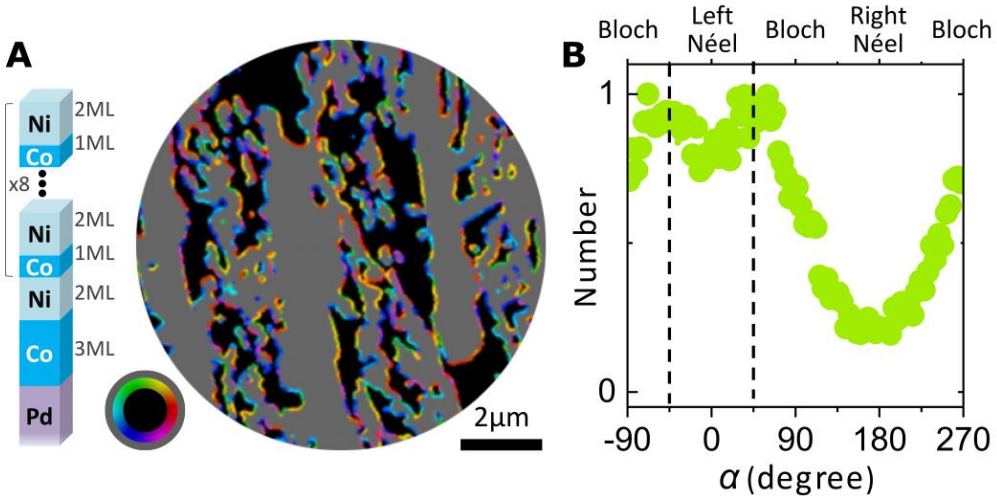

Fig. S6. Quantification of the DMI at the Co/Pd(111) interface. (**A**) (left) The sketch shows the structure of the $[Ni/Co]_9/Pd(111)$ multilayer, (right) compound SPLEEM image of the multilayer. Grey/black regions show out-of-plane components of the magnetization pointing down (-z)/up (+z). The color wheel indicates the magnetization orientation of the in-plane components within domain walls. (**B**) Histogram of angle α in $[Ni/Co]_9/Pd(111)$ multilayer, where double peaks at ±45° indicate that in this sample the dipolar force exceeds the DMI strength at the Co/Pd interface.