Mechanically tunable exchange coupling of Co/CoO bilayers on flexible muscovite substrate

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Figure S1. Temperature and pressure dependence of oxide-MBE deposition (a) XPS and (b) XRD spectra of films grown in varied oxygen pressures at 260 °C; (c) XPS and (d) XRD spectra of films grown at varied temperatures in oxygen pressure of $10^{-6}$ mbar.
**Figure S2.** The quality of CoO/mica films by checking the rocking-curve (a) CoO(111)/mica; (b) CoO(100)/mica

![Figure S2](image)

**Figure S3.** The Raman spectra of CoO films with/without bending.

The Raman spectra of (a) CoO (111); and (b) CoO (100) films on mica substrate were collected during bending-out with various stages. The radius was changed from 12.5 mm to 3.5 mm.

![Figure S3](image)
Figure S4. Temperature and thickness dependence of exchange coupling in Co/CoO (111). M-H loops at 50 K, 200 K and 300K of: (a) Co_{15nm}/CoO_{20nm}, (b) Co_{5nm}/CoO_{30nm} and (c) Co_{5nm}/CoO_{20nm}; (d) Extracted $H_{EB}$.

Figure S5. M-H loops of Co/CoO (100) film at different temperatures, different bending radii. (a) Illustration of the bending tests: samples were measured when non-bended, bended-in, bended-out and released; (b)-(f) M-H loops at different temperatures with varied bending states: no bend, bend in with radius of 3 mm, bend out with radius of 3 mm, bend in with radius of 2 mm, bend in with radius of 2 mm, respectively.
Figure S6. M-H loops of Co/CoO(111) film at different temperatures, different bending radii. (a)-(c) M-H loops at different temperatures with varied bending states: no bend, bend in with radius of 3 mm, bend out with radius of 3 mm, respectively.

Figure S7. Schematics of AMR measurements (a) Illustration of measurement setup: samples were bended by a cylinder, the applied field was perpendicular to the excitation current; (b) the geometry of measurements: the excitation current was along y-axis, samples were rotated along x-axis, and applied field was along z-axis.
**Figure S8.** (a) Schematics of conventional epitaxy and vdW epitaxy; (b) RHEED patterns of mica and CoO(111) film; (c) Real-time monitoring RHEED intensity of the growth; (d) Thickness dependence of XAS for CoO(111) films on mica, in comparisons with CoO bulk.

### Computational details

First-principles calculations were carried out with the Vienna *ab initio* Simulation Package (VASP)\(^1\) by using the projector-augmented wave (PAW) method\(^2\) and the generalized gradient approximation (GGA). The exchange-correlation potential is adopted in the PBE\(^3\) (Perdew-Burke-Ernzerh) form of GGA+U\(^4\) method with \(U=7.1\) eV and \(J=1\) eV for cobalt 3\(d\) electrons. Both structural relaxation and self-consistent calculations were carried out with the tetrahedral method with Blöchl corrections\(^5\), and the energy cut-off is set to 500 eV. We fully optimize each ionic position until the residual forces converged less than 0.001 eV/Å and self-consistent convergence for electronic energy is 10\(^{-6}\) eV. A 3×3×3 and 7×7×2 Monkhorst-Pack \(k\)-point mesh are adopted for CoO (100) and CoO (111) calculations, respectively.
The calculated lattice constant of 4.261 Å is very close to experimental lattice parameter. We use a 2×2×2 supercell to simulate the type-II AFM structure of CoO (100). And for CoO (111), the bulk structure restructured along [111] direction. Figure S9 shows the atomic structures of CoO (100) and CoO (111). In consideration of applying strain, the lattice parameter of ab-plane is artificially altered. Then we newly relax the c/a of the structures that are used to calculate the MAE.

**Figure S9.** The atomic structure of (a) CoO (100) and (b) CoO (111). Bule and red spheres represent Co and O atoms, respectively.

The main origin of the magnetic anisotropy is the spin-orbit coupling (SOC)⁶. In this work, the calculations about MAE include two steps: first, the charge density is obtained through self-consistent calculations without the spin-orbital coupling. Then, we calculate the total energy for different magnetization axes, for which we use the same charge density and include the spin-orbital coupling.

For CoO without stress, the easy axis is along [001] (or [100], [010]) direction. When the (001) strain is applied, however, the easy axis will be canted away from [001] direction. As shown in Table 1, the magnetic anisotropy energy \( E_{[001]-[011]} \) changes from negative (strain free) to positive (0.5% strain), suggesting the canting of the easy axis. However, when a 0.5% strain is applied in the (111) plane, the MAE remains negative, meaning that the easy axis is unchanged (Table 1). These calculated results are in nice agreement with the experiments.

**Table 1.** The calculated magnetic anisotropy energies (MAE, in units of meV) of CoO under (100) and (111) strain. The MAE is calculated as \( E_{[001]-[011]} \).

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<thead>
<tr>
<th></th>
<th>(001)</th>
<th>(111)</th>
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<tr>
<td>0%</td>
<td>-7.08</td>
<td>-7.08</td>
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<tr>
<td>0.5%</td>
<td>5.01</td>
<td>-6.66</td>
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References