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Supporting Information

First-principles study of the magnetic exchange forces between the RuO_2(110) surface and Fe tip

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Supporting Information

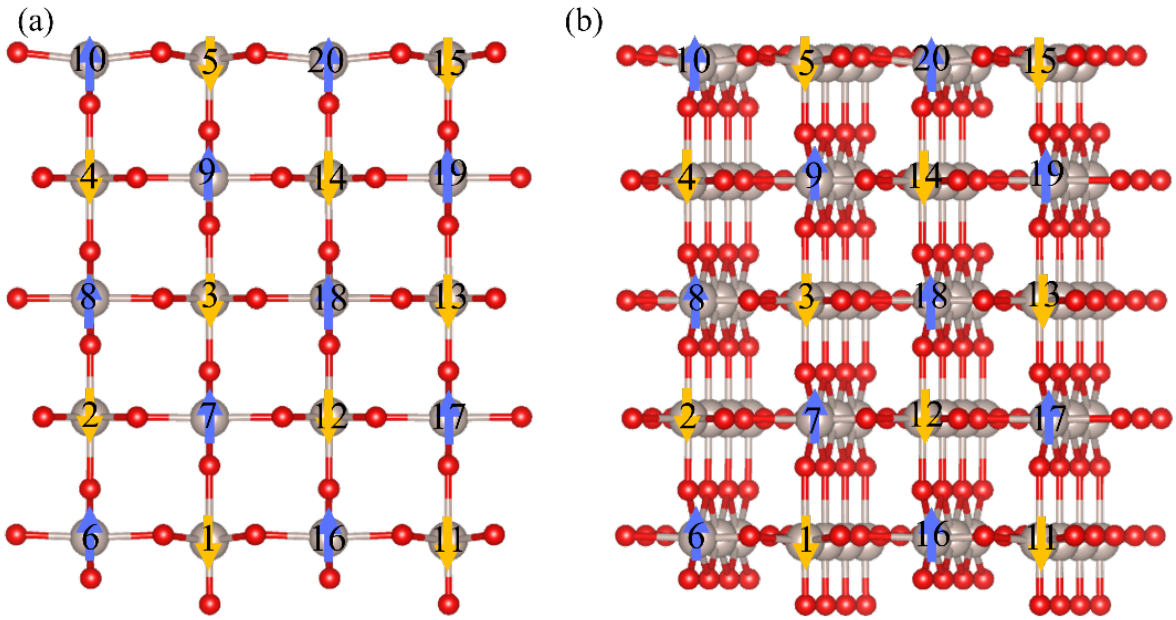

Figure S1. Side and rotated views of a five-layer RuO_2 slab with a (110) surface at the top. Such a clean (110) surface can be obtained under ultra-high vacuum conditions. The AFM ordering of the magnetic moments on the Ru atoms is indicated by the orange and blue arrows. The atom labels correspond to those in Table S1.

Table S1. Magnetic moments on the Ru atoms of the five-layer RuO_2 slab shown in Figure S1. Positive/negative numbers correspond to spin-up/spin-down.

Ru atoms	Magnetic moment (μ_B)
1-5	−0.24, −1.17, −1.20, −0.95, −0.91
6-10	1.46, 1.14, 1.17, 1.11, 1.14
11-15	−0.24, −1.16, −1.20, −0.94, −0.92
16-20	1.46, 1.14, 1.16, 1.11, 1.14

Magnetic moments as function of the tip-surface distance

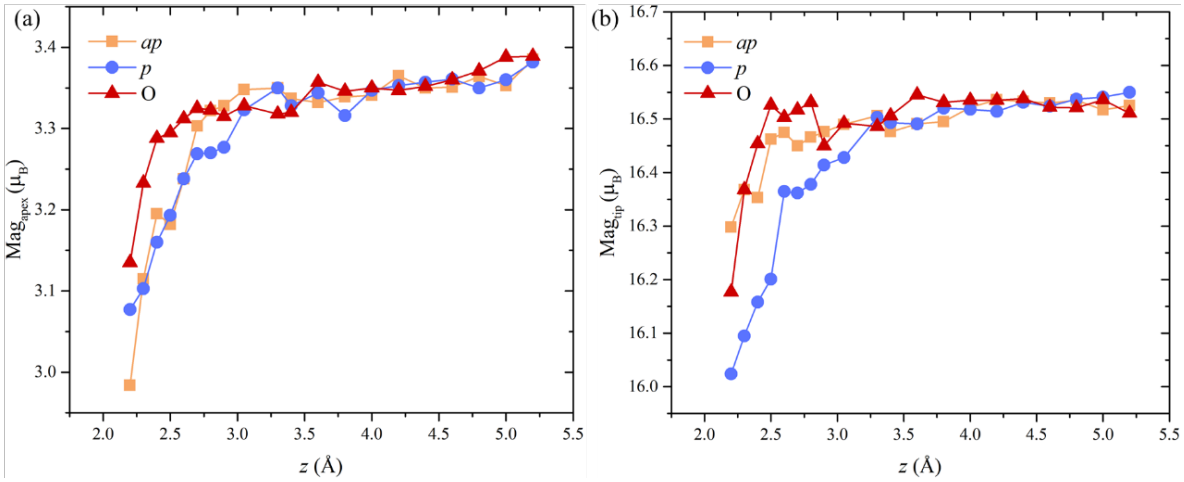

Figure S2. The magnetic moments of (a) the Fe tip apex atom and the (b) whole Fe tip cluster with the tip positioned over different atoms of the RuO_2(110) substrate, as a function of the tip-surface distance *z*.

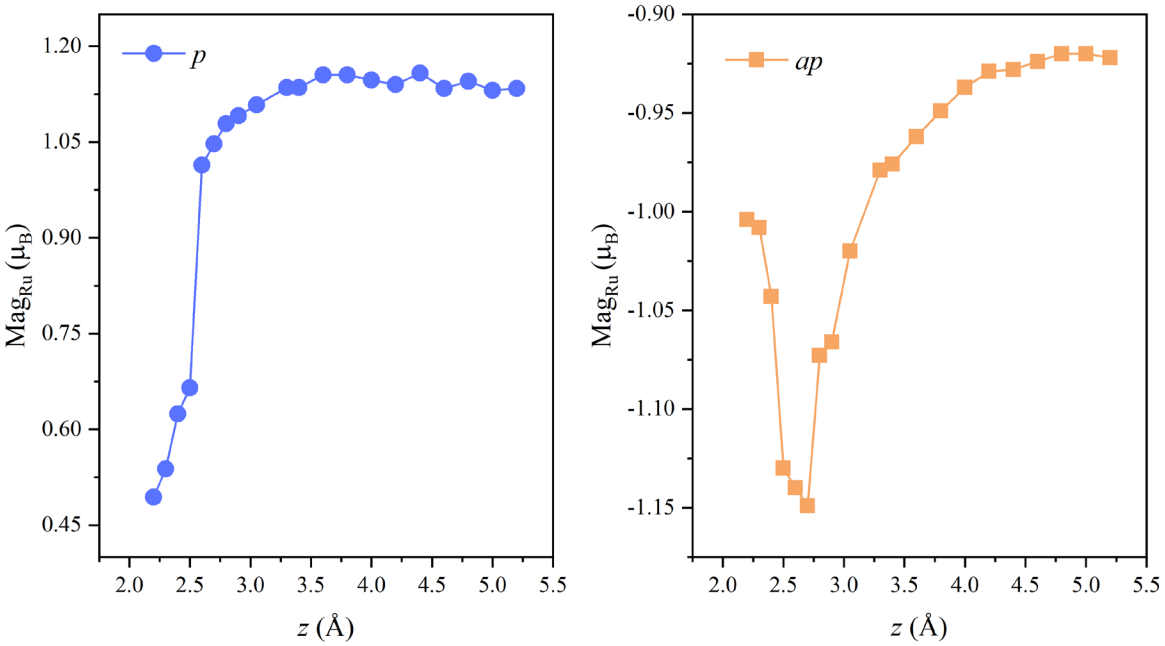

Figure S3. The magnetic moments of (a) the Ru↑ atom and the (b) Ru↓ atom, as a function of the tip-surface distance *z*.

Tip-surface interaction

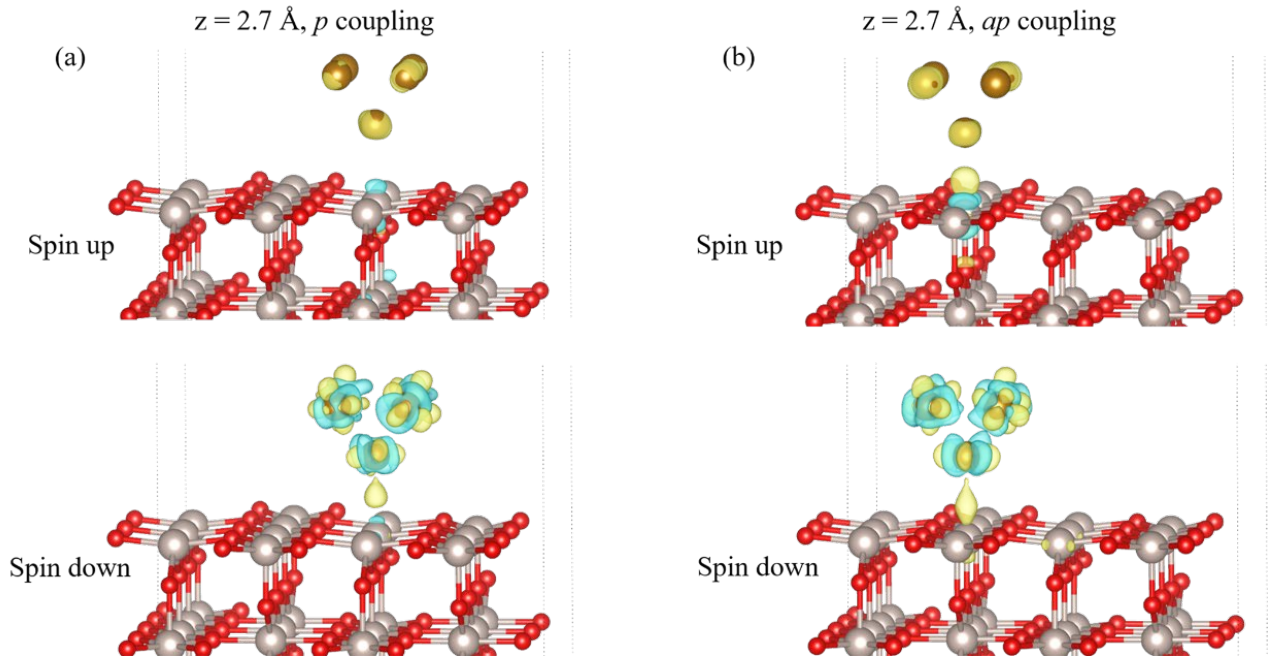

Figure S4. Changes of the spin densities $\Delta\rho^{\sigma}$ at z = 2.7 Å for (a) *p* and (b) *ap* systems. Positive values (yellow) indicate an accumulation and negative values (cyan) a depletion of spin density.

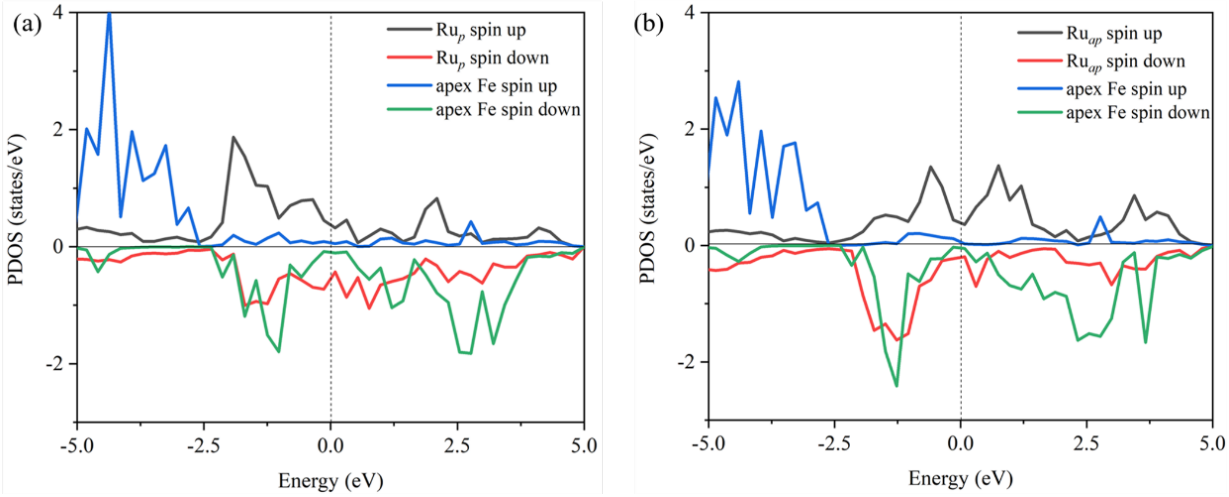

Figure S5. Spin-polarized projected density of states (PDOS) of (a) the Fe apex atom and Ru↑ atom (*p* coupling), and (b) the Fe apex atom and Ru↓ atom (*ap* coupling).

Spin-resolved charge density differences are plotted Figure S4, for a tip-surface distance z = 2.7 Å. The tip-surface interaction affects the spin-down (minority) channel of the Fe tip most, which is not surprising as the frontier orbitals, i.e., the Fe d-states with energies closest to the Fermi level, have minority spin character, see Figure S5. Overall, there is some depletion of spin-up electrons on the Ru↑ atom, and some accumulation of spin-down electrons on the Ru↓ atom, which is consistent with the change in magnetic moments, Figure S3. That would be compatible with the formation of a Heitler-London like bond (valence bond) involving the spin-down electrons on the tip and the spin-up electrons on the Ru atom. That bond then would be strongest for the Ru↑ atom, which then leads to the total energy being lower for the *p* coupling.

Difference between the two surface Ru atoms

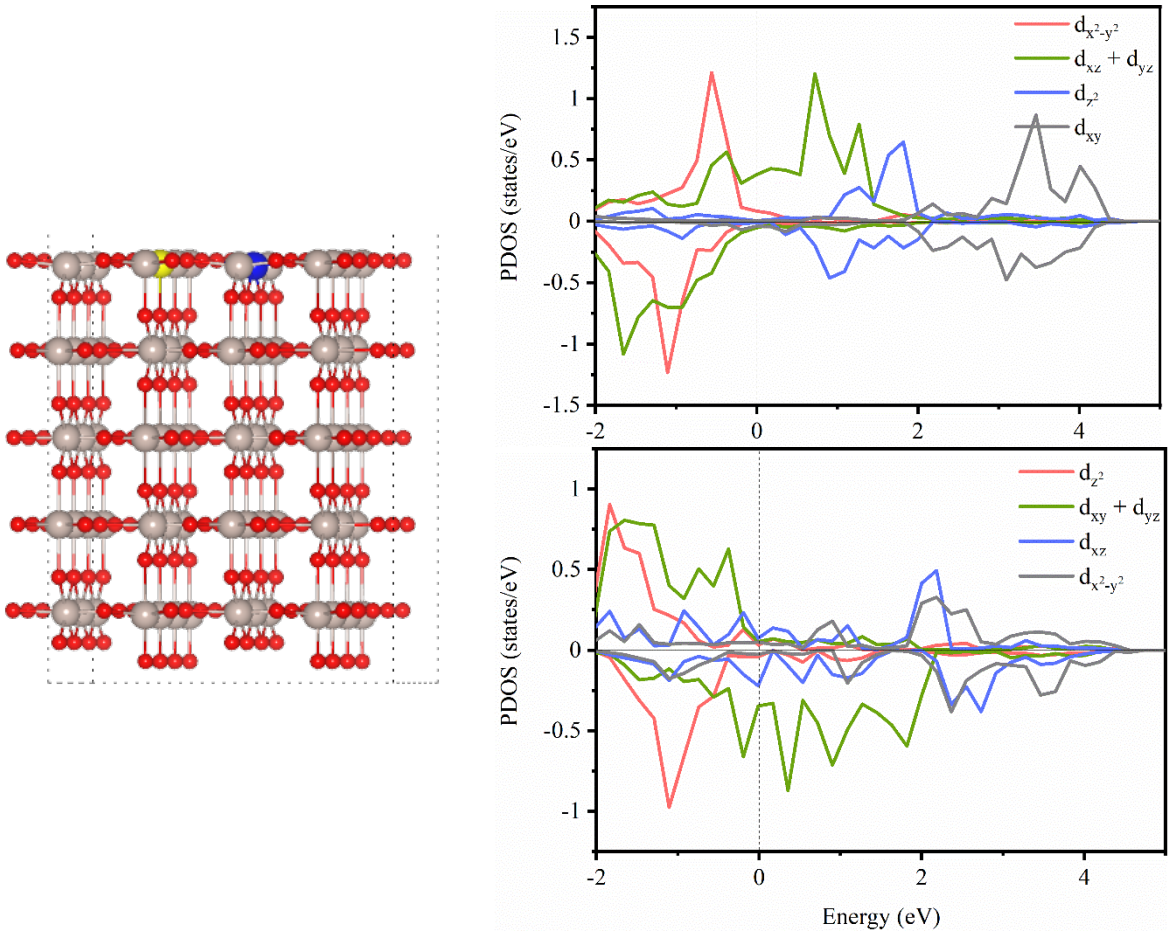

Figure S6. Left: side view of the RuO_2(110) surface without the Fe tip. The surface Ru↓ atom is highlighted in yellow and the Ru↑ atom is highlighted in blue. Right top: projected density of states (PDOS) of the Ru↓ atom. Right bottom: PDOS of the Ru↑ atom.

The two Ru atoms in the (110) surface have a different coordination by oxygen atoms, the Ru↓ atom is fivefold coordinated, whereas the Ru↑ atom is fourfold coordinated, see Figure S6. The magnetic moment on these two atoms is quite similar (apart from the sign), 0.91 versus 1.14 μ_B, see Figure S1 and Table S1, indicating that their electronic structure is quite similar. The chemical similarity of the two Ru atoms in the (110) surface can be illustrated by projecting the density of states on the two atoms. Whereas the PDOS of a bulk Ru atom can be assigned to Ru^{4+} with d^4 configuration, see ref. 12 in the main text, the PDOS of a surface Ru more resembles Ru^{3+} with d^5 configuration, see the Figure S6. However, apart from a relabeling of local axes, and a reversal of spin directions, the PDOSs of the occupied states of the two surface Ru atoms are quite similar.