

Supplementary Information

Electrical switching of spin-polarized current in multiferroic tunneling junctions

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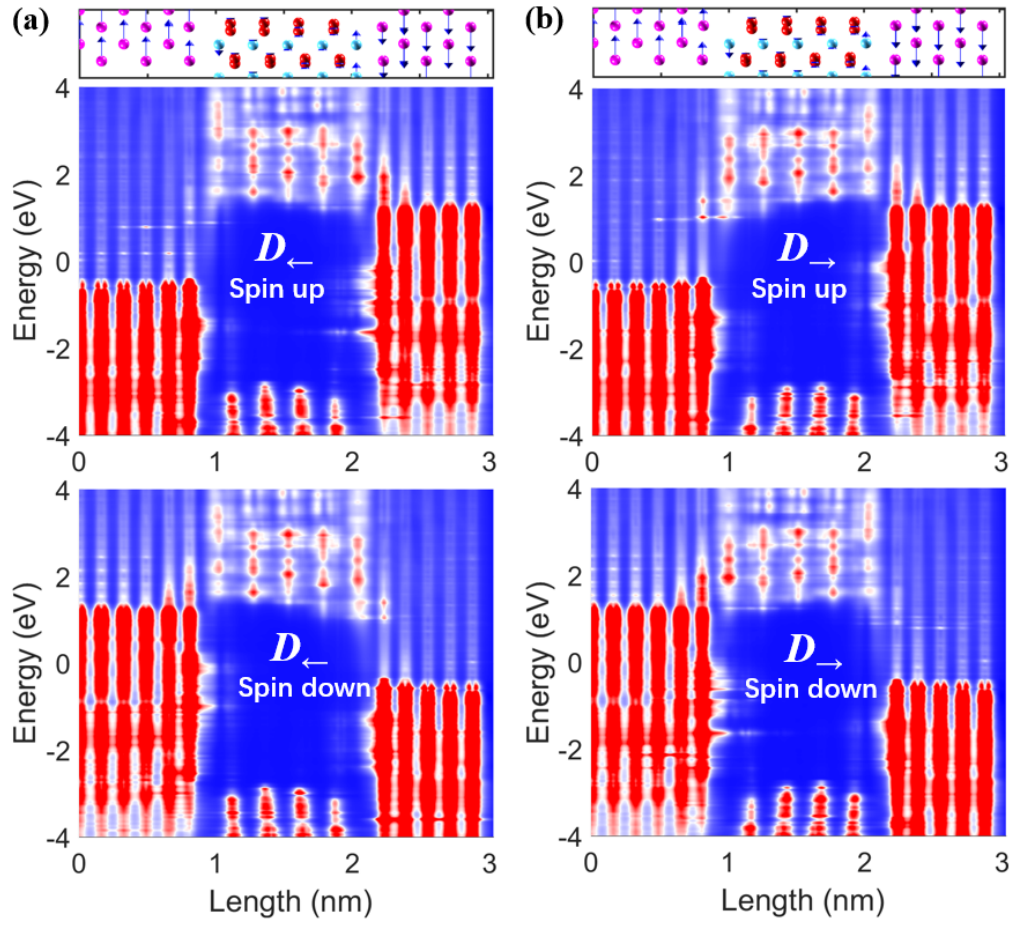
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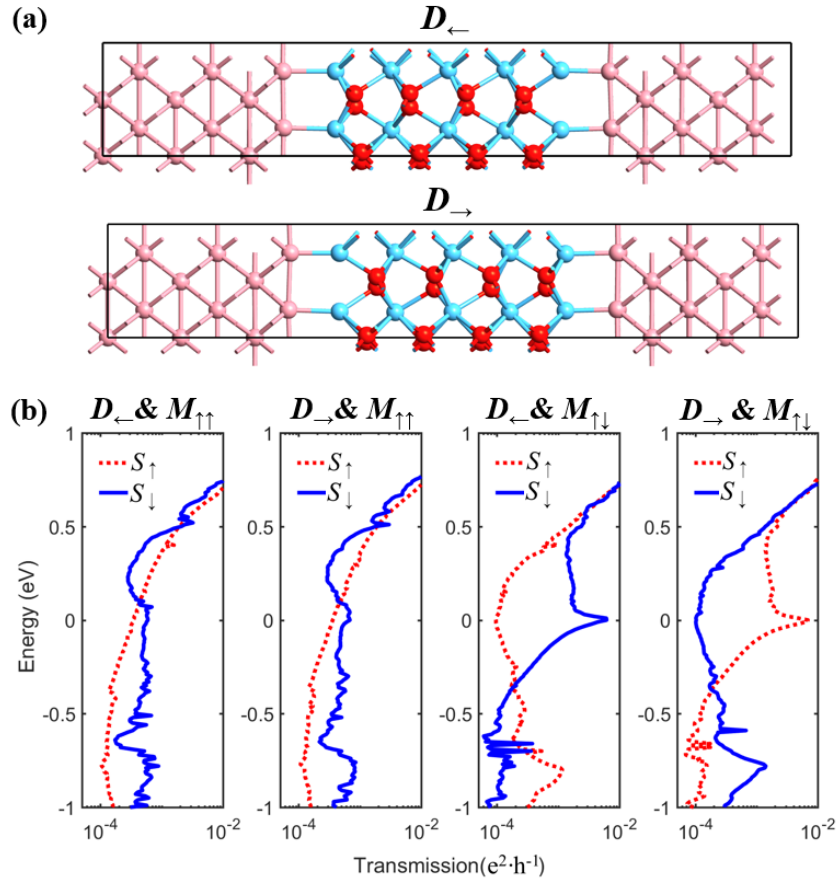
Supplementary Discussion

Supplementary Figure 1 shows local spin DOS of Co/HfO₂/Co junctions at antiparallel magnetic states. It is observed that there is metallization of the FE tunnel barrier at both interfaces of the Co/HfO₂/Co junction and an obvious magnetoelectric coupling effect at interfaces, which can be modulated by ferroelectric switching. Because the two leads are set at antiparallel magnetic states, there are different spin-polarized states of Hf atoms at interfaces. As a result, there are different spin tunnel barriers that play a role in changing the spin polarization. With the reversal of ferroelectric polarization, spin polarization orientation can be switched due to symmetric interfaces of Co/HfO₂/Co junctions.

In order to investigate the possibility to enhance the magnetoelectric coupling effect at interfaces, we constructed Co/HfO₂/Co junctions with Hf atoms on the top sites of Co atoms as shown in Supplementary Figure 2 (a). The optimized distances between Co and Hf atoms are 2.33 Å and 2.21 Å for the left and right interfaces of Co/HfO₂/Co junctions at $D_{\leftarrow}$ in Supplementary Figure 2 (a). The magnetic moments of Hf atoms at the left and right interface are 0.24 μ_B and 0.39 μ_B , respectively. Spin-dependent transmissions are plotted in Supplementary Figure 2 (b). It can be found that the spin polarization ratio of junctions with two leads at antiparallel states gets larger than that in Fig. 3, which is -94% at $D_{\leftarrow}$ and 94% at $D_{\rightarrow}$.



Supplementary Figure 1. Local magnetic moments and local spin density of states. Local magnetic moments and local spin density of states along Co/HfO₂/Co junctions with Hf-terminated interfaces at an antiparallel state: (a) left $D_{\leftarrow}$ and (b) right $D_{\rightarrow}$ oxygen displacements.



Supplementary Figure 2. Atomistic structures and transmissions of the Co/HfO₂/Co junction. (a) Atomistic structures and (b) transmissions of the Co/HfO₂/Co junction with Hf atoms on top of Co atoms.