## Supplementary Information

## Molecular spinterface in $\mathbf{F}_{4}$ TCNQ-doped polymer spin valves

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Fig. S1 Band alignment diagram of LSMO/P3HT/F4TCNQ/Co.


Fig. S2 Optical microscope image of $\mathrm{F}_{4}$ TCNQ-doped P3HT layer on LSMO/STO substrate. In the figure, the organic layer is relatively smooth, and the small granules are $\mathrm{F}_{4} \mathrm{TCNQ}$.


Fig. S3 MR curve of the LSMO/P3HT/Co/Au device before doping. The temperature was 2 K , and the input current was $0.03 \mu \mathrm{~A}$.


Fig. S4 AFM images of P3HT before and after $\mathrm{F}_{4}$ TCNQ doping. (a), (d) The morphology and thickness of the pristine P3HT layer on the bottom ITO electrode. (b), (e) The morphology and thickness of $\mathrm{F}_{4}$ TCNQ-doped P3HT on the bottom ITO electrode. (c), (f) The morphology and thickness of $\mathrm{F}_{4} \mathrm{TCNQ}$-doped P3HT on the bottom LSMO electrode.


Fig. S5 MR curves of the doped ITO/P3HT/F4TCNQ/Co/Au device at 5 K . Each measurement was conducted with different currents of (a) $0.01 \mu \mathrm{~A}$, (b) $0.02 \mu \mathrm{~A}$, (c) $0.03 \mu \mathrm{~A}$. and (d) 0.05 $\mu \mathrm{A}$.


Fig. S6 MR curves of the doped ITO/P3HT/F4TCNQ/Co/Au device with the input current of $0.01 \mu \mathrm{~A}$. Each measurement was conducted at different temperatures of (a) 2 K , (b) 10 K , (c) 20 K , and (d) 30 K .

