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Electronic Supplementary Information

Revealing the origin of magnetoresistance in unipolar organic field-effect transistors

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Figure S1. MR effects observed in four identical Spiro-TAD field-effect transistors (labeled as #1, #2, #3 and #4) fabricated on bare SiO₂ and measured at $V_d = V_g = -40$ V under influence of a magnetic field of 96 mT and at room temperature.



Figure S2. MR effects of a Spiro-TAD field-effect transistor fabricated on HMDS-treated SiO₂ gate dielectric and measured at (a) $V_d = V_g = -80$ V and (b) $V_d = V_g = -50$, each at a magnetic field of 96 mT and at room temperature.



Figure S3. Transfer curves of F6-TNAP (x nm)/Spiro-TAD (40 nm) field-effect transistors fabricated on bare SiO_2 and recorded at different V_g sweep rate. Here, x is 1 nm, 5 nm, 10 nm and 15 nm, respectively.



Figure S4. Bias stress effect of F6-TNAP (x nm)/Spiro-TAD (40 nm) field-effect transistors fabricated on bare SiO₂ and measured in the devices with different thickness of F6-TNAP. In this case, x is 1 nm, 5 nm, 10 nm and 15 nm, respectively. The relative drain current I_d/I_0 was measured at $V_d = V_g = -30$ V.



Figure S5. Output curves of F6-TNAP/Spiro-TAD (40 nm) field-effect transistor fabricated on bare SiO₂ and with different thickness of F6-TNAP. (a) F6-TNAP = 1 nm, (b) F6-TNAP = 5 nm, (c) F6-TNAP = 10 nm and (d) F6-TNAP = 15 nm.



Figure S6. Transfer curves of F6-TNAP/Spiro-TAD (40 nm) field-effect transistor fabricated on bare SiO_2 and with different thickness of F6-TNAP. (a) F6-TNAP = 1 nm, (b) F6-TNAP = 5 nm, (c) F6-TNAP = 10 nm and (d) F6-TNAP = 1 nm.



Figure S7. (a) Cyclic voltammogram of Spiro-TAD measured in $CH_2Cl_2/0.1$ M TBAHFP. It exhibits half-wave potentials of the oxidation at -0.23 V, -0.38 V and -0.58 V vs. ferrocene/ferrocenium (Fc/Fc⁺), respectively. (b) In-situ UV-Vis-NIR spectroelectrochemical measurement of Spiro-TAD in MeCN/0.1 M TBAHFP at room temperature.



Figure S8. In-situ UV-Vis-NIR absorption spectra of Spiro-TAD (concentration 31 μ M) measured in CH₂Cl₂ and with additional concentration of organic molecular *p*-dopant F6-TNAP.



Figure S9. (a) Transfer characteristics and (b) square roots of absolute drain currents of a bilayer of F6-TNAP (5 nm)/Spiro-TAD (40 nm) field-effect transistor fabricated on bare SiO₂ measured at $V_d = -30$ V, without and under a magnetic field of 96 mT.