

Electronic Supplementary Information

Magnetoresistive Field-Effect Transistors Based on Organic Donor/Acceptor Blends

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EXPERIMENT

Bottom-contact field-effect transistor substrates were purchased from Fraunhofer IPMS (Dresden, Germany) with channel lengths (L) between 2.5 and 20 μm and channel width (W) of 10 mm. The isolation layer consists of 230 ± 10 nm thick SiO_2 and the source and drain electrodes are 30 nm Au with 10 nm ITO as adhesion layer. For all experiments transistors were used with $L = 2.5 \mu\text{m}$ and $W = 10\text{mm}$ which are measured in a glove box ($\text{O}_2, \text{H}_2\text{O} < 0.1$ ppm) at room temperature. Spiro-TTB and HAT-CN were synthesized and purified in our laboratory. Prior to the deposition of Spiro-TTB and HAT-CN, the predefined substrates were cleaned with acetone, 2-propanol and deionized water, followed by oxygen-plasma treatment and exposure to hexamethyldisilazane to replace the natural hydroxyl end group of SiO_2 with an apolar methoxy group. Finally, Spiro-TTB and HAT-CN were

deposited by thermal evaporation at a base pressure of 1×10^{-7} Torr ($T_{\text{substrate}} = 298$ K) with a thickness of 40 nm. For mixed system the composition was controlled by using different evaporation rates for Spiro-TTB and HAT-CN, respectively. The evaporation rates were monitored by two independent oscillating quartz-sensors. The uncertainty of our deposition process is $\pm 2.5\%$. From the vacuum chamber the samples were directly transferred to a glove box ($O_2, H_2O < 0.1$ ppm) and placed in a homebuilt sample holder. This sample holder was placed between the poles of an (unshielded) electromagnet with the magnetic field being perpendicular to the direction of the current flow in OFETs. The magnetic field was varied between -85 mT and $+85$ mT. Due to the (unshielded) earth magnetic field, reminiscence effects of the electromagnet and the uncertainty of our magnetic-field sensor, the uncertainty of our magnetic-field strength is $\pm 80\ \mu\text{T}$. Current-voltage measurements were performed by using a Keithley 4200 semiconductor characterization system equipped with preamplifiers for improving low-current measurements. All measurements were performed at room temperature (≈ 298 K).

Table S1. Field-effect mobility μ , on/off ratio, OFET behavior and magnetoresistance MR of Spiro-TTB, HAT-CN and various blending ratios of both materials.

S-TTB/HAT-CN [%]	μ / $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$	on/off ratio	OFET behavior	MR
100 / 0	$\mu_h = 3.2 \times 10^{-4}$	1×10^5	✓	✗
78 / 22	n.d	~3	✗	✓
50 / 50	n.d	~1	✗	✓
20 / 80	n.d	~1	✗	✓
0 / 100	$\mu_e = 5.6 \times 10^{-5}$	2×10^5	✓	✗

n.d = cannot be determined.

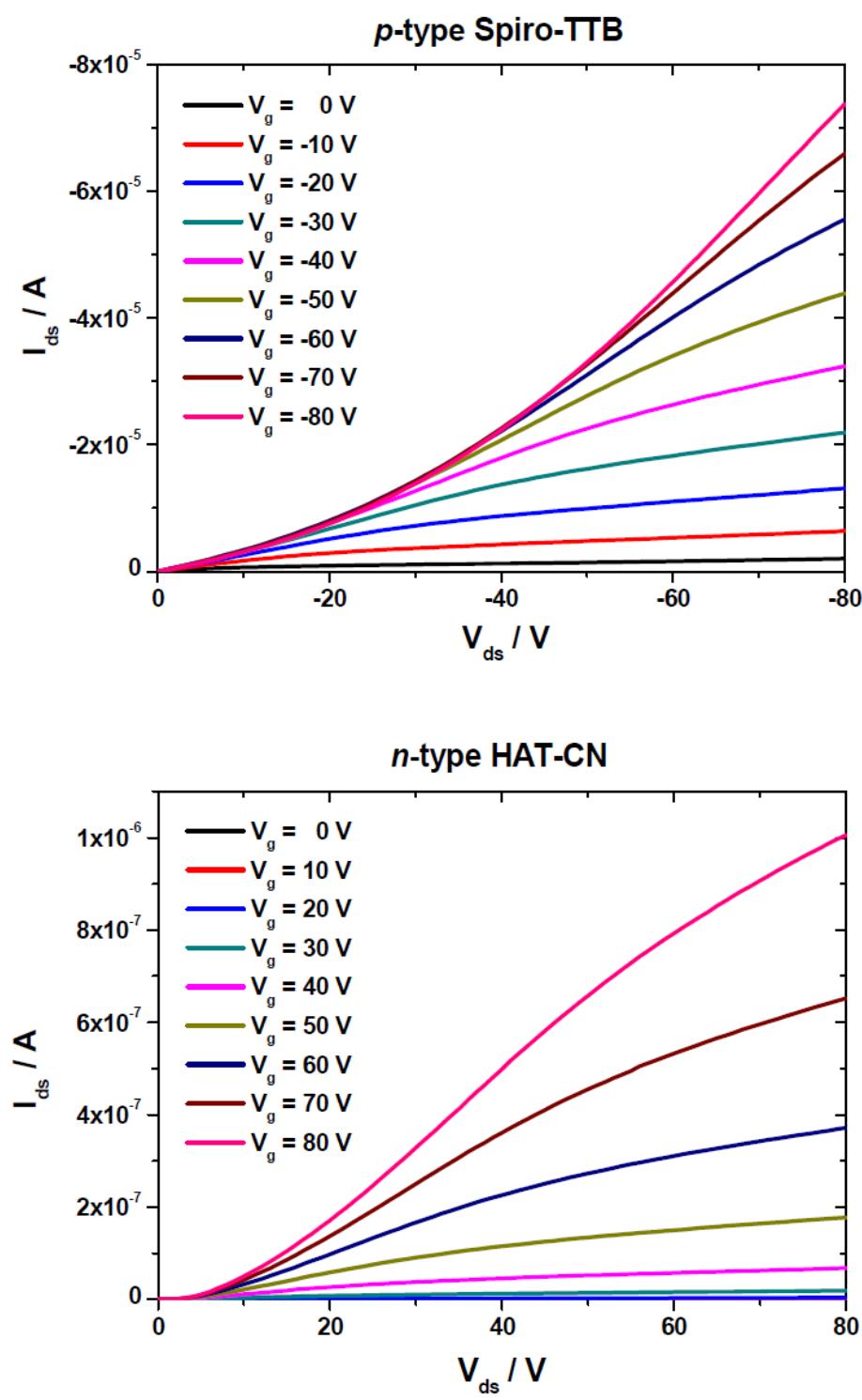


Figure S1. Output characteristics of pristine Spiro-TTB and pristine HAT-CN organic field-effect transistors.

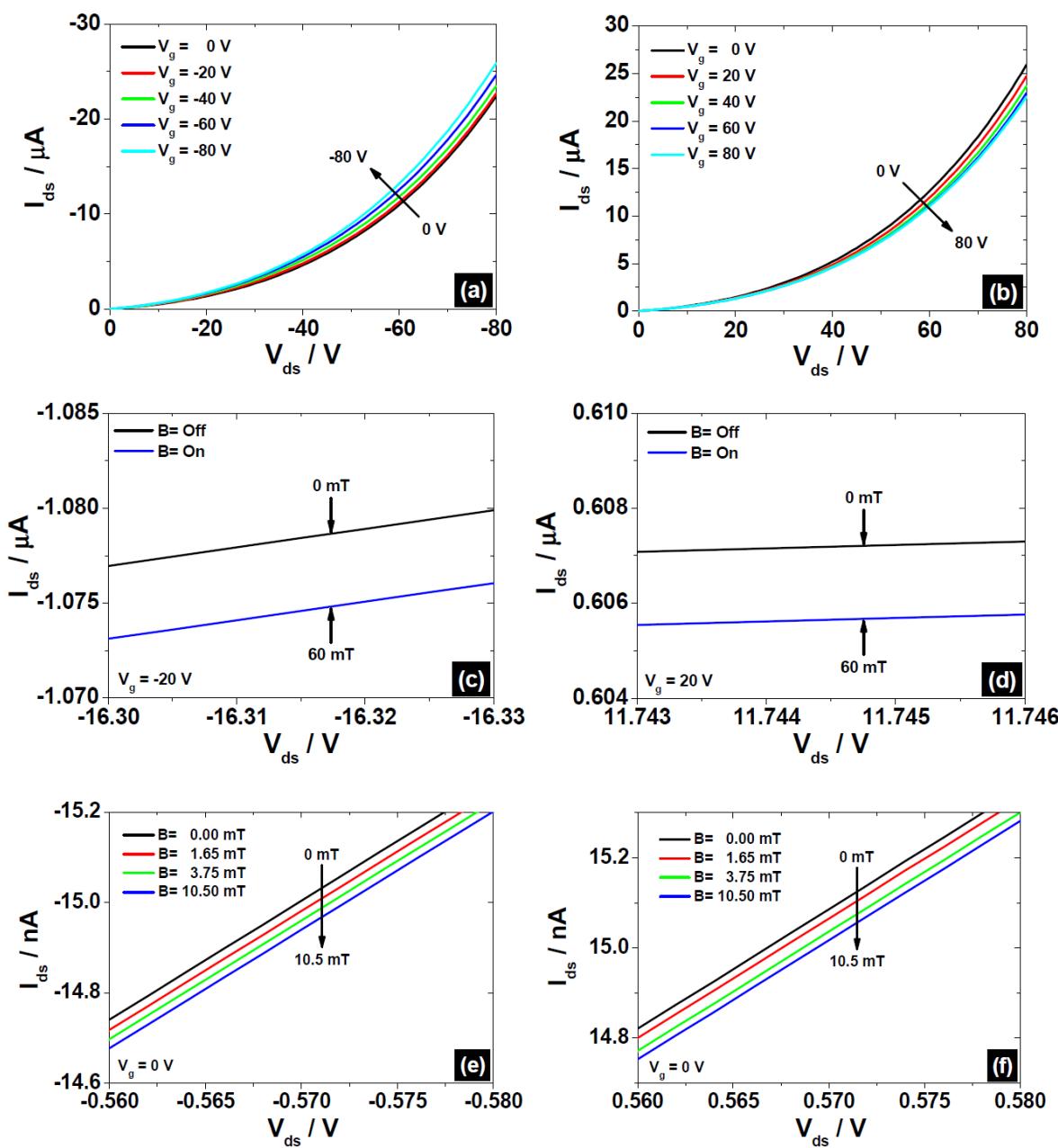


Figure S2. I_{ds} - V_{ds} curves of devices based on evenly blended Spiro-TTB/HAT-CN devices for *p*-channel on the left (a, c, e) and *n*-channel on the right side (b, d, f). I_{ds} - V_{ds} -curves are shown with V_g being swept from (a) 0 V to -80 V (*p*-channel) and (b) 0 V to 80 V (*n*-channel) without an external magnetic field. The magnetoresponse of the I_{ds} - V_{ds} -measurements for low magnetic fields of 60 mT (c, d) and very low magnetic fields below 11 mT (e, f) are depicted as well. All measurements show very high reproducibility.

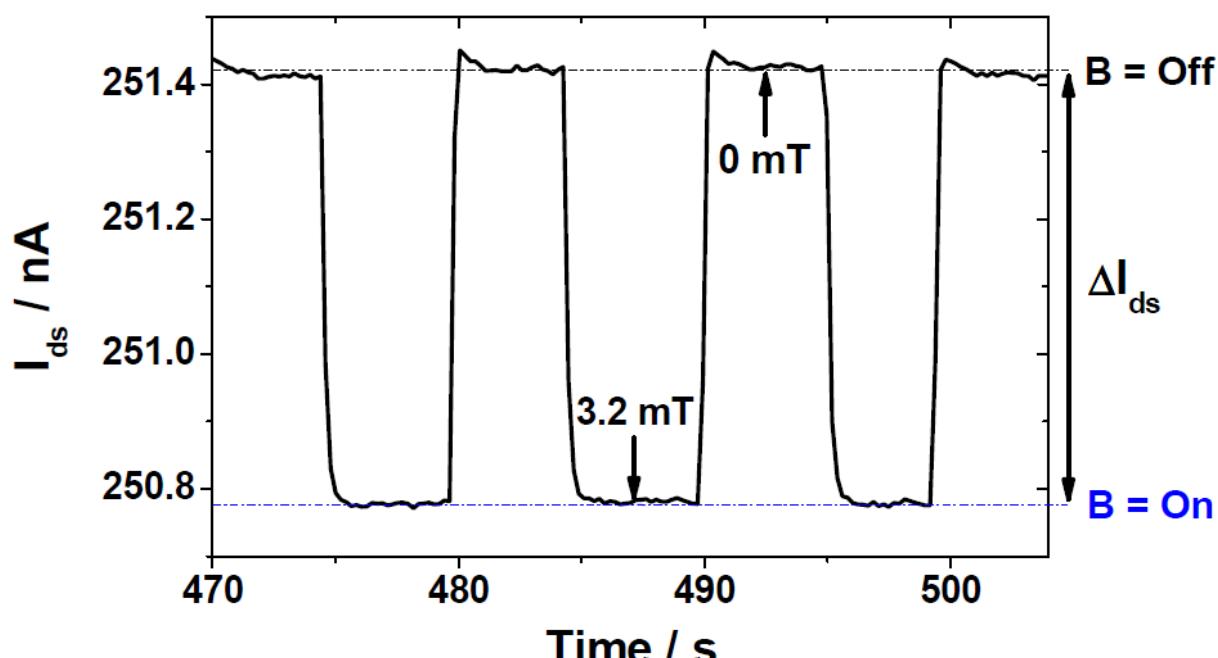


Figure S3. Determination of the MR values. With constant voltage conditions ($V_{ds} = \text{const.}; V_g = \text{const.}$) the magnetic-field dependency of I_{ds} is recorded and analyzed. During the I_{ds} -time measurement an external magnetic-field is switched on and off three times in a row and I_{ds} -time curves like the shown one is obtained. The magnetic-field induces a current decrease ΔI_{ds} , which is defined as $\Delta I_{ds} = I_{ds}(B) - I_{ds}(0)$, where $I_{ds}(B)$ is the current with and $I_{ds}(0)$ the current without an external magnetic-field, respectively. The MR values are calculated with $\text{MR} = \{[R(B) - R(0)]/R(0)\} \times 100\%$. $R(B)$ and $R(0)$ are resistances with and without an external magnetic-field, and are defined as $V_{ds}/I_{ds}(B)$ and $V_{ds}/I_{ds}(0)$. With constant V_{ds} the magnetoresistance therefore is determined by $I_{ds}(B)$ and $I_{ds}(0)$. The dash dotted lines are used for guiding the eyes.

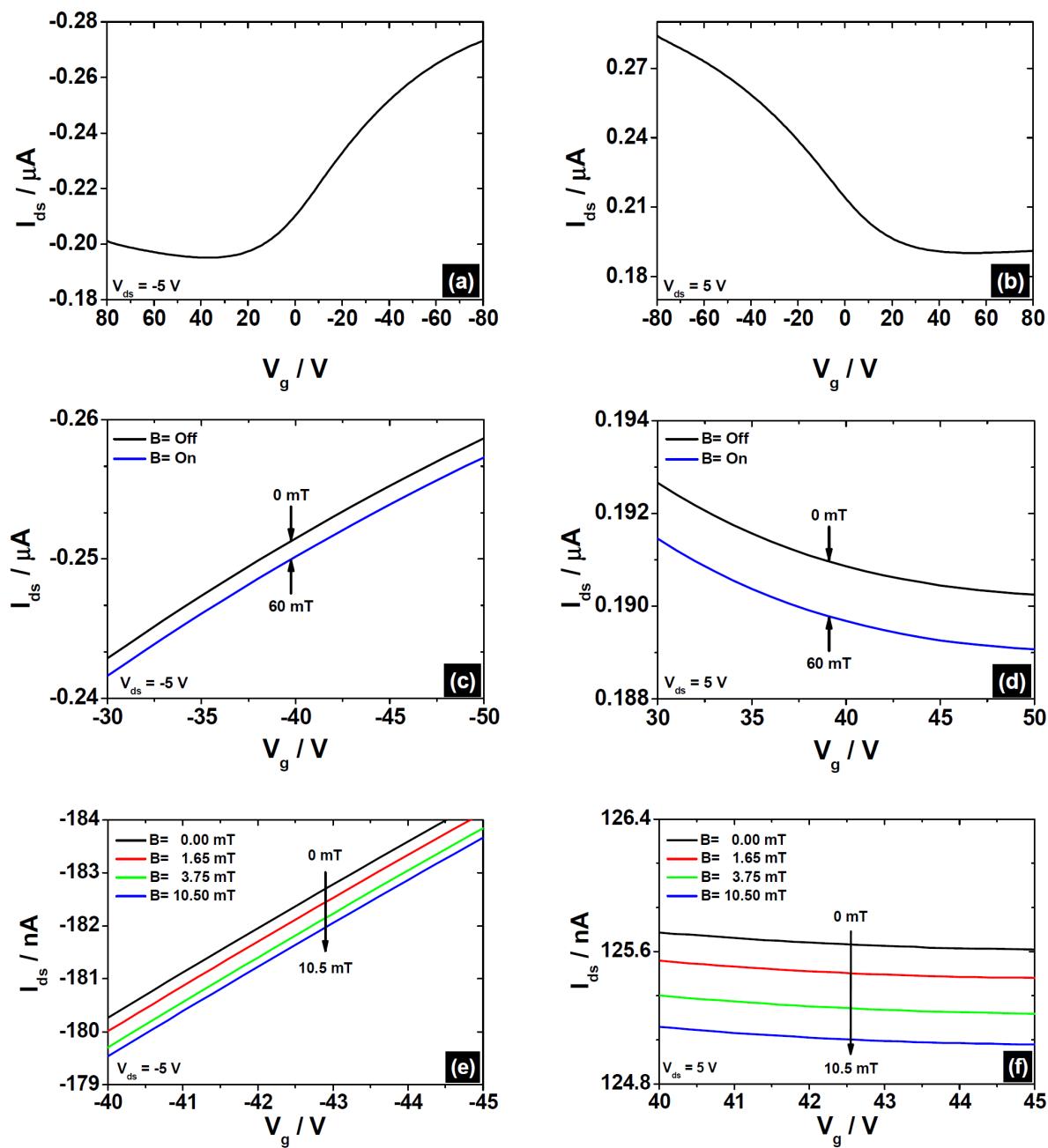


Figure S4. I_{ds} - V_g curves of devices based on evenly blended Spiro-TTB/HAT-CN devices for *p*-channel ($V_{ds} = -5$ V) on the left (a, c, f) and *n*-channel ($V_{ds} = 5$ V) on the right side (b, d, f). I_{ds} - V_g -curves are shown with V_g being swept from (a) 80 V to -80 V at $V_{ds} = -5$ V (*p*-channel) and (b) -80 V to 80 V at $V_{ds} = 5$ V (*n*-channel) without an external magnetic field. The magnetoresponse of the I_{ds} - V_g -measurements for low magnetic fields of 60 mT (c, d) and very low magnetic fields below 11 mT (e, f) are depicted as well. All measurements also show very high reproducibility.

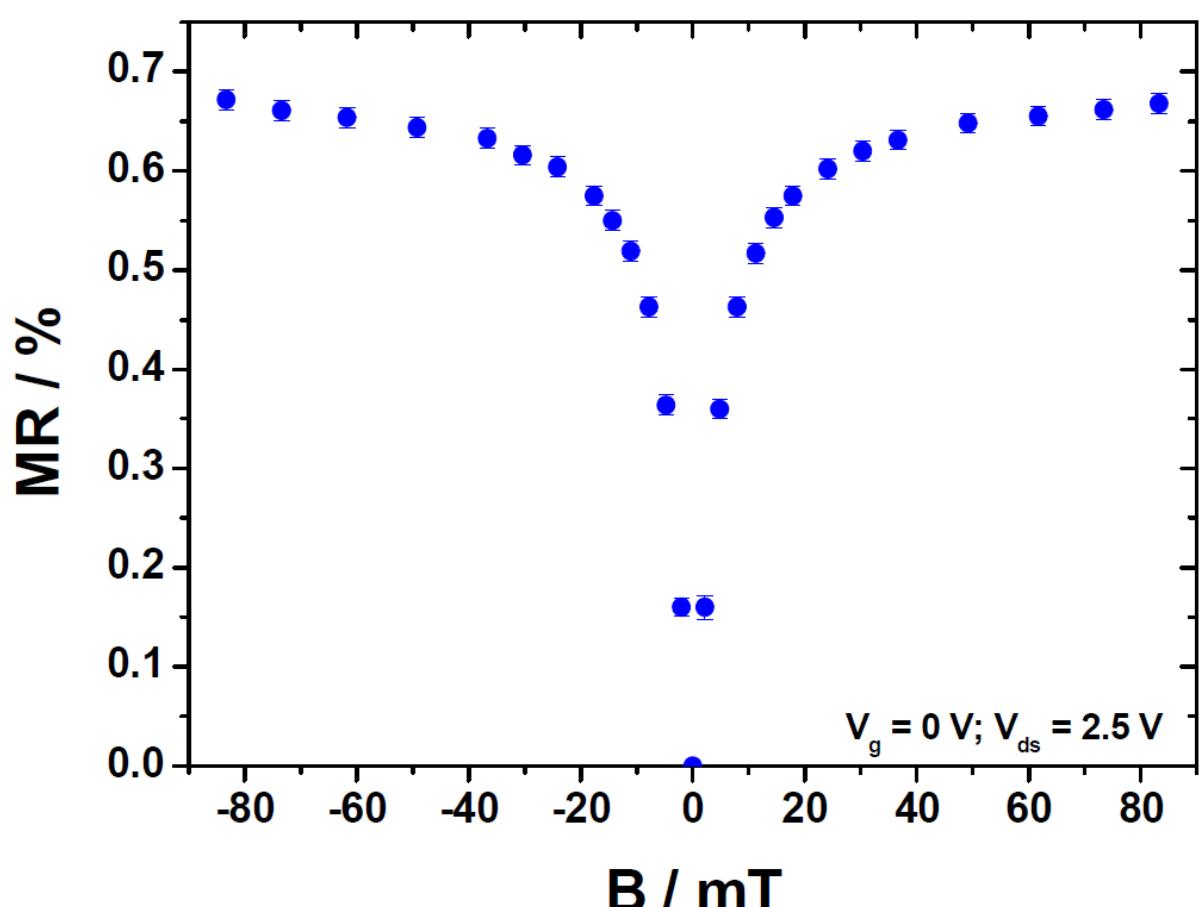


Figure S5. Dependence of MR values on the strength of the external magnetic field for devices based on evenly blended Spiro-TTB/HAT-CN transport layers.