Supplementary Information (SI) for Physical Chemistry Chemical Physics.

First-Principles Study of Spin-Selective Transport Properties and Quantum Logic Operation in Transition-Metal-Capped Nanowires.

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Supporting information

Table S1: (Both electrode and TM are parallel)

System (TM-C_7-TM) FM	Conductance(in nS)			
	Spin Up	Spin Down	Ic (charge current)	Is (spin current)
Sc	142.86	3054.61	3197.47	2911.75
Ti	223.06	6209.99	6433.05	5986.93
V	1305.69	18520.45	19826.14	17214.76
Cr	41203.47	28206.06	69409.53	-12997.4
Mn	507.97	25170.54	25678.51	24662.57
Fe	128.70	20623.87	20752.57	20495.17
Co	107.91	24288.73	24396.64	24180.82
Ni	59.16	2423.73	2482.89	2364.57

Table S2: (Electrode are anti-parallel & both TM are parallel)

System (TM-C_n-TM) AFM	Conductance(in nS)			
	Spin Up	Spin Down	Ic (charge current)	Is (spin current)
Sc	20274.07	20234.34	40508.41	-39.73
Ti	25291.11	18054.56	43345.67	-7236.55
V	31238.28	16157.55	47395.83	-15080.7
Cr	62109.29	1799.49	63908.78	-60309.8
Mn	1280.59	1717.82	2998.41	437.23
Fe	769.37	3335.98	4105.35	2566.61
Co	1000.24	9742.53	10742.77	8742.29
Ni	21551.05	1555.75	23106.8	-19995.3

Table S3: (TMR-1)

TM	TMR= (Ipara-Ianti-para)/Ianti-para	TMR (%)
Sc	-0.92107	-92.1067
Ti	-0.85159	-85.1587
V	-0.58169	-58.169
Cr	0.086072	8.60719
Mn	7.564042	756.4042
Fe	4.055006	405.5006
Co	1.270982	127.0982
Ni	-0.89255	-89.2547

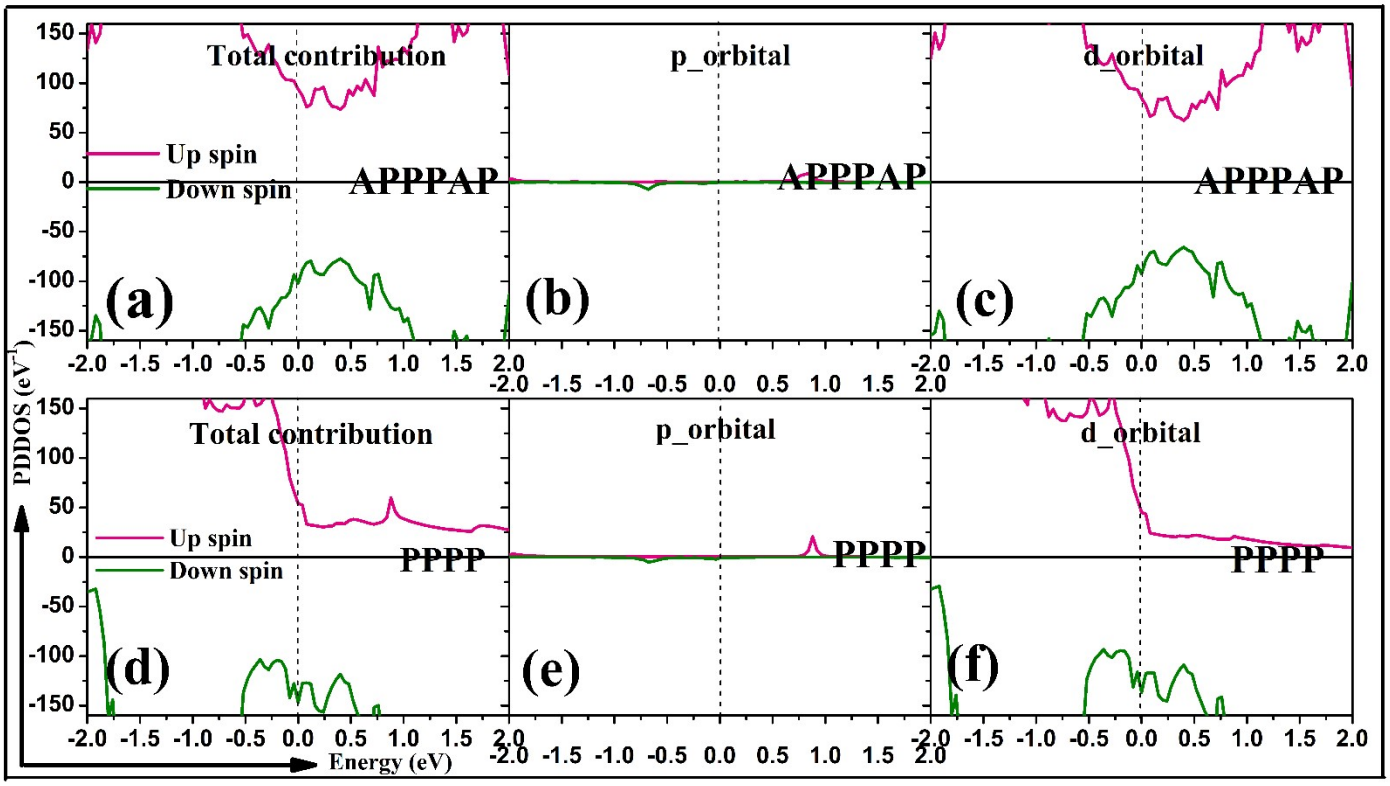


Figure S1: Spin-polarised PDDOS plot under two-probe device configuration at zero-bias at a constat spin configuration of the Mn-C_7-Mn sample: (a) Total contribution in APPPAP state; (b) Contribution from p-orbital in APPPAP state; (c) Contribution from d-orbital in APPPAP state; (d) Total contribution in APPPAP state; (e) Total contribution in APPPAP state; (f) Contribution from d-orbital in APPPAP state.

Table S4: PDDOS data at the Fermi-level.

CONFIGURATION	TOTAL		p-orbital		d-orbital	
	Up-spin	Down-spin	Up-spin	Down-spin	Up-spin	Down-spin
PPPP	54.07	146.99	0.10	0.81	45.17	136.68
APPPAP	93.91	102.04	0.19	0.21	84.83	92.46

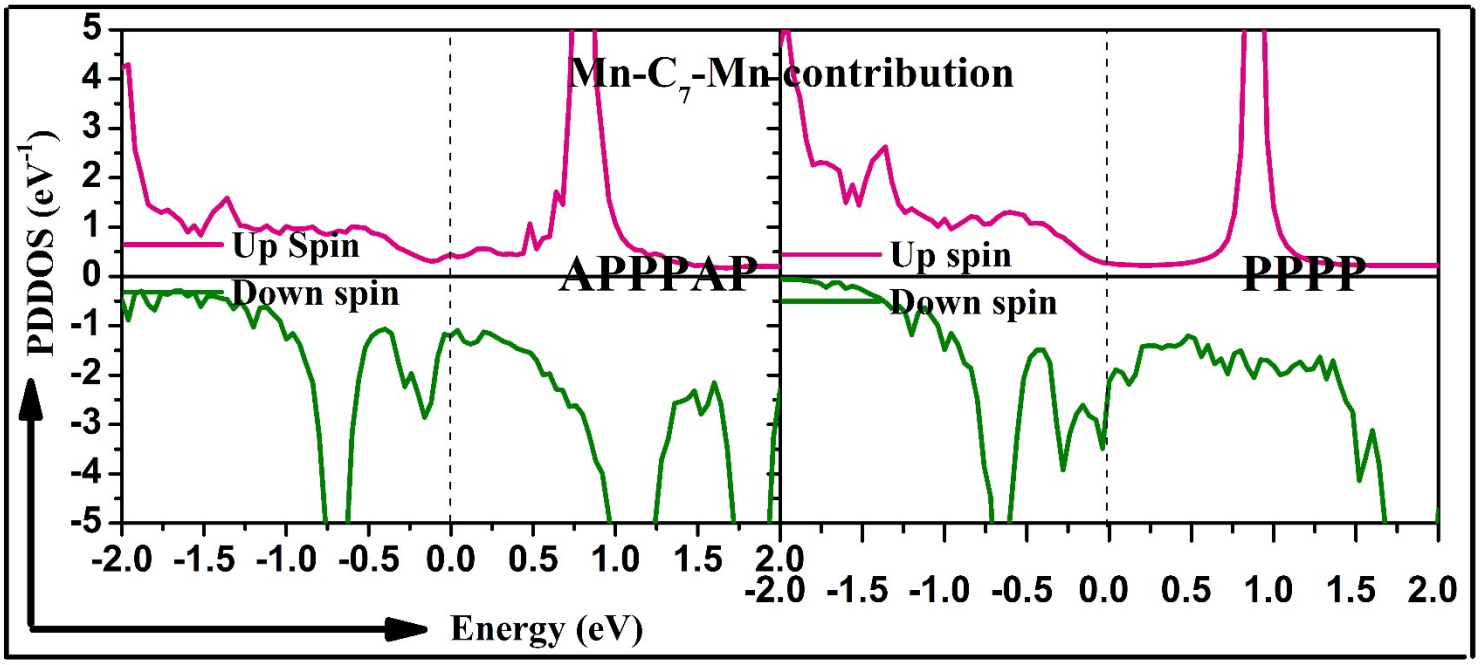


Figure S2: PDDOS of the molecular sample Mn-C7-Mn for APPPAP and PPPP states.

Table S5: Table for PDDOS contribution of molecular sapmle $Mn–C_7–Mn$ at the Fermi level

Constibution for	FM				AFM			
	Up spin (↑)	Down spin (↓)	Total PDDOD (↑+↓)	Δ_{PDDOS} (↑-↓)	Up spin (↑)	Down spin (↓)	Total (↑+↓)	Δ_{PDDOS} (↑-↓)
Mn-C7-Mn	0.265	2.129	2.394	1.864	0.445	1.204	1.649	0.759

Table S6: Magnetic moment calculation from PDDOS data.

	FM Configuration		AFM Configuration
	PPPP		APPPAP
Down-spin	2.129		1.204
Up-spin	0.265		0.445
Difference	1.864		0.759
Magnetic Moment	1.864 Bohr Magneton		0.759 Bohr Magneton

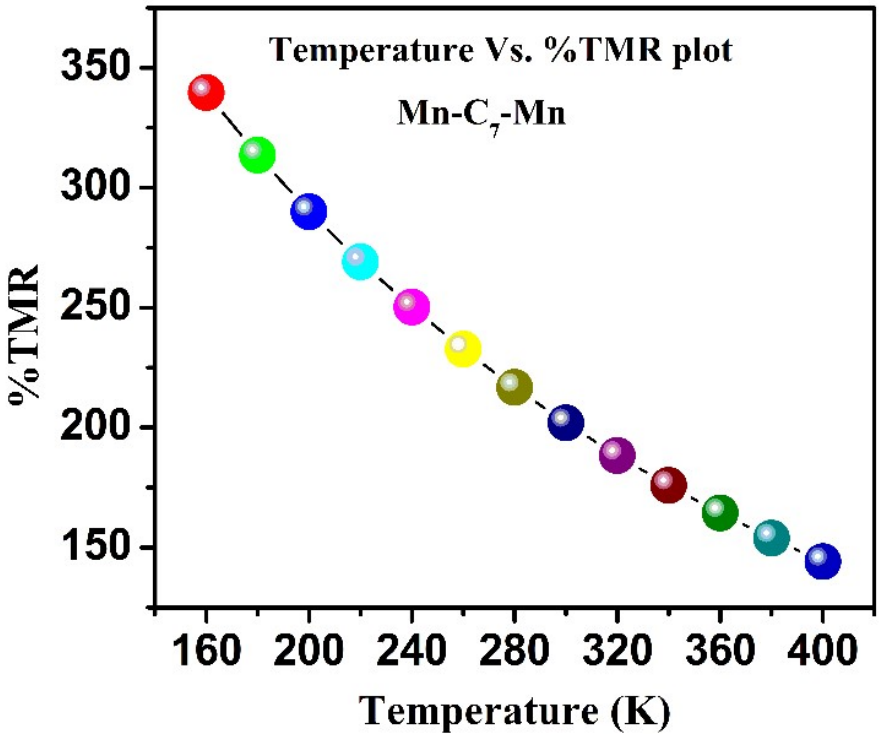


Figure S3: Temperature response of % TMR of the sample Mn-C_7-Mn.

Table S7: %of TMR of the sample Mn-C7-Mn consider the Hubbard-U effect and compare with without Hubbard-U effect.

Configuration	Conductance (nS)		Charge conductance (nS)	TMR	%TMR
	Up spin	Down spin			
PPPP	3878.63	4338.99	8217.62	35.43531	3543.531
APPPAP	192.63	32.91	225.54		
Without Hubbard-U potential					
PPPP	507.97	25170.54	24662.57	7.564042	756.4042
APPPAP	1280.59	1717.82	2998.41		

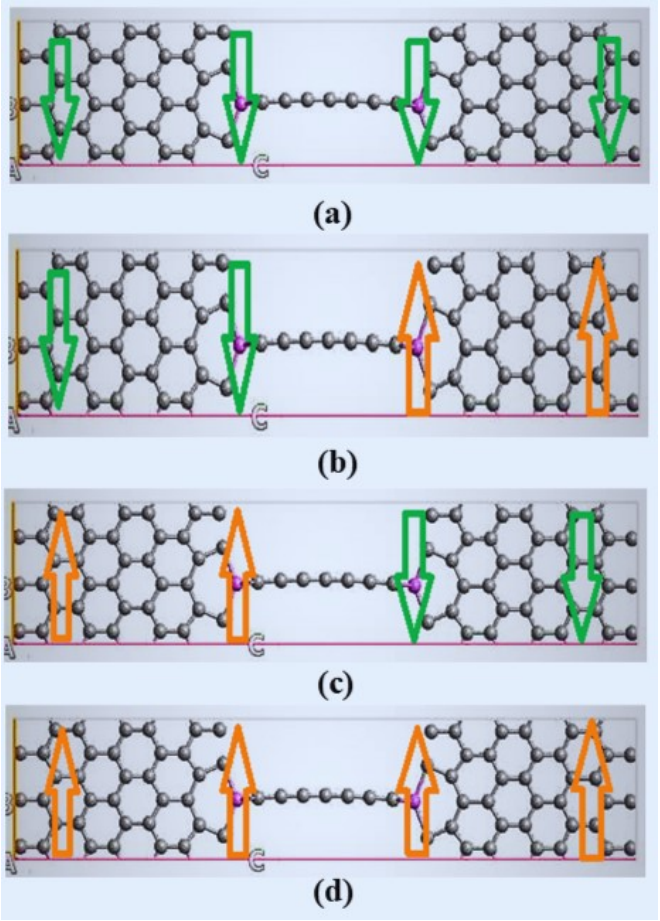


Figure S4: Spin resolved eigenstates of spin molecular logic device (SMLD): (a) APAP-APAP; (b) APAP-PP; (c) PP-APAP; (d) PP-PP

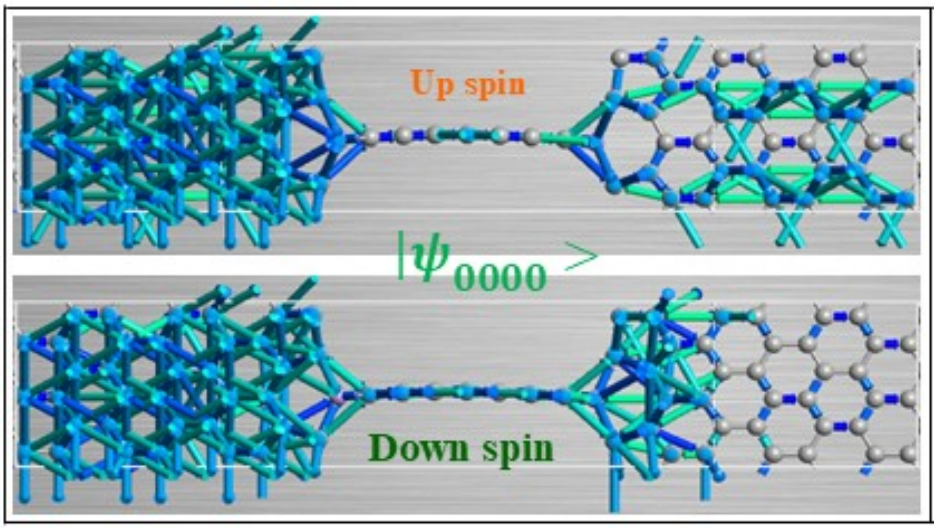


Figure S5: Transmission pathways of electrons through the central scattering region of the sample spin molecular logic device (SMLD) at the bias voltage of -0.8 V for quantum eigenstate $|\psi_{0000}>$

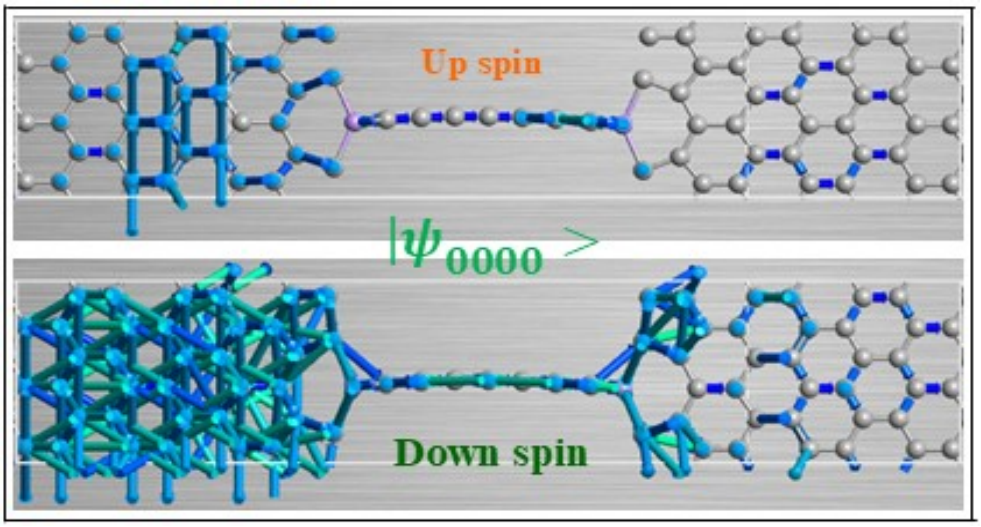
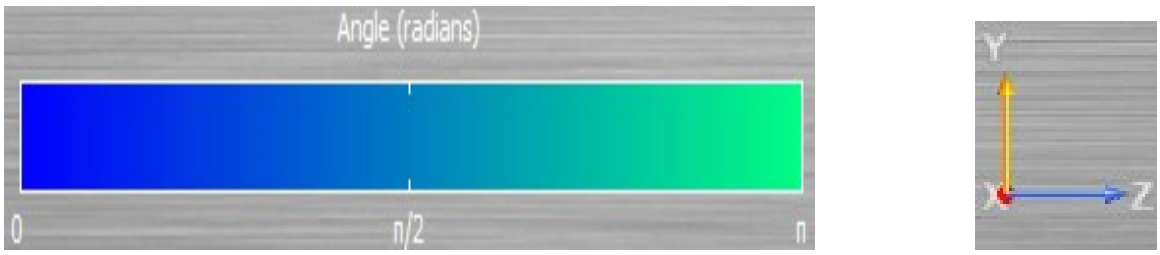


Figure S6: Transmission pathways of electrons through the central scattering region of the sample spin molecular logic device (SMLD) at the bias voltage of -0.9 V for quantum eigenstate $|\psi_{0000}>$



(a) (b)

Figure S7: (a) atom-to-atom relative phase difference; (b) Axial representation of lattice vector.

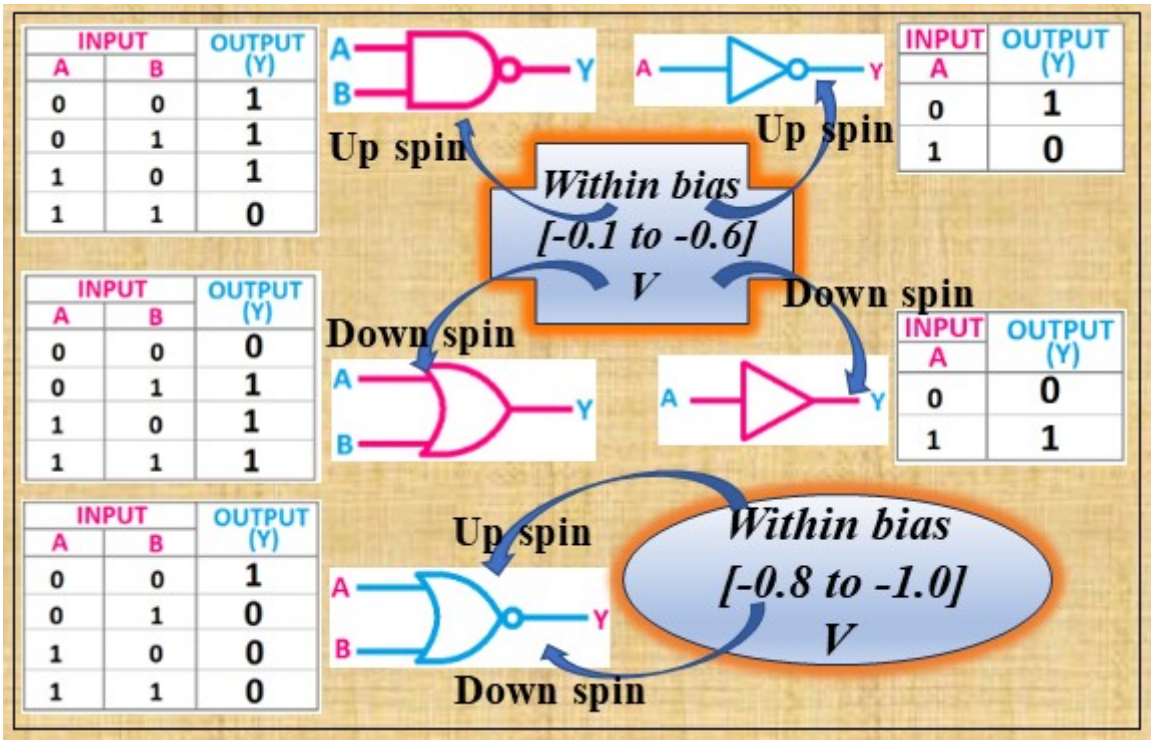


Figure S8: Truth table and gate symbol for the corresponding SMLG

Table S8: Transmission coefficient data within the scattering region for the eigenstates $|\psi_{0000}>$ and $|\psi_{0011}>$ at a bias voltage of -0.5 V.

# Item: 1	# Item: 2
# Input eigenstate $\|\psi_{0000}>$	# Input eigenstate $\|\psi_{0011}>$
Pathways for Spin = Up Down	Pathways for Spin = Up Down
30 -> 32 (-1, 0, 0) : 2.819840e-07 -6.710968e-08	30 -> 30 (-1, 0, 0) : 1.134675e-01 9.305300e-02
30 -> 33 (-1, 0, 0) : 1.814691e-01 7.750989e-02	30 -> 30 (-1, 0, 0) : 1.134675e-01 9.305300e-02
30 -> 36 (-1, 0, 0) : 3.503568e-02 2.255515e-03	32 -> 32 (-1, 0, 0) : 1.690480e-01 8.830121e-02
31 -> 31 (-1, 0, 0) : 1.227225e-01 8.902077e-02	32 -> 35 (-1, 0, 0) : 9.474300e-02 1.402427e-01
31 -> 34 (-1, 0, 0) : 1.919208e-01 8.060372e-02	33 -> 33 (-1, 0, 0) : 2.007482e-01 1.417838e-01
31 -> 33 (1, 0, 0) : 2.207049e-03 4.290567e-04	33 -> 33 (1, 0, 0) : -1.656410e-01 -1.352460e-01
32 -> 34 (0, 0, 0) : 1.813738e-02 4.686324e-03	34 -> 34 (-1, 0, 0) : 1.717298e-01 1.444020e-01
32 -> 35 (0, 0, 0) : 1.425405e-02 7.387631e-03	34 -> 35 (1, 0, 0) : 2.276337e-02 2.146583e-02
32 -> 34 (1, 0, 0) : 3.705829e-02 1.523544e-03	34 -> 36 (1, 0, 0) : -1.224966e-01 1.009120e-02
32 -> 33 (1, 1, 0) : 2.880543e-02 3.553270e-03	35 -> 35 (-1, 0, 0) : 8.881919e-02 1.446316e-01
33 -> 34 (0,-1, 0) : 3.251260e-03 5.902124e-04	35 -> 36 (-1, 0, 0) : -8.145169e-04 -8.251983e-04
33 -> 34 (2, 0, 0) : 3.083286e-06 -3.432494e-07	36 -> 37 (0, 0, 0) : 7.537476e-01 9.012600e-02
34 -> 36 (-1, 0, 0) : 9.596119e-02 -1.671182e-02	37 -> 38 (0, 0, 0) : 6.404375e-02 7.588325e-02
34 -> 36 (-1, 1, 0) : 1.147575e-06 8.859385e-07	40 -> 40 (-1, 0, 0) : 5.956594e-02 3.235692e-02
34 -> 36 (0, 0, 0) : 2.678415e-02 1.787595e-02	40 -> 41 (1, 0, 0) : -6.204671e-02 4.338580e-03
34 -> 35 (1, 0, 0) : 4.301194e-02 -9.932899e-04	41 -> 43 (2, 0, 0) : 4.388882e-07 7.486994e-09
34 -> 37 (2, 0, 0) : 1.003001e-04 -3.535975e-04	42 -> 44 (-1, 0, 0) : -1.140021e-01 -1.380423e-01
35 -> 37 (1, 1, 0) : 1.614205e-04 -8.791571e-05	43 -> 43 (-1, 0, 0) : 2.109069e-01 2.256057e-01
36 -> 37 (-1, 0, 0) : 2.792519e-03 -1.361770e-03	43 -> 44 (-2, 0, 0) : 5.569288e-03 2.499182e-04
36 -> 38 (-1, 0, 0) : 1.442048e-03 6.432330e-04	43 -> 44 (0, 0, 0) : 9.333826e-02 -1.770861e-03
36 -> 37 (0, 0, 0) : 4.320144e-01 4.235353e-02	43 -> 45 (0, 0, 0) : 1.828749e-01 1.641947e-02
36 -> 39 (2, 0, 0) : 1.584576e-05 1.300587e-06	43 -> 46 (0, 0, 0) : 1.442376e-01 2.310051e-02
37 -> 37 (-1, 0, 0) : 2.842891e-02 1.773130e-02	44 -> 46 (-1, 0, 0) : 1.553656e-01 1.001416e-01
37 -> 38 (0, 0, 0) : 1.818981e-01 3.146864e-02	45 -> 47 (-1,-1, 0) : 1.980572e+00 1.798186e-02
37 -> 37 (2, 0, 0) : 1.227371e-05 -2.809364e-06	45 -> 48 (-1, 0, 0) : -1.784598e+00 9.309056e-03
38 -> 39 (0, 0, 0) : 1.719432e-01 3.737539e-02	46 -> 46 (-1, 0, 0) : 8.930724e-01 3.004658e-01
38 -> 38 (1, 0, 0) : 2.361993e-02 -6.930640e-02	46 -> 47 (-1, 0, 0) : -1.621395e+00 1.536315e-02
39 -> 40 (0, 0, 0) : 1.866358e-01 3.919513e-02	47 -> 48 (1, 1, 0) : 1.678528e+00 -5.310494e+00

39 -> 40 (1, 0, 0) : 5.304405e-02 5.625367e-04 40 -> 41 (0, 0, 0) : 1.694712e-01 3.576934e-02 40 -> 42 (0, 0, 0) : 2.606773e-02 -4.736983e-04 41 -> 42 (-1, 0, 0) : 1.681816e-01 -5.673895e-03 41 -> 43 (-1, 0, 0) : 1.510783e-02 2.090594e-03 41 -> 41 (1, 0, 0) : 1.615648e-01 -4.090993e-03 42 -> 43 (0, 0, 0) : 1.235433e-01 4.327434e-02 42 -> 43 (1, 0, 0) : 2.892485e-01 -2.854256e-02 43 -> 43 (-1, 0, 0) : 2.910850e-01 1.198755e-02 43 -> 44 (-1, 0, 0) : 2.056083e+00 -2.688362e-02 44 -> 45 (0, 0, 0) : 3.572969e-02 2.302971e-03 44 -> 46 (0, 0, 0) : 5.183803e-02 -4.507922e-04 45 -> 45 (-1, 0, 0) : 1.100095e-01 5.379151e-02 46 -> 46 (-1, 0, 0) : 8.087401e-02 5.427339e-02 46 -> 48 (-1, 0, 0) : 1.778613e-03 -6.047709e-05 47 -> 47 (-1, 0, 0) : 4.724469e-02 2.128091e-02 47 -> 57 (0, 1, 0) : 1.366981e-04 9.968479e-06 47 -> 49 (1, 0, 0) : 1.655536e-03 -9.683900e-03 48 -> 50 (-1,-1, 0) : 3.834752e-03 -1.623691e-02 48 -> 51 (-1, 0, 0) : 2.401729e-02 -3.324170e-02 48 -> 54 (1, 0, 0) : 1.205514e-02 -3.952712e-04 49 -> 49 (-1, 0, 0) : 1.965793e-02 2.953093e-02 49 -> 52 (-1, 0, 0) : 1.132581e-02 -5.535526e-03 49 -> 53 (-1, 0, 0) : 1.595539e-02 -3.245943e-02 49 -> 54 (-1, 0, 0) : 7.005891e-05 1.983430e-05 49 -> 50 (0, 0, 0) : 3.050232e-03 -2.900621e-03 49 -> 52 (0, 0, 0) : 1.292082e-02 6.284463e-03 49 -> 53 (0, 0, 0) : 5.815014e-02 -2.856273e-02 50 -> 54 (0, 1, 0) : 2.787742e-03 -2.982811e-03 50 -> 56 (1, 0, 0) : 1.007612e-02 7.591849e-03 50 -> 54 (1, 1, 0) : 1.027574e-02 7.287220e-03	48 -> 50 (-1,-1, 0) : -1.362922e-01 -5.915283e-01 48 -> 52 (-1, 0, 0) : -1.500243e-01 -2.494517e-01 49 -> 51 (-1, 0, 0) : 2.739898e-02 1.593424e-02 49 -> 50 (0, 0, 0) : 2.554193e+00 2.715851e+00 49 -> 53 (0, 0, 0) : -9.511553e+00 1.333679e-02 50 -> 50 (1, 0, 0) : -3.625742e+00 -2.216529e+00 50 -> 52 (1, 0, 0) : 1.252967e-02 -1.964483e-02 50 -> 53 (1, 0, 0) : 2.912503e+00 -2.004096e-02

In the transmission pathways analysis, values of the local bond contributions T_{ij}(E) with relatively high and positive magnitudes are areas of interest.

Explanation with example: (a) Quantum Eigenstate$|\psi_{0000}>$

30 -> 33 (-1, 0, 0) :

Contribution to the up spin channel: 1.814691e-01

Contribution to the down spin channel: 7.750989e-02

This suggests transmission from atomic site 30 to site 33. (-1, 0, 0) represent direction of transport. 1.814691e-01 and 7.750989e-02 are the transmission coefficients corresponding to the spin up channel and down spin channel during the transmission from the site 30 to 33. Positive value imply transmission from the atomic site 30 to atomic site 33. Up spin has higher transmission rate than down spin channel.

41 -> 42 (-1, 0, 0) :

Contribution to the up spin channel: 1.681816e-01

Contribution to the down spin channel: -5.673895e-03

1.681816e-01 is the transmission coefficient corresponding to the spin up channel from the atomic site 41 to atomic site 42. On the other hand, -5.673895e-03 is the transmission coefficient corresponding to the down spin channel during the transmission. Negative value implies transmission from the atomic site 42 to atomic site 41.

The tabulated results reveal that the spin-up channel exhibits a greater number of strong positive contributions, indicating coherent transmission between atomic sites. In contrast, the spin-down channel shows far fewer positive values, reflecting reduced electronic transmission. This asymmetry between spin channels is consistent with the spin-resolved transport behaviour and corroborates the features observed in Fig. 13(a) of the revised manuscript.

(b) Quantum Eigenstate $|\psi_{0011}>$:

44 -> 46 (-1, 0, 0) :

Contribution to the up spin channel: 1.553656e-01

Contribution to the down spin channel: 1.001416e-01

49 -> 50 (0, 0, 0) :

Contribution to the up spin channel: 2.554193e+00

Contribution to the down spin channel: 2.715851e+00

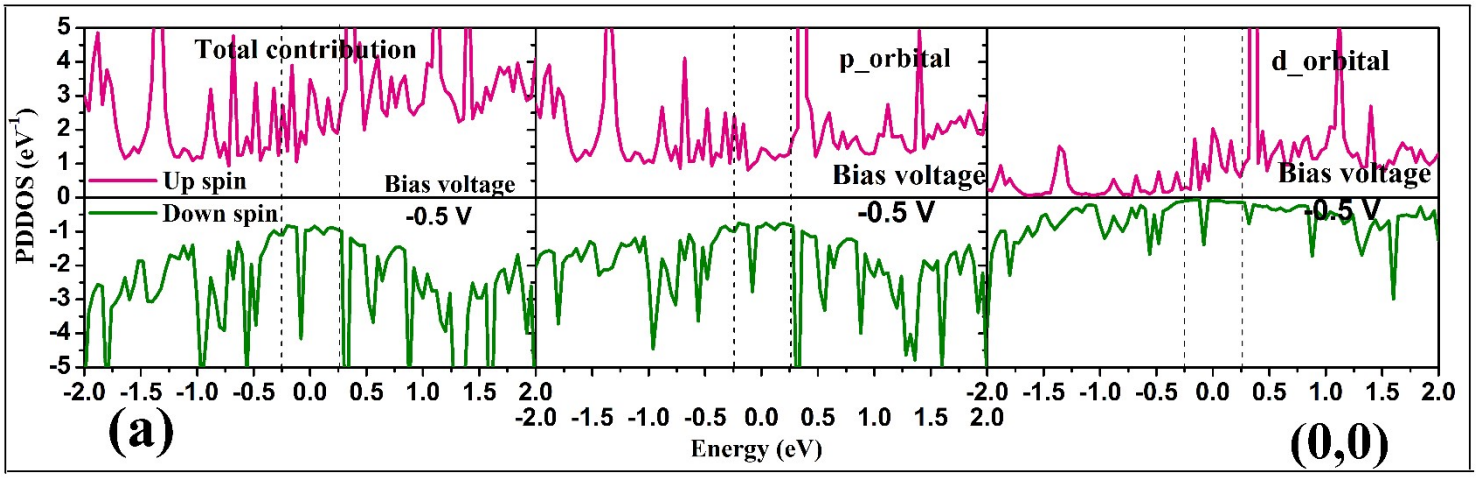
The data set in item 2 of table S8 clearly shows that both spin channels exhibit relatively high positive transmission coefficients, indicating efficient electron transport between the corresponding atomic sites. This nearly symmetric transmission behaviour across the two spin channels aligns well with the spin-resolved transport characteristics and supports the features presented in Fig. 13(b) of the revised manuscript.

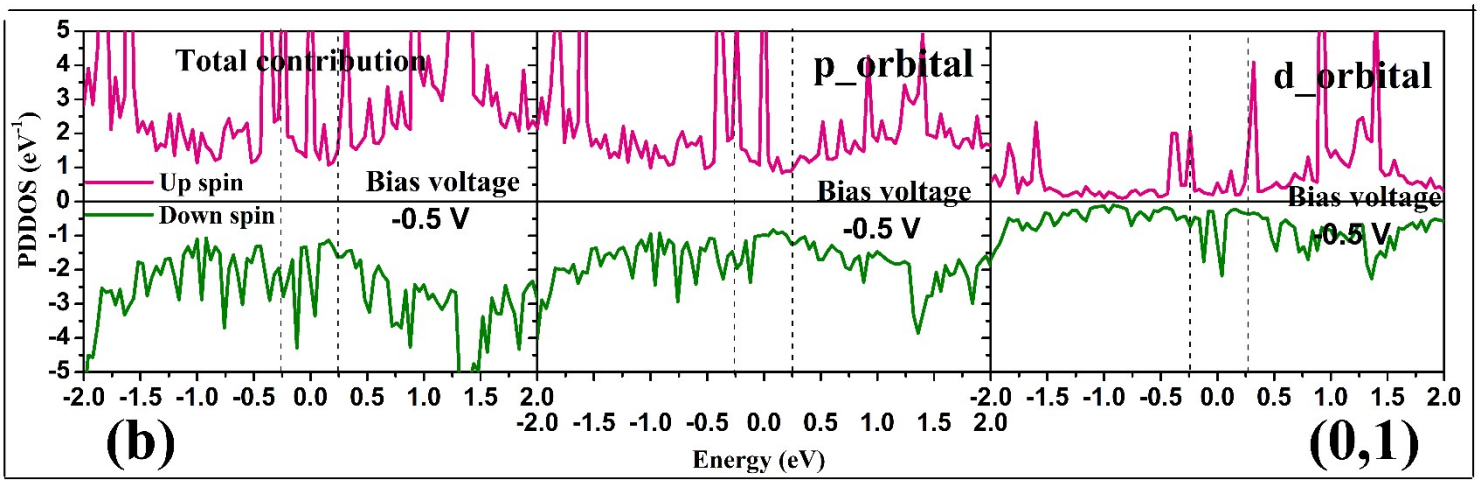
Table S9: Biased dependent spin logic operation

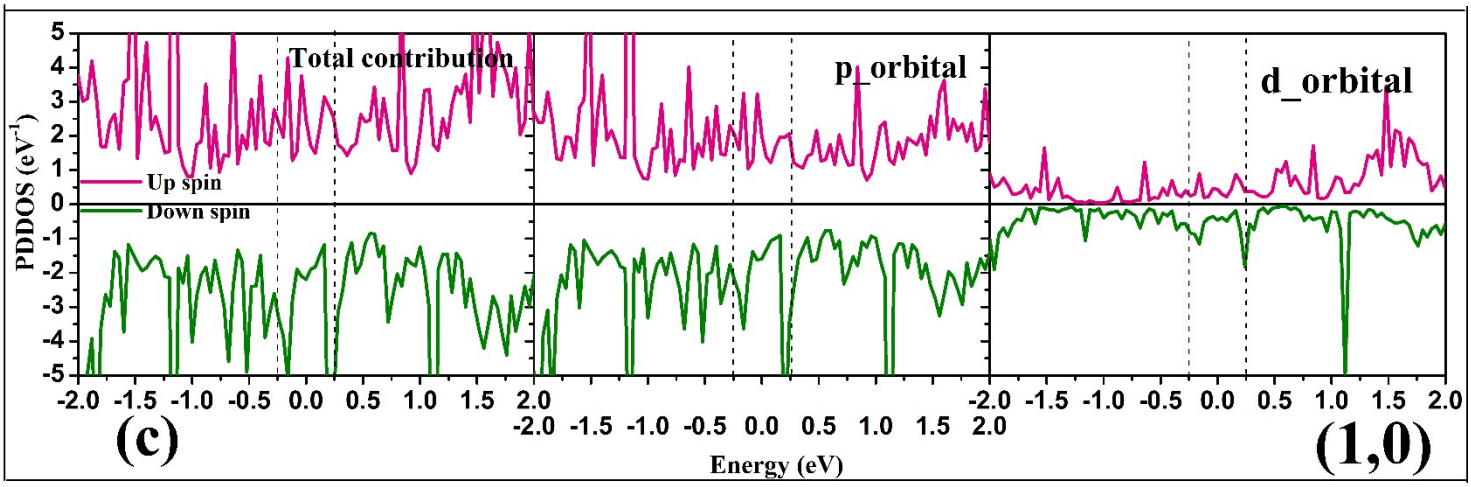
Inputs	Bias voltage (in Volt)				More symmetric inputs	Bias voltage (in Volt)			
	[-0.1 to -0.6]		[-0.8 to -1.0]			[-0.1 to -0.6]		[-0.8 to -1.0]	
	↑	↓	↑	↓		↑	↓	↑	↓
(0,0)	1	0	1	1	0	1	0	1	1
(0,1)	1	1	0	0					
(1,0)	1	1	0	0					
(1,1)	0	1	0	0	1	0	1	0	0
SMLD Functions as	NAND	OR	NOR			NOT	YES	NOT	

* Note: The input representation is based on the probable spin effect of left electrode and left Mn atom as input A and spin effect of right electrode and right Mn atom as input B. 0 for spin down effect and I for spin up effect.

** Bias voltage region of [-0.1 to -0.6 V] has spin selective and [-0.8 to 1.0 V] has spin independent logic response.







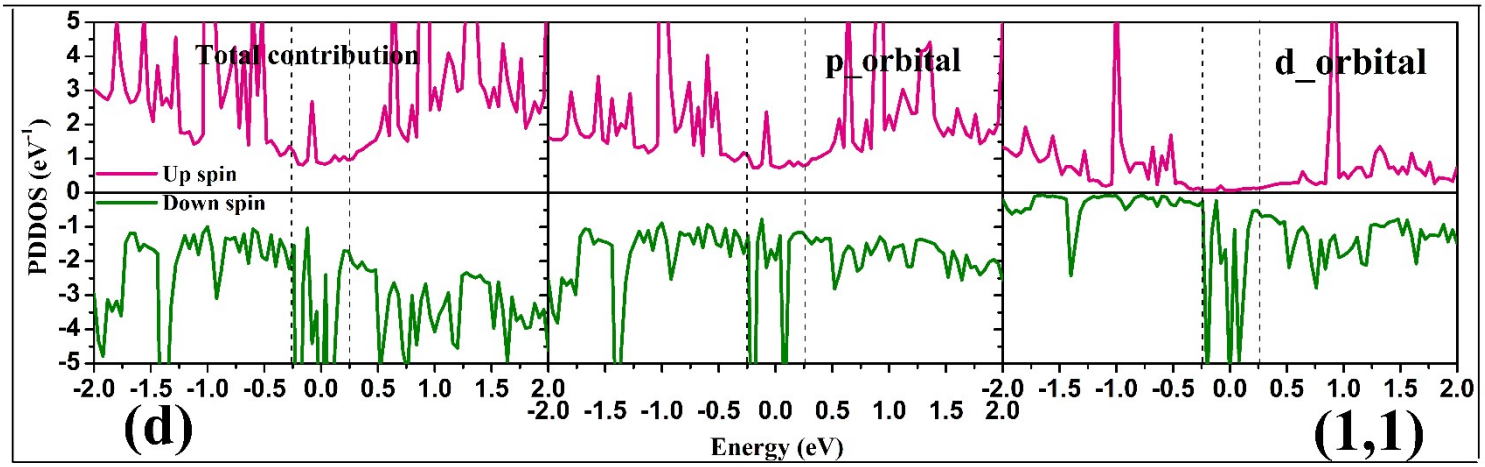


Figure S9: Visual mapping of LDOS from the scattering region of the two-probe device in a row of total contribution, p-orbital and d-orbital contribution for the eigenstate of (a) $|\psi_{0000}>$; (b) $|\psi_{0011}>$; (c) $|\psi_{1100}>$ and (d) $|\psi_{1111}>$. All the PDDOS from the scattering region are map at bias of -0.5 V.

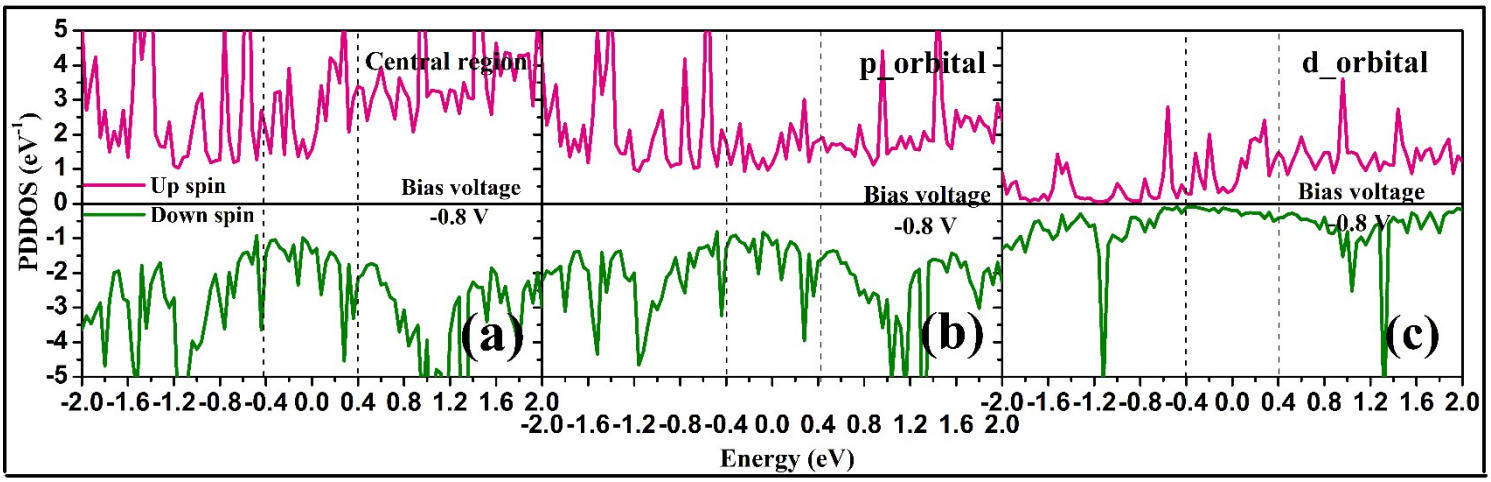


Figure S10: PDDOS analysis of the central scattering region within the bias window of -0.8 V; (a) total contribution; (b) p-orbital contribution; (c) d-orbital contribution.

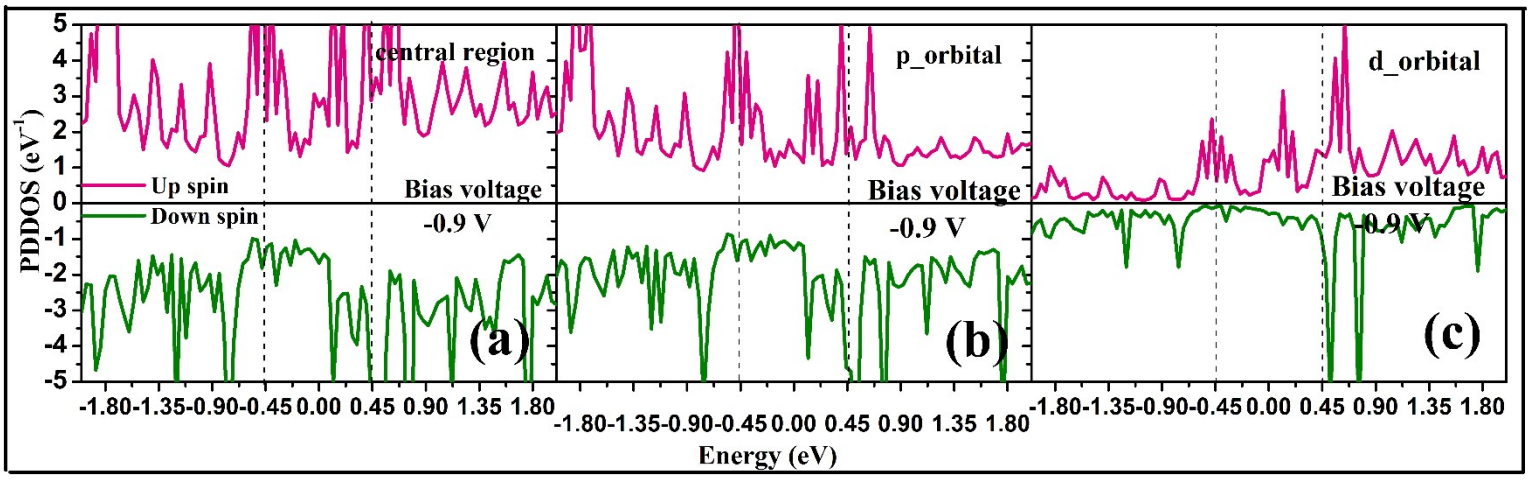


Figure S11: PDDOS analysis of the central scattering region within the bias window of -0.8 V; (a) total contribution; (b) p-orbital contribution; (c) d-orbital contribution.