Electronic Supplementary Material (ESI) for Nanoscale.


**Supplementary Information**

# Room temperature giant magnetoresistance in half-metallic $Cr_2C$ based two-dimensional tunnel junctions.

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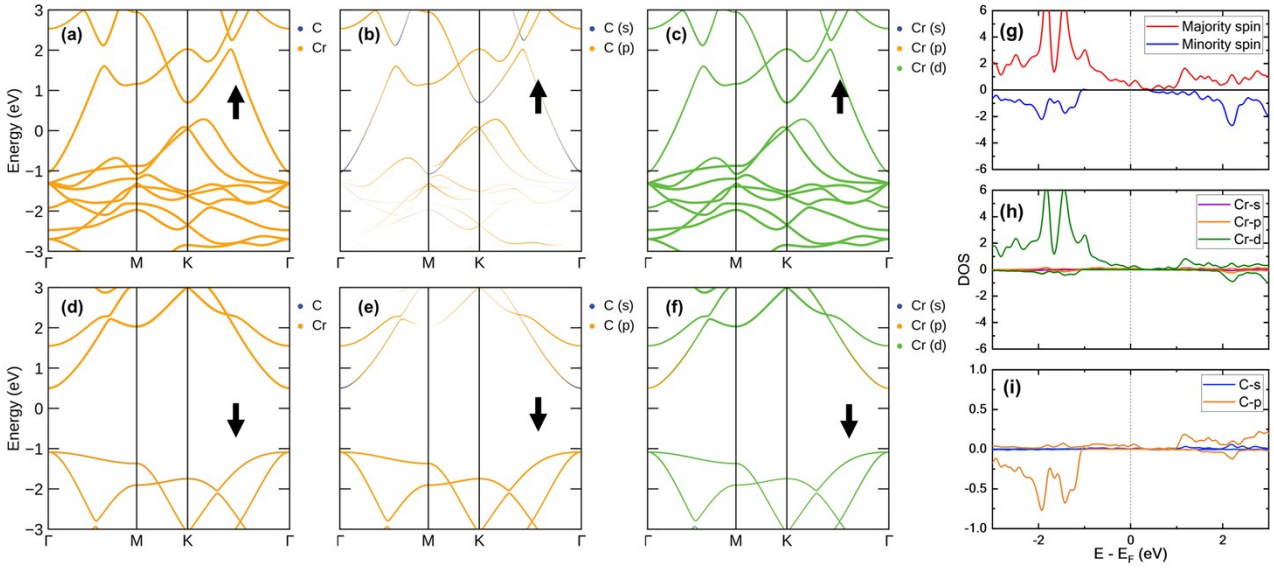


Fig. S1: Orbital projected band structures showing contribution of C, Cr, C-s, C-p, Cr-s, Cr-p, Cr-d orbitals for (a, b, c) majority spin up states and (d, e, f) minority spin down states. (g) Total and (h, i) projected density of states for Cr2C monolayer.

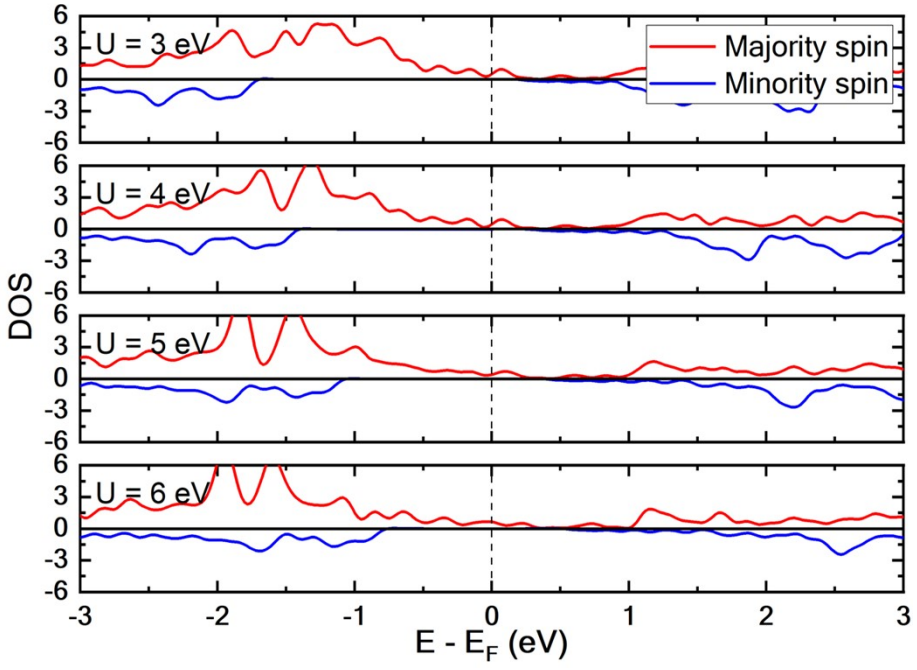


Fig. S2: Density of states at different levels of on-site Hubbard correction of U = 3, 4, 5, and 6 eV.

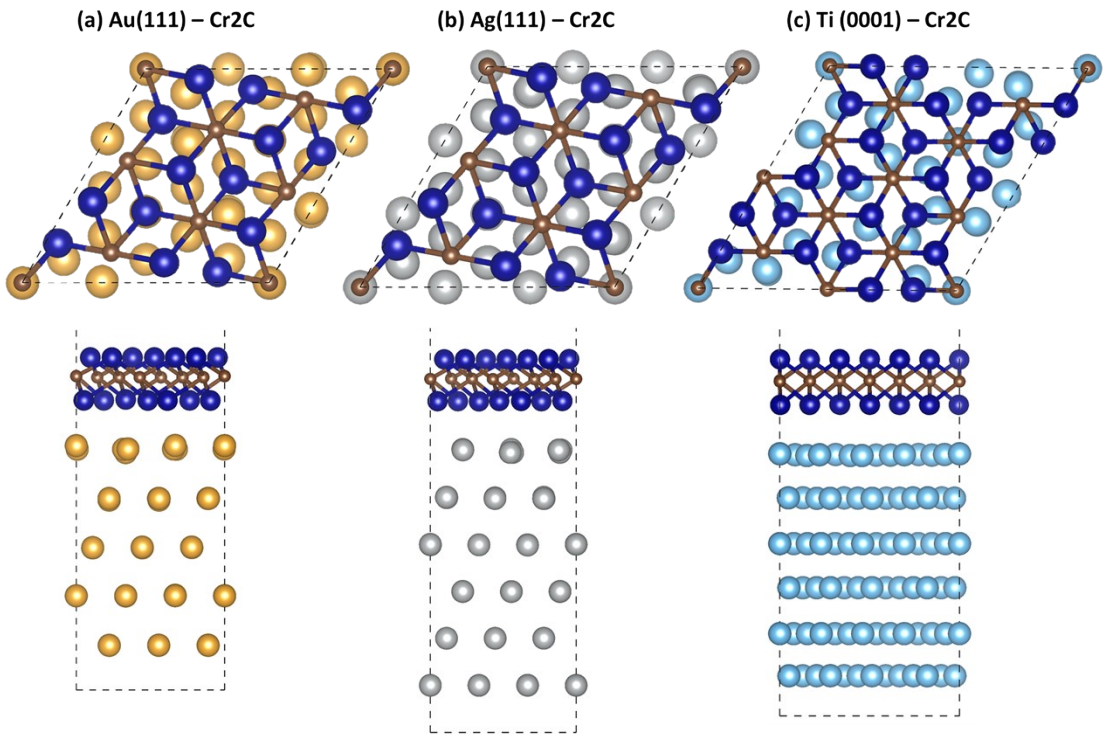


Fig. S3: Relaxed atomic structures (top and side views) of three different metal contact – Cr2C interface: (a) Au (111) – Cr2C, (b) Ag(111)-Cr2C, and (c) Ti (0001)-Cr2C

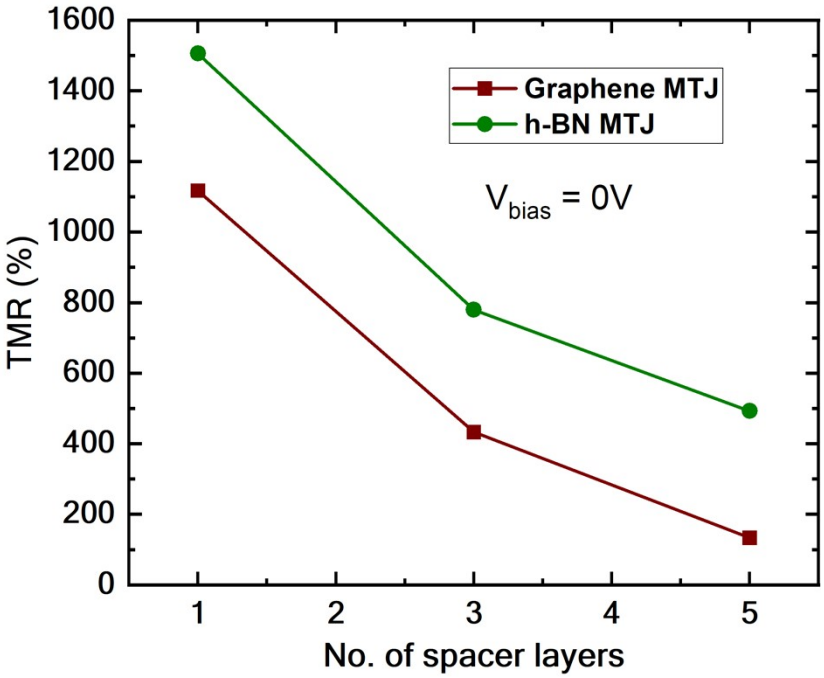


Fig. S4: Layer dependence of TMR of graphene and h-BN MTJs at zero voltage bias.

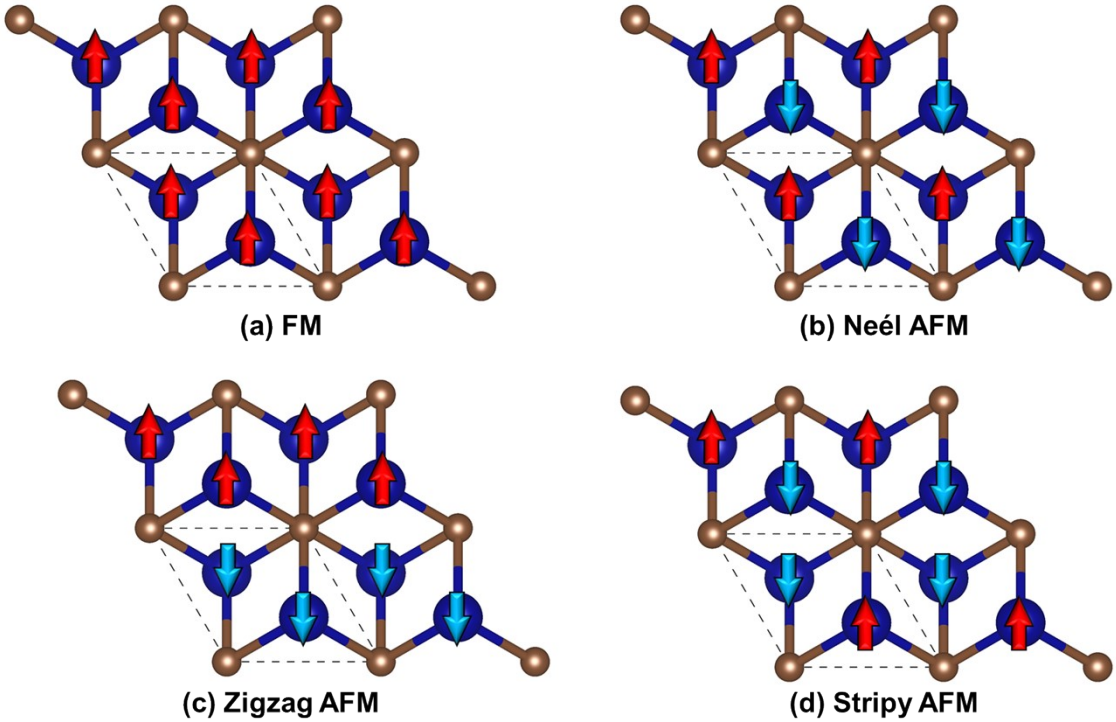


Fig. S5: Different magnetic configurations of Cr2C considered in this study with up spin denoted by red arrows and down spins by blue arrows; (a) Ferromagnetic (FM) ordering, (b) Neel antiferromagnetic (AFM) ordering, (c) Zigzag antiferromagnetic ordering, and (d) Stripy antiferromagnetic ordering.

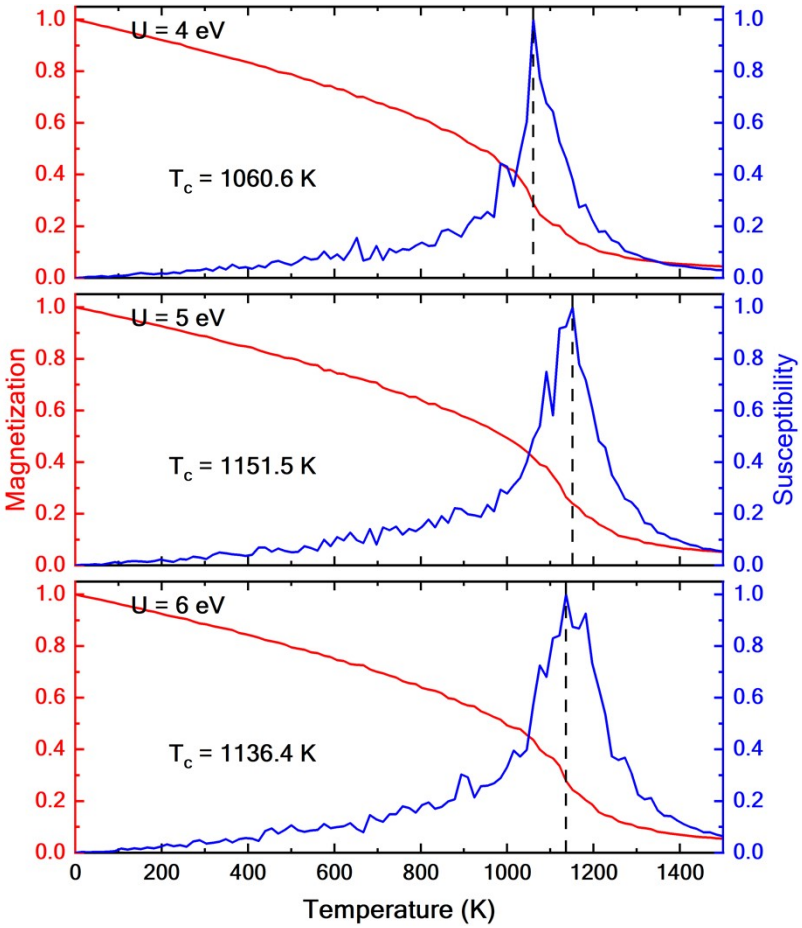


Fig. S6: Curie temperatures calculated from magnetization and susceptibility curves using different Hubbard correction parameters of U = 4, 5, and 6 eV.

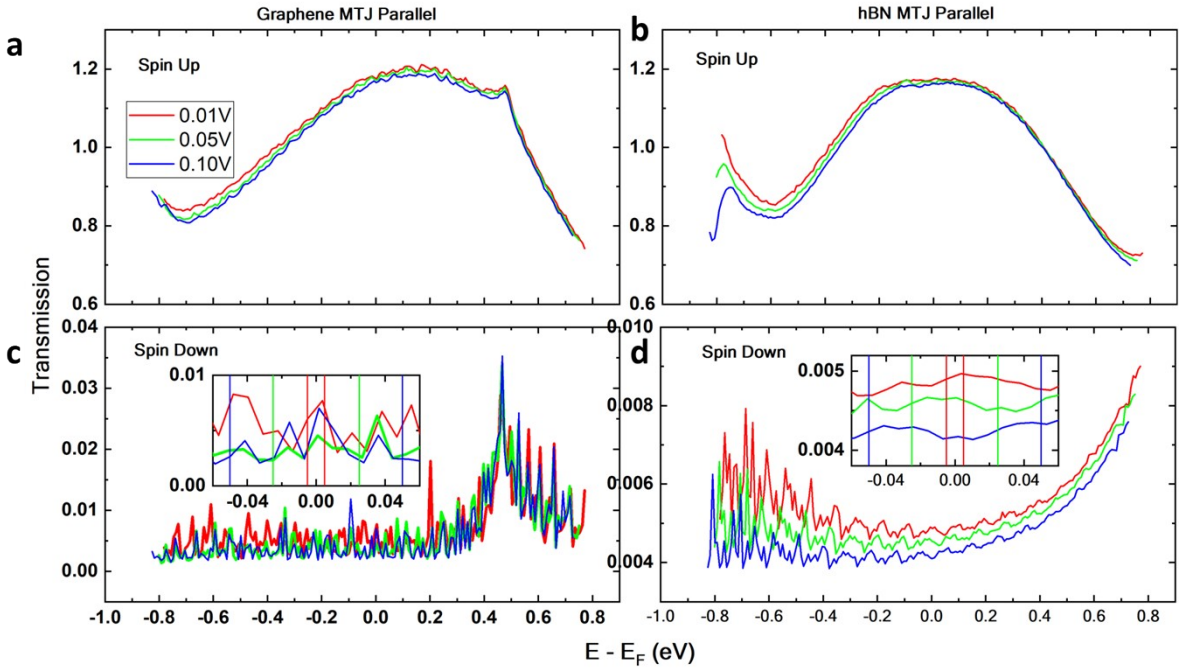


Fig. S7: Transmission coefficients at different bias voltages for (a, c) spin up and (b, d) spin-down components of the charge carriers for graphene barrier MTJ and hBN barrier MTJ in parallel configuration, respectively. The transmission coefficients for the spin-down carriers around the bias window are shown in the insets of (c, d), where vertical lines denote the bias window.

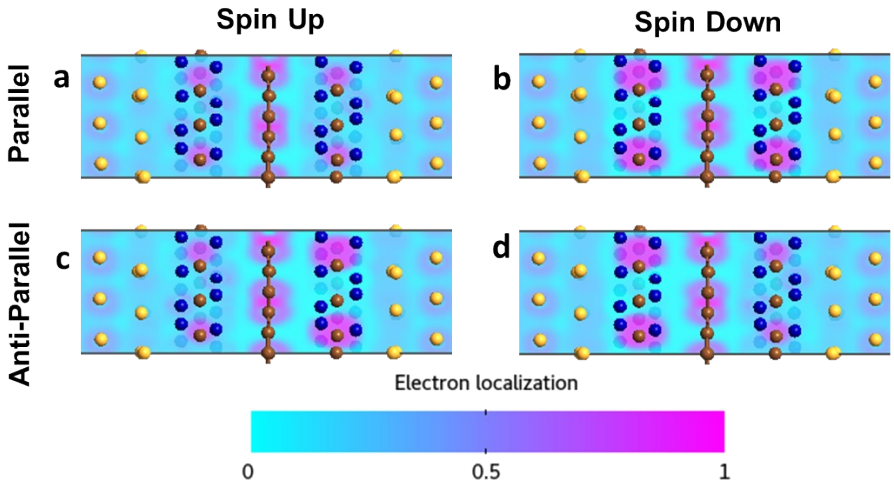


Fig. S8: Electron localization function along the transport direction at equilibrium for graphene MTJ. (a) Spin up, and (b) Spin down localizations for parallel configuration, (c) Spin up, and (d) Spin down localizations for anti-parallel configuration.

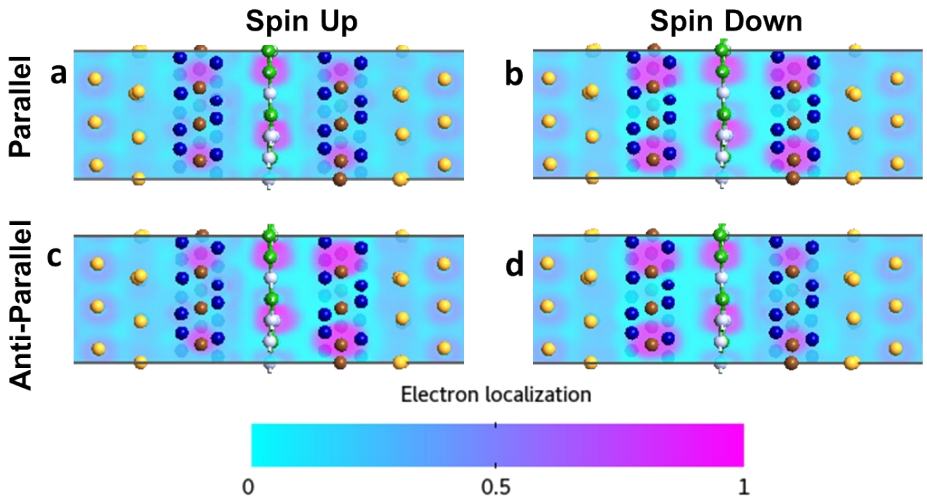


Fig. S9: Electron localization function along the transport direction at equilibrium for hBN MTJ. (a) Spin up, and (b) Spin down localizations for parallel configuration, (c) Spin up, and (d) Spin down localizations for anti-parallel configuration.

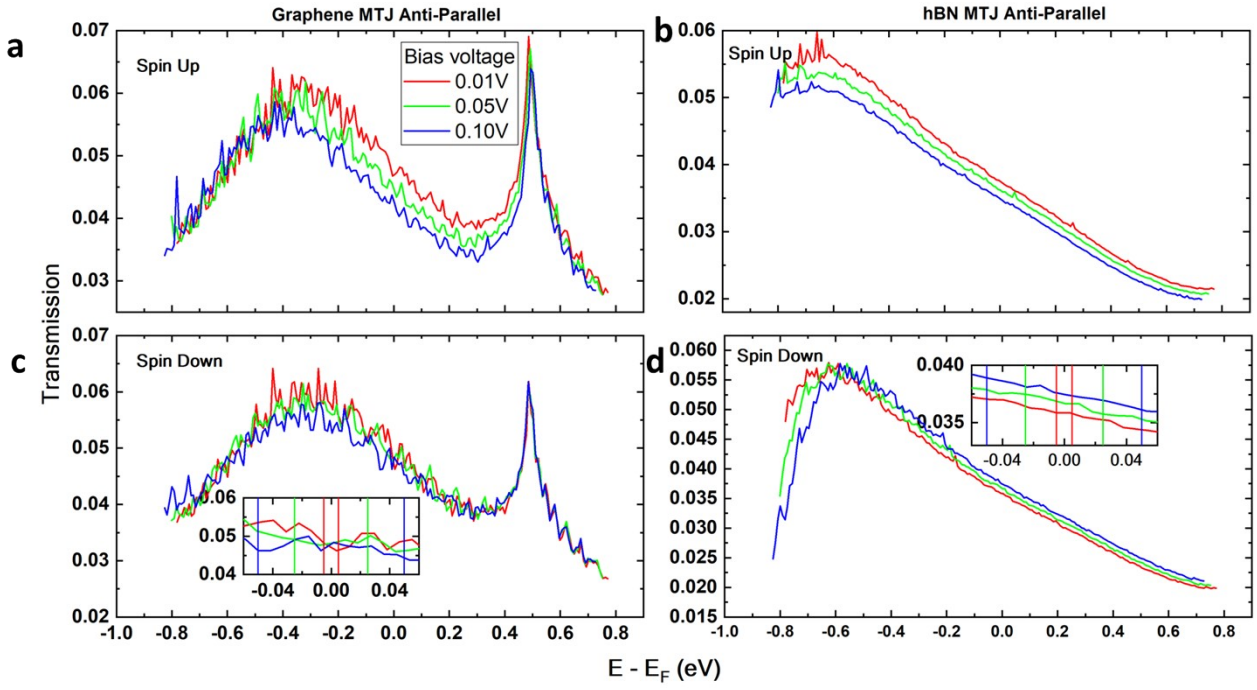


Fig. S10: Transmission coefficients at different bias voltages for (a, c) spin up and (b, d) spin-down components of the charge carriers for graphene barrier MTJ and hBN barrier MTJ in anti-parallel configuration, respectively. The transmission coefficients for the spin-down carriers around the bias window are shown in the insets of (c, d), where vertical lines denote the bias window.

Table S1: Heisenberg Hamiltonian parameters calculation

| U (eV) | Total collinear energy of different spin configurations (eV/atom). | Equations to Solve | Solutions (meV/link or Cr) | | Magnetic Moment ($\mu_B$) | Curie Temperature (K) | Magnetic Anisotropy Energy (μeV/Cr) | Easy Magnetic Axis |
|---|---|---|---|---|---|---|---|---|
| 5 | -9.81, -9.68, -9.57, -9.42 | E0 - 48.0*J1 - 96.0*J2 - 48.0*J3 + 78.50 = 0<br>E0 + 48.0*J1 - 96.0*J2 + 48.0*J3 + 77.44 = 0<br>E0 - 16.0*J1 + 32.0*J2 + 48.0*J3 + 76.58 = 0<br>E0 + 16.0*J1 + 32.0*J2 - 48.0*J3 + 75.35 = 0<br>-32.0*Ax - 48.0*K1x - 96.0*K2x - 48.0*K3x + 8.72e-2 = 0<br>-32.0*Ax + 48.0*K1x - 96.0*K2x + 48.0*K3x + 8.69e-2 = 0<br>-32.0*Ax - 16.0*K1x + 32.0*K2x + 48.0*K3x + 8.94e-2 = 0<br>-32.0*Ax + 16.0*K1x + 32.0*K2x - 48.0*K3x + 8.77e-2 = 0<br>-32.0*Az - 48.0*K1z - 96.0*K2z - 48.0*K3z + 8.77e-2 = 0<br>-32.0*Az + 48.0*K1z - 96.0*K2z + 48.0*K3z + 8.70e-2 = 0<br>-32.0*Az - 16.0*K1z + 32.0*K2z + 48.0*K3z + 8.94e-2 = 0<br>-32.0*Az + 16.0*K1z + 32.0*K2z - 48.0*K3z + 8.81e-2 = 0 | J1<br>J2<br>J3<br>K1x<br>K2x<br>K3x<br>K1z<br>K2z<br>K3z<br>Ax<br>Az | 17.91<br>15.64<br>-6.82<br>16.24e-3<br>-11.92e-3<br>-12.51e-3<br>15.98e-3<br>-11.05e-3<br>8.40e-3<br>2.75<br>2.76 | 3.755 | 1151.5 | 60.88 | z |

Table S2: Mulliken population analysis for MTJ with graphene spacer.

| | Atom Index | Element | Spin-up charge | Spin-down charge |
|---|---|---|---|---|
| Left Cr2C electrode | 54 | Chromium | 5.047 | 0.647 |
| | 55 | Chromium | 5.047 | 0.647 |
| | 56 | Chromium | 5.047 | 0.647 |
| | 57 | Chromium | 5.062 | 0.622 |
| | 58 | Chromium | 5.062 | 0.622 |
| | 59 | Chromium | 5.062 | 0.622 |
| | 60 | Chromium | 5.126 | 0.718 |
| | 61 | Carbon | 1.408 | 2.837 |
| | 62 | Carbon | 1.408 | 2.837 |
| | 63 | Carbon | 1.408 | 2.837 |
| | 64 | Carbon | 1.403 | 2.842 |
| | 65 | Carbon | 1.403 | 2.842 |
| | 66 | Carbon | 1.403 | 2.842 |
| | 67 | Carbon | 1.429 | 2.858 |
| | 68 | Chromium | 5.192 | 0.667 |
| | 69 | Chromium | 5.192 | 0.667 |
| | 70 | Chromium | 5.192 | 0.667 |
| | 71 | Chromium | 5.209 | 0.674 |
| | 72 | Chromium | 5.18 | 0.686 |
| | 73 | Chromium | 5.18 | 0.686 |
| | 74 | Chromium | 5.18 | 0.686 |
| Right Cr2C electrode | 99 | Chromium | 5.204 | 0.647 |
| | 100 | Chromium | 5.193 | 0.669 |
| | 101 | Chromium | 5.193 | 0.669 |
| | 102 | Chromium | 5.193 | 0.669 |
| | 103 | Chromium | 5.18 | 0.689 |
| | 104 | Chromium | 5.18 | 0.689 |
| | 105 | Chromium | 5.18 | 0.689 |
| | 106 | Carbon | 1.433 | 2.853 |
| | 107 | Carbon | 1.404 | 2.844 |
| | 108 | Carbon | 1.404 | 2.844 |
| | 109 | Carbon | 1.404 | 2.844 |
| | 110 | Carbon | 1.407 | 2.836 |
| | 111 | Carbon | 1.407 | 2.836 |
| | 112 | Carbon | 1.407 | 2.836 |
| | 113 | Chromium | 5.124 | 0.72 |
| | 114 | Chromium | 5.063 | 0.621 |
| | 115 | Chromium | 5.063 | 0.621 |
| | 116 | Chromium | 5.063 | 0.621 |
| | 117 | Chromium | 5.046 | 0.646 |
| | 118 | Chromium | 5.046 | 0.646 |
| | 119 | Chromium | 5.046 | 0.646 |

| | | | | |
|---|---|---|---|---|
| | 75 | Carbon | 2.019 | 2.006 |
| | 76 | Carbon | 2.014 | 2.008 |
| | 77 | Carbon | 2.014 | 2.008 |
| | 78 | Carbon | 2.014 | 2.008 |
| | 79 | Carbon | 2.019 | 2.007 |
| | 80 | Carbon | 2.019 | 2.007 |
| | 81 | Carbon | 2.019 | 2.007 |
| | 82 | Carbon | 2.017 | 2.008 |
| | 83 | Carbon | 2.017 | 2.008 |
| | 84 | Carbon | 2.017 | 2.008 |
| | 85 | Carbon | 2.017 | 2.008 |
| Graphene Spacer | 86 | Carbon | 2.017 | 2.008 |
| | 87 | Carbon | 2.017 | 2.008 |
| | 88 | Carbon | 2.019 | 2.007 |
| | 89 | Carbon | 2.018 | 2.007 |
| | 90 | Carbon | 2.018 | 2.007 |
| | 91 | Carbon | 2.018 | 2.007 |
| | 92 | Carbon | 2.016 | 2.007 |
| | 93 | Carbon | 2.016 | 2.007 |
| | 94 | Carbon | 2.016 | 2.007 |
| | 95 | Carbon | 2.019 | 2.007 |
| | 96 | Carbon | 2.019 | 2.007 |
| | 97 | Carbon | 2.019 | 2.007 |
| | 98 | Carbon | 2.013 | 2.009 |

Table S3: Mulliken population analysis for MTJ with h-BN spacer.

| | Index | Element | Spin-up charge | Spin-down charge |
|---|---|---|---|---|
| | 54 | Chromium | 4.863 | 0.099 |
| | 55 | Chromium | 4.863 | 0.099 |
| | 56 | Chromium | 4.863 | 0.099 |
| | 57 | Chromium | 4.874 | 0.094 |
| | 58 | Chromium | 4.874 | 0.094 |
| | 59 | Chromium | 4.874 | 0.094 |
| | 60 | Chromium | 4.941 | 0.1 |
| | 61 | Carbon | 1.703 | 0.046 |
| Left Cr2C layer | 62 | Carbon | 1.703 | 0.046 |
| | 63 | Carbon | 1.703 | 0.046 |
| | 64 | Carbon | 1.7 | 0.045 |
| | 65 | Carbon | 1.7 | 0.045 |
| | 66 | Carbon | 1.7 | 0.045 |
| | 67 | Carbon | 1.725 | 0.047 |
| | 68 | Chromium | 4.985 | 0.104 |
| | 69 | Chromium | 4.985 | 0.104 |
| | 70 | Chromium | 4.985 | 0.104 |
| | 71 | Chromium | 4.967 | 0.11 |

| | | | | |
|---|---|---|---|---|
| | 72 | Chromium | 4.967 | 0.11 |
| | 73 | Chromium | 4.967 | 0.11 |
| | 74 | Chromium | 5.037 | 0.104 |
| | 99 | Chromium | 4.976 | 0.104 |
| | 100 | Chromium | 4.976 | 0.104 |
| | 101 | Chromium | 4.976 | 0.104 |
| | 102 | Chromium | 5.025 | 0.105 |
| | 103 | Chromium | 4.979 | 0.11 |
| | 104 | Chromium | 4.979 | 0.11 |
| | 105 | Chromium | 4.979 | 0.11 |
| | 106 | Carbon | 1.724 | 0.046 |
| | 107 | Carbon | 1.705 | 0.046 |
| Right | 108 | Carbon | 1.705 | 0.046 |
| Cr2C | 109 | Carbon | 1.705 | 0.046 |
| layer | 110 | Carbon | 1.698 | 0.045 |
| | 111 | Carbon | 1.698 | 0.045 |
| | 112 | Carbon | 1.698 | 0.045 |
| | 113 | Chromium | 4.94 | 0.101 |
| | 114 | Chromium | 4.877 | 0.094 |
| | 115 | Chromium | 4.877 | 0.094 |
| | 116 | Chromium | 4.877 | 0.094 |
| | 117 | Chromium | 4.861 | 0.099 |
| | 118 | Chromium | 4.861 | 0.099 |
| | 119 | Chromium | 4.861 | 0.099 |
| | 75 | Boron | 1.867 | 1.796 |
| | 76 | Nitrogen | 2.234 | 0.013 |
| | 77 | Nitrogen | 2.234 | 0.013 |
| | 78 | Nitrogen | 2.234 | 0.013 |
| | 79 | Boron | 1.88 | 1.787 |
| | 80 | Boron | 1.88 | 1.787 |
| | 81 | Boron | 1.88 | 1.787 |
| | 82 | Boron | 1.869 | 1.778 |
| | 83 | Nitrogen | 2.226 | 0.01 |
| | 84 | Nitrogen | 2.226 | 0.01 |
| | 85 | Nitrogen | 2.226 | 0.01 |
| h-BN | 86 | Nitrogen | 2.231 | 0.012 |
| Spacer | 87 | Nitrogen | 2.231 | 0.012 |
| | 88 | Nitrogen | 2.231 | 0.012 |
| | 89 | Boron | 1.874 | 1.782 |
| | 90 | Boron | 1.874 | 1.782 |
| | 91 | Boron | 1.874 | 1.782 |
| | 92 | Boron | 1.872 | 1.774 |
| | 93 | Boron | 1.872 | 1.774 |
| | 94 | Boron | 1.872 | 1.774 |
| | 95 | Nitrogen | 2.236 | 2.218 |
| | 96 | Nitrogen | 2.236 | 2.218 |
| | 97 | Nitrogen | 2.236 | 2.218 |
| | 98 | Boron | 1.863 | 1.769 |