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**Supporting Information (SI)**

# H4,4,4-graphyne with double Dirac points as high-efficiency bifunctional electrocatalysts for water splitting

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**Table S1** Calculated energy of single TM atom ($E_{TM}$), and the unit is eV.

| TM | Sc | Ti | Mn | Fe | Co | Ni | Pd | Ir | Pt |
|---|---|---|---|---|---|---|---|---|---|
| $E_{TM}$ | –2.08 | –2.05 | –5.16 | –2.96 | –1.66 | –0.69 | –1.48 | –1.47 | –0.62 |

**Table S2** Calculated structural parameters of H4,4,4-GY: lattice constant, bond length $l_1$, $l_2$, $l_3$ and $l_4$, the unit is Å.

| | $a$ (Å) | $l_1$ | $l_2$ | $l_3$ | $l_4$ |
|---|---|---|---|---|---|
| H4,4,4-graphyne | 11.82 | 1.247 | 1.351 | 1.453 | 1.489 |

**Table S3** Geometry coordinates of H4,4,4-GY unit cell.

| | $x$ | $y$ | $z$ |
|---|---|---|---|
| C1 | 0.4498080594913604 | 0.2776521240056888 | 0.5000000002002796 |
| C2 | 0.7223457607876440 | 0.5501917866205626 | 0.4999999997521376 |
| C3 | 0.5501918687566763 | 0.7223490392824031 | 0.4999999998362595 |
| C4 | 0.2776493357252811 | 0.4498070415013354 | 0.5000000002251881 |
| C5 | 0.4725260261095471 | 0.4015497774807891 | 0.5000000006507673 |
| C6 | 0.5984467891407294 | 0.5274714884365177 | 0.4999999993096790 |
| C7 | 0.5274684115093161 | 0.5984494527829730 | 0.4999999993661225 |
| C8 | 0.4015480188996772 | 0.4725275926465286 | 0.5000000006311618 |
| C9 | 0.4498084125337444 | 0.1721344007632919 | 0.4999999998397184 |
| C10 | 0.8278627770641404 | 0.5501905885519126 | 0.5000000001945941 |
| C11 | 0.5501914465550511 | 0.8278665334369966 | 0.5000000001456874 |
| C12 | 0.1721334659034351 | 0.4498084308991612 | 0.4999999997954062 |
| C13 | 0.4725688394477252 | 0.0709890304923592 | 0.4999999997405423 |
| C14 | 0.9290085798887817 | 0.5274313114199366 | 0.5000000002301028 |
| C15 | 0.5274310718434376 | 0.9290116358972150 | 0.5000000002362344 |

| | | | |
|---|---|---|---|
| C16 | 0.0709876242390127 | 0.4725692154635029 | 0.4999999998148684 |
| C17 | 0.9290409176543298 | 0.4015352244210509 | 0.5000000001763447 |
| C18 | 0.4015348151832717 | 0.9290414319062611 | 0.5000000001411027 |
| C19 | 0.0709573600485594 | 0.5984650993466738 | 0.4999999998418171 |
| C20 | 0.5984653271793091 | 0.0709587278383544 | 0.4999999998344000 |
| C21 | 0.8279243046676785 | 0.2775968888065787 | 0.4999999998832971 |
| C22 | 0.2775969665094481 | 0.8279244658454973 | 0.4999999999113018 |
| C23 | 0.1720751815149288 | 0.7224032801241296 | 0.5000000001307827 |
| C24 | 0.7224033818786912 | 0.1720754394808605 | 0.5000000001122042 |

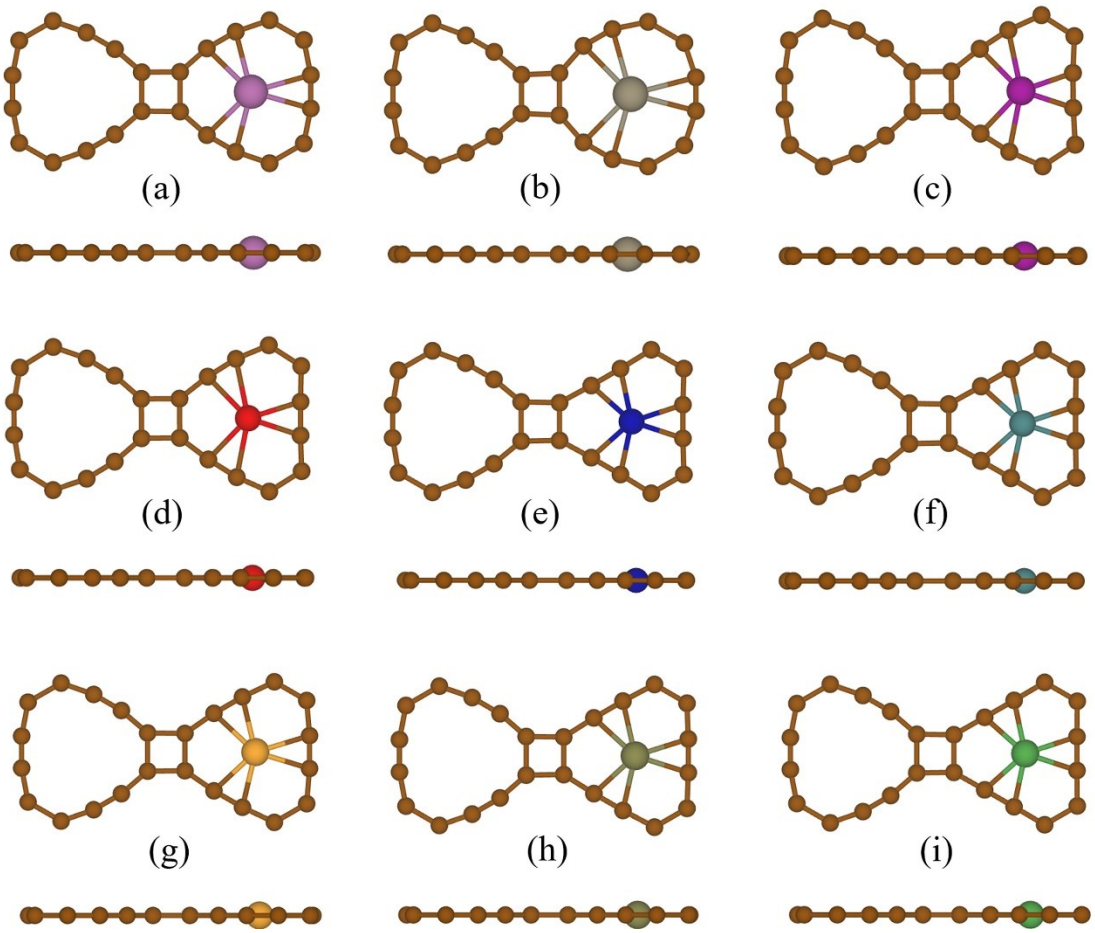


Figure S1. Optimized adsorption configuration of (a) Sc, (b) Ti, (c) Mn, (d) Fe, (e) Co, (f) Ni, (g) Pd, (h) Ir, and (i) Pt supported H4,4,4-GY.

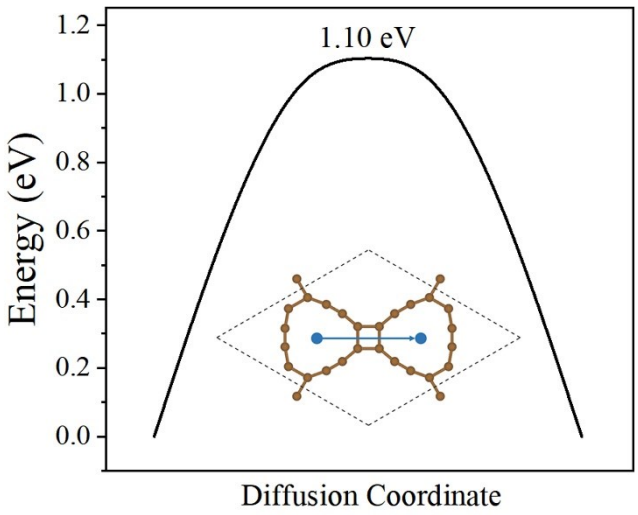


Figure S2. Diffusion pathway and energy barrier for single Co atom on H4,4,4-GY.

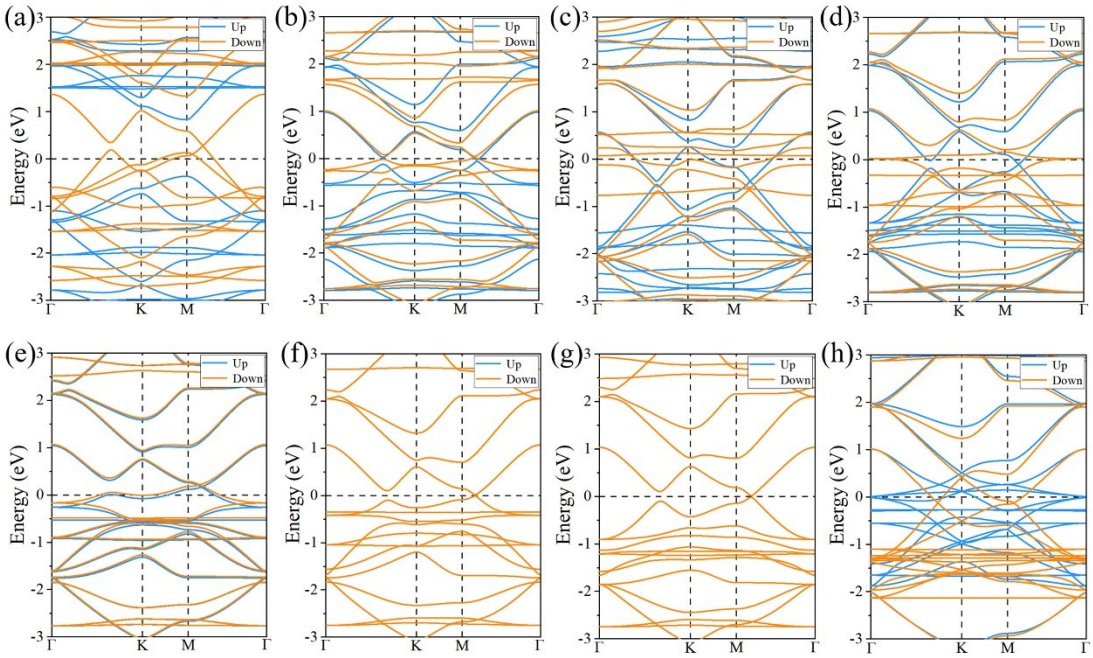


Figure S3. Spin-polarized band structure of (a) Sc@H4,4,4-GY, (b) Mn@H4,4,4-GY, (c) Fe@H4,4,4-GY, (d) Co@H4,4,4-GY, (e) Ni@H4,4,4-GY, (f) Ir@H4,4,4-GY, (g) Pt@H4,4,4-GY, and (h) $Co_2$@H4,4,4-GY, the Fermi level is set to zero.

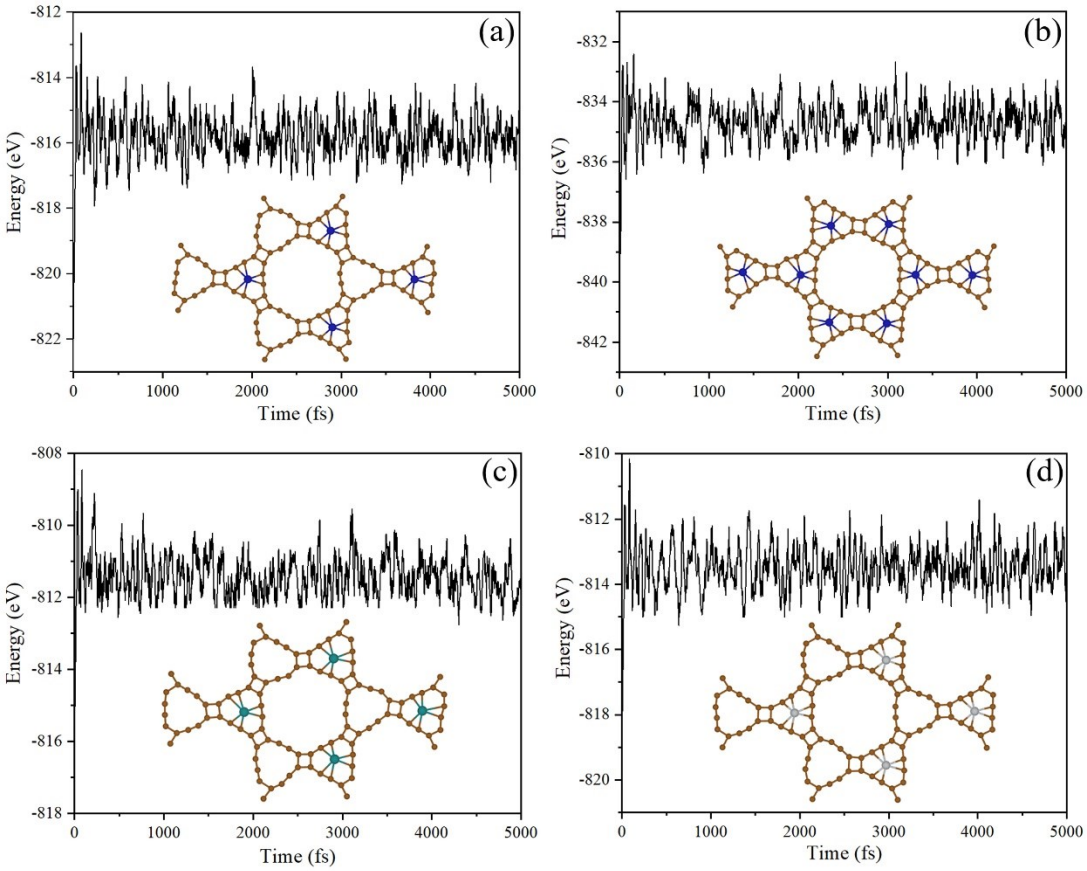


Figure S4. AIMD simulation results for (a) Co@H4,4,4-GY, (b) $Co_2$@H4,4,4-GY, (c) Ni@H4,4,4-GY, and (d) Pt@H4,4,4-GY at 500 K lasting for 5 ps. Insets are snapshots of the final structures of TM@H4,4,4-GY.

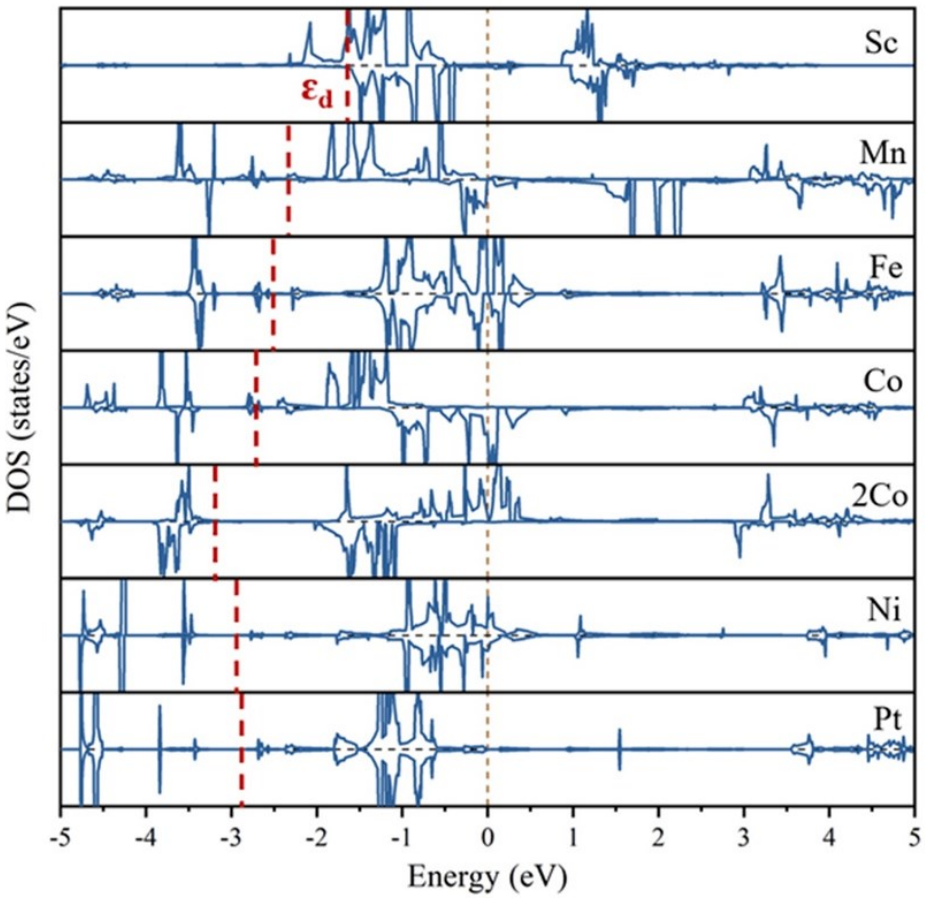


Figure S5. *d* band center of TM atoms supported on H4,4,4-GY, indicated by the red lines. The Fermi level is marked by brown dotted lines.