Electronic Supplementary Information

Acetylene Coupler Builds Strong and Tunable Diradical Organic Molecular Magnets

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Table S1. Detailed energy information of PO-PO radicals under diradical effect.

System	Dihedral Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
PO-≡- PO/	0	-1317.68250911 (2.0505)	-1317.69779782 (0.5648)	-2258.52
	10	-1317.68262476 (2.0509)	-1317.69746807 (0.5801)	-2214.93
	20	-1317.68283122 (2.0521)	-1317.69646061 (0.6227)	-2092.69
	30	-1317.68303309 (2.0539)	-1317.69472868 (0.6861)	-1876.65
	40	-1317.68350220 (2.0563)	-1317.69271546 (0.7730)	-1575.68
	50	-1317.68404536 (2.0584)	-1317.69078391 (0.8601)	-1234.19
	60	-1317.68435309 (2.0603)	-1317.68880554 (0.9412)	-873.20
	70	-1317.68445672 (2.0620)	-1317.68683923 (1.0151)	-499.47
	80	-1317.68487561 (2.0631)	-1317.68591985 (1.0630)	-229.16
	90	-1317.68515712 (2.0633)	-1317.68583983 (1.0761)	-151.78
PO-(≡)$_2$-PO	0	-1393.84253571 (2.0540)	-1393.85314007 (0.7411)	-1772.70
	10	-1393.84267295 (2.0545)	-1393.85297653 (0.7510)	-1734.84
	20	-1393.84293679 (2.0559)	-1393.85242747 (0.7784)	-1630.49
	30	-1393.84324239 (2.0580)	-1393.85141634 (0.8537)	-1489.64
	40	-1393.84383828 (2.0606)	-1393.85037021 (0.8817)	-1216.04
	50	-1393.84447369 (2.0628)	-1393.84940342 (0.9402)	-963.79
	60	-1393.84485487 (2.0648)	-1393.84828690 (0.9970)	-705.41
	70	-1393.84504503 (2.0667)	-1393.84705675 (1.0522)	-435.21

PO-(≡)$_3$-PO	80	-1393.84550106 (2.0677)	-1393.84671991 (1.0838)	-271.88
	90	-1393.84578371 (2.0678)	-1393.84678152 (1.0927)	-224.58
	0	-1470.00265398 (2.0579)	-1470.01042174 (0.8582)	-1421.04
	10	-1470.00278174 (2.0585)	-1470.01034192 (0.8651)	-1390.37
	20	-1470.00301661 (2.0601)	-1470.01001221 (0.8844)	-1305.90
	30	-1470.00328676 (2.0626)	-1470.00936938 (0.9166)	-1164.90
	40	-1470.00382520 (2.0655)	-1470.00878403 (0.9579)	-982.60
	50	-1470.00439790 (2.0681)	-1470.00826508 (0.9999)	-794.55
	60	-1470.00471932 (2.0704)	-1470.00755819 (1.0415)	-605.55
	70	-1470.00487830 (2.0726)	-1470.00671303 (1.0837)	-407.19
	80	-1470.00529348 (2.0736)	-1470.00661176 (1.1061)	-299.04
	90	-1470.00556099 (2.0739)	-1470.00673435 (1.1124)	-267.83

Table S2. Detailed energy information of NN-NN radicals under diradical effect.

System	Dihedral Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
NN-≡-NN	0	-1143.75958359 (2.1109)	-1143.76294618 (1.2164)	-825.05
	10	-1317.68262476 (2.0509)	-1317.69746807 (0.5801)	-798.36
	20	-1317.68283122 (2.0521)	-1317.69646061 (0.6227)	-720.69
	30	-1317.68303309 (2.0539)	-1317.69472868 (0.6861)	-619.89
	40	-1317.68350220 (2.0563)	-1317.69271546 (0.7730)	-482.44
	50	-1317.68404536 (2.0584)	-1317.69078391 (0.8601)	-358.394
	60	-1317.68435309 (2.0603)	-1317.68880554 (0.9412)	-245.594
	70	-1317.68445672 (2.0620)	-1317.68683923 (1.0151)	-155.17
	80	-1317.68487561 (2.0631)	-1317.68591985 (1.0630)	-96.09
	90	-1143.76132242 (2.1505)	-1143.76167538 (1.0761)	-78.68
NN-(≡)$_2$-NN/	0	-1219.91800465 (2.1367)	-1219.92029231 (1.2310)	-554.35
	10	-1393.84267295 (2.0545)	-1393.85297653 (0.7510)	-537.61
	20	-1393.84293679 (2.0559)	-1393.85242747 (0.7784)	-489.61
	30	-1393.84324239 (2.0580)	-1393.85141634 (0.8537)	-432.82
	40	-1393.84383828 (2.0606)	-1393.85037021 (0.8817)	-340.80
	50	-1393.84447369 (2.0628)	-1393.84940342 (0.9402)	-265.56
	60	-1393.84485487 (2.0648)	-1393.84828690 (0.9970)	-193.80
	70	-1393.84504503 (2.0667)	-1393.84705675 (1.0522)	-136.53

NN-(≡)$_3$-NN	80	-1393.84550106 (2.0677)	-1393.84671991 (1.0838)	-95.41
	90	-1219.91896373 (2.1739)	-1219.91934528 (1.1986)	-85.86
	0	-1296.0765836 (2.1567)	-1296.07823967 (1.2435)	-398.01
	10	-1470.00278174 (2.0585)	-1470.01034192 (0.8651)	-388.14
	20	-1470.00301661 (2.0601)	-1470.01001221 (0.8844)	-357.20
	30	-1470.00328676 (2.0626)	-1470.00936938 (0.9166)	-319.01
	40	-1470.00382520 (2.0655)	-1470.00878403 (0.9579)	-260.67
	50	-1470.00439790 (2.0681)	-1470.00826508 (0.9999)	-209.05
	60	-1470.00471932 (2.0704)	-1470.00755819 (1.0415)	-160.03
	70	-1470.00487830 (2.0726)	-1470.00671303 (1.0837)	-121.42
	80	-1470.00529348 (2.0736)	-1470.00661176 (1.1061)	-92.91
	90	-1296.07708901 (2.1891)	-1296.07746963 (1.2209)	-86.28

Table S3. Detailed energy information of PO-NN radicals under diradical effect.

System	Dihedral Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
NN-≡-PO	0	-1230.73256126 (2.1094)	-1230.72327745 (1.0668)	1954.30
	10	-1230.73238710 (2.1098)	-1230.72325670 (1.0676)	1922.75
	20	-1230.73176249 (2.1109)	-1230.72324262 (1.0700)	1796.42
	30	-1230.73074566 (2.1128)	-1230.72307232 (1.0739)	1621.04
	40	-1230.72941905 (2.1149)	-1230.72299575 (1.0792)	1361.15
	50	-1230.72791236 (2.1169)	-1230.72288246 (1.0854)	1070.22
	60	-1230.72649992 (2.1188)	-1230.72298814 (1.0921)	750.70
	70	-1230.72508099 (2.1194)	-1230.72304402 (1.0979)	437.65
	80	-1230.72415270 (2.1199)	-1230.72314180 (1.1024)	218.05
	90	-1230.72372066 (2.1203)	-1230.72318319 (1.1048)	116.16
NN-(≡)$_2$-PO	0	-1306.887375 (2.1402)	-1306.880919 (1.0840)	1341.52
	10	-1306.887288 (2.1406)	-1306.880944 (1.0847)	1318.67
	20	-1306.886972 (2.1413)	-1306.881076 (1.0874)	1227.82
	30	-1306.886420 (2.1426)	-1306.881184 (1.0916)	1093.56
	40	-1306.885720 (2.1442)	-1306.881299 (1.0968)	926.41
	50	-1306.884933 (2.1457)	-1306.881404 (1.1025)	742.40
	60	-1306.884236 (2.1470)	-1306.881766 (1.1089)	522.17
	70	-1306.883513 (2.1476)	-1306.881958 (1.1138)	330.05

System	Dihedral Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
NN-(≡)$_3$-PO	80	-1306.883049 (2.1480)	-1306.882110 (1.1178)	200.05
	90	-1306.882852 (2.1484)	-1306.882185 (1.1197)	142.45
	0	-1383.044653 (2.1673)	-1383.039957 (1.0989)	964.61
	10	-1383.044604 (2.1663)	-1383.039990 (1.0997)	948.38
	20	-1383.044426 (2.1680)	-1383.040139 (1.1023)	882.97
	30	-1383.044087 (2.1688)	-1383.040278 (1.1062)	786.62
	40	-1383.043653 (2.1698)	-1383.040421 (1.1111)	670.06
	50	-1383.043165 (2.1709)	-1383.040565 (1.1168)	541.25
	60	-1383.042787 (2.1718)	-1383.040868 (1.1219)	401.17
	70	-1383.042355 (2.1720)	-1383.041069 (1.1268)	270.16
	80	-1383.042080 (2.1723)	-1383.041177 (1.1298)	190.08
	90	-1383.041969 (2.1726)	-1383.041241 (1.1316)	153.46

Table S4. Detailed energy information of IA-NN radicals under diradical effect.

System	Dihedral Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
IA-≡-NN	0	-1397.94284074 (2.1184)	-1397.93999065 (1.0729)	598.29
	10	-1397.94269194 (2.1178)	-1397.93992097 (1.0732)	582.19
	20	-1397.94229423 (2.1158)	-1397.93974163 (1.0743)	537.90
	30	-1397.94169572 (2.1131)	-1397.93948803 (1.0767)	467.51
	40	-1397.94096721 (2.1097)	-1397.93916770 (1.0786)	383.03
	50	-1397.94028587 (2.1058)	-1397.93895773 (1.0815)	284.57
	60	-1397.93953560 (2.1021)	-1397.93866566 (1.0845)	187.62
	70	-1397.93897511 (2.0991)	-1397.93845498 (1.0868)	112.76
	80	-1397.93869022 (2.0971)	-1397.93841115 (1.0891)	60.76
	90	-1397.93857168 (2.0965)	-1397.93837644 (1.0890)	42.53
IA-(≡)$_2$-NN	0	-1474.09814317 (2.1349)	-1474.09616513 (1.0880)	414.681
	10	-1474.09812900 (2.1342)	-1474.09620934 (1.0887)	402.98
	20	-1474.09793978 (2.1329)	-1474.09616597 (1.0898)	373.22
	30	-1474.09771421 (2.1306)	-1474.09616930 (1.0920)	326.46
	40	-1474.09747767 (2.1277)	-1474.09622644 (1.0947)	265.84
	50	-1474.09714272 (2.1244)	-1474.09618769 (1.0971)	204.03
	60	-1474.09693454 (2.1217)	-1474.09625290 (1.1000)	146.42
	70	-1474.09669572 (2.1195)	-1474.09625975 (1.1024)	94.07

IA-(≡)$_3$-NN	80	-1474.09657410 (2.1179)	-1474.09628622 (1.1039)	62.31
	90	-1474.09650834 (2.1175)	-1474.09627000 (1.1043)	51.62
	0	-1550.25587131 (2.1474)	-1550.25442624 (1.0995)	302.65
	10	-1550.25588295 (2.1467)	-1550.25447945 (1.0998)	294.23
	20	-1550.25577638 (2.1458)	-1550.25447269 (1.1013)	273.93
	30	-1550.25567197 (2.1438)	-1550.25452917 (1.1036)	241.12
	40	-1550.25557644 (2.1413)	-1550.25463546 (1.1060)	199.47
	50	-1550.25540776 (2.1391)	-1550.25468047 (1.1089)	154.94
	60	-1550.25523922 (2.1367)	-1550.25469900 (1.1112)	115.61
	70	-1550.25514203 (2.1346)	-1550.25474802 (1.1128)	84.63
	80	-1550.25512959 (2.1334)	-1550.25484042 (1.1141)	62.26
	90	-1550.25507950 (2.1333)	-1550.25482318 (1.1148)	55.23

Table S5. Detailed energy information of IA-IN radicals under diradical effect.

System	Dihedral Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
IA-≡-IN	0	-1322.77065350 (2.0486)	-1322.76964083 (1.0255)	217.23
	10	-1322.77049431 (2.0482)	-1322.76960009 (1.0255)	191.90
	20	-1322.77018029 (2.0476)	-1322.76934251 (1.0262)	180.01
	30	-1322.76976248 (2.0463)	-1322.76903483 (1.0273)	156.72
	40	-1322.76934867 (2.0448)	-1322.76874763 (1.0289)	129.84
	50	-1322.76884296 (2.0431)	-1322.76839205 (1.0306)	97.74
	60	-1322.76834685 (2.0415)	-1322.76802490 (1.0322)	70.00
	70	-1322.76798063 (2.0403)	-1322.76776853 (1.0337)	46.24
	80	-1322.76777149 (2.0394)	-1322.76763754 (1.0347)	29.26
	90	-1322.76771322 (2.0391)	-1322.76760397 (1.0351)	23.88
IA-(≡)$_2$-IN	0	-1398.92587799 (2.0537)	-1398.92523053 (1.0318)	139.05
	10	-1398.92593652 (2.0533)	-1398.92530842 (1.0320)	134.97
	20	-1398.92575262 (2.0528)	-1398.92516666 (1.0327)	126.06
	30	-1398.92559993 (2.0516)	-1398.92509140 (1.0335)	109.62
	40	-1398.92548031 (2.0506)	-1398.92504865 (1.0349)	93.27
	50	-1398.92529972 (2.0491)	-1398.92496655 (1.0361)	72.18
	60	-1398.92509820 (2.0480)	-1398.92484667 (1.0376)	54.63
	70	-1398.92496868 (2.0469)	-1398.92479422 (1.0389)	37.98

	80	-1398.92488370 (2.0462)	-1398.92475386 (1.0397)	28.31
	90	-1398.92489194 (2.0460)	-1398.92477731 (1.0399)	25.00
	0	-1475.08357597 (2.0578)	-1475.08309786 (1.0367)	102.76
	10	-1475.08361154 (2.0574)	-1475.08314695 (1.0367)	99.89
	20	-1475.08348389 (2.0569)	-1475.08304920 (1.0374)	93.57
	30	-1475.08343199 (2.0560)	-1475.08305013 (1.0382)	82.34
IA-(≡)$_3$-IN	40	-1475.08334164 (2.0552)	-1475.08301015 (1.0392)	71.60
	50	-1475.08325210 (2.0539)	-1475.08298716 (1.0403)	57.36
	60	-1475.08314527 (2.0530)	-1475.08293827 (1.0415)	44.91
	70	-1475.08311942 (2.0521)	-1475.08296579 (1.0425)	33.39
	80	-1475.08308579 (2.0516)	-1475.08296198 (1.0431)	26.94
	90	-1475.08309427 (2.0514)	-1475.08298117 (1.0434)	24.62

Table S6. Detailed energy information of PO-PO diradicals under bending effect.

Diradicals/ Dihedral Angle	Bending Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
PO-≡-PO	140°	-1317.66326753 (2.0500)	-1317.67879444 (0.5700)	-2302.54
	150°	-1317.67155300 (2.0502)	-1317.68704356 (0.5635)	-2286.79
	160°	-1317.67698804 (2.0503)	-1317.69238312 (0.5634)	-2272.39
	170°	-1317.68143010 (2.0505)	-1317.69669825 (0.5658)	-2257.00
	180°	-1317.68250911 (2.0505)	-1317.69779782 (0.5648)	-2258.52
PO-(≡)$_2$-PO	140°	-1393.82923938 (2.0559)	-1393.83942810 (0.7590)	-1724.23
	150°	-1393.83475073 (2.0549)	-1393.84512506 (0.7515)	-1746.89
	160°	-1393.83913467 (2.0543)	-1393.84966269 (0.7444)	-1763.97
	170°	-1393.84180114 (2.0540)	-1393.85239613 (0.7415)	-1771.68
	180°	-1393.84253571 (2.0540)	-1393.85314007 (0.7411)	-1772.70
PO-(≡)$_3$-PO	140°	-1469.99227884 (2.0583)	-1470.00002472 (0.8609)	-1419.762
	150°	-1469.99657039 (2.0581)	-1470.00433466 (0.8594)	-1421.59
	160°	-1470.00011365 (2.0580)	-1470.00787901 (0.8585)	-1420.84
	170°	-1470.00182522 (2.0580)	-1470.00958892 (0.8586)	-1420.65
	180°	-1470.00265398 (2.0579)	-1470.01042174 (0.8582)	-1421.04

Table S7. Detailed energy information of NN-NN diradicals under bending effect.

Diradicals/ Dihedral Angle	Bending Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
NN-≡- NN	140°	-1143.74548457 (2.1129)	-1143.74906889 (1.2339)	-894.95
	150°	-1143.75108650 (2.1119)	-1143.75456827 (1.2258)	-862.38
	160°	-1143.75896823 (2.1109)	-1143.76232662 (1.2159)	-823.55
	170°	-1143.75851756 (2.1109)	-1143.76187313 (1.2158)	-822.77
	180°	-1143.75958359 (2.1109)	-1143.76294618 (1.2164)	-825.04
NN-$(≡)_2$-NN	140°	-1219.90922419 (2.1394)	-1219.91136249 (1.2232)	-517.30
	150°	-1219.91305834 (2.1381)	-1219.91531612 (1.2317)	-546.69
	160°	-1219.91596454 (2.1371)	-1219.91824291 (1.2313)	-552.04
	170°	-1219.91768996 (2.1367)	-1219.91997670 (1.2309)	-554.07
	180°	-1219.91800465 (2.1367)	-1219.92029231 (1.2310)	-554.35
NN–$(≡)_3$-NN/	140°	-1296.06814106 (2.1571)	-1296.06979656 (1.2453)	-398.48
	150°	-1296.07181888 (2.1569)	-1296.07347502 (1.2444)	-398.33
	160°	-1296.07456462 (2.1567)	-1296.07621822 (1.2435)	-397.41
	170°	-1296.07608510 (2.1567)	-1296.07773980 (1.2434)	-397.64
	180°	-1296.07658360 (2.1567)	-1296.07823967 (1.2435)	-398.01

Table S8. Detailed energy information of NN-PO diradicals under bending effect.

Diradicals	Bending Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
NN–≡–PO	140°	-1230.72377822 (2.1104)	-1230.71497736 (1.0670)	1851.22
	150°	-1230.72790335 (2.1097)	-1230.71886996 (1.0669)	1901.22
	160°	-1230.73053246 (2.1094)	-1230.72136389 (1.0668)	1930.04
	170°	-1230.73205606 (2.1094)	-1230.72280818 (1.0668)	1946.74
	180°	-1230.73256126 (2.1094)	-1230.72327745 (1.0668)	1954.30
NN–(≡)$_2$–PO	140°	-1306.87781693 (2.1424)	-1306.87150615 (1.0856)	1310.61
	150°	-1306.88178107 (2.1416)	-1306.87540299 (1.0848)	1324.59
	160°	-1306.88495596 (2.1408)	-1306.87852149 (1.0843)	1336.68
	170°	-1306.88686101 (2.1402)	-1306.88041594 (1.0840)	1339.26
	180°	-1306.88737534 (2.1402)	-1306.88091939 (1.0840)	1341.52
NN–(≡)$_3$–PO	140°	-1383.03682277 (2.1700)	-1383.03210837 (1.0995)	966.54
	150°	-1383.03897445 (2.1687)	-1383.03427561 (1.0990)	964.07
	160°	-1383.04195790 (2.1680)	-1383.03726971 (1.0990)	962.52
	170°	-1383.04398146 (2.1678)	-1383.03929253 (1.0996)	963.39
	180°	-1383.04465287 (2.1673)	-1383.03995715 (1.0989)	964.61

Table S9. Detailed energy information of NN-IA diradicals under bending effect.

Diradicals	Bending Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
IN–≡–NN	140°	-1397.93348605 (2.1159)	-1397.93072459 (1.0711)	580.08
	150°	-1397.93739688 (2.1179)	-1397.93461567 (1.0725)	583.89
	160°	-1397.94076873 (2.1174)	-1397.93796796 (1.0723)	588.17
	170°	-1397.94220571 (2.1182)	-1397.93935556 (1.0726)	598.25
	180°	-1397.94284074 (2.1184)	-1397.93999065 (1.0729)	598.29
IN–(≡)$_2$–NN	140°	-1474.08776015 (2.1366)	-1474.08585441 (1.0898)	399.56
	150°	-1474.09225124 (2.1358)	-1474.09030499 (1.0888)	407.97
	160°	-1474.09548170 (2.1353)	-1474.09350966 (1.0883)	413.38
	170°	-1474.09752478 (2.1348)	-1474.09554901 (1.0879)	414.20
	180°	-1474.09814317 (2.1349)	-1474.09616513 (1.0880)	414.68
IN–(≡)$_3$–NN	140°	-1550.24717698 (2.1490)	-1550.24572697 (1.0995)	303.23
	150°	-1550.25103681 (2.1484)	-1550.24958788 (1.0995)	303.17
	160°	-1550.25390575 (2.1478)	-1550.25245766 (1.0995)	303.17
	170°	-1550.25545071 (2.1472)	-1550.25400691 (1.0991)	302.33
	180°	-1550.25587131 (2.1474)	-1550.25442624 (1.0995)	302.65

Table S10. Detailed energy information of IN-IA diradicals under bending effect.

Diradicals	Bending Angle	E_T (au) (S^2)	E_{BS} (au) (S^2)	J (cm^{-1})
IN–≡–IA	140°	-1322.76020573 (2.0481)	-1322.75930868 (1.0246)	192.35
	150°	-1322.76630280 (2.0480)	-1322.76540324 (1.0247)	192.93
	160°	-1322.76834237 (2.0483)	-1322.76743198 (1.0250)	195.25
	170°	-1322.77004688 (2.0485)	-1322.76912958 (1.0252)	196.74
	180°	-1322.77065350 (2.0486)	-1322.76964083 (1.0255)	217.23
IN–(≡)$_2$–IA	140°	-1398.91542576 (2.0537)	-1398.91480168 (1.0320)	134.06
	150°	-1398.92034396 (2.0538)	-1398.91970942 (1.0320)	136.29
	160°	-1398.92341728 (2.0537)	-1398.92277449 (1.0319)	138.06
	170°	-1398.92539346 (2.0537)	-1398.92474777 (1.0318)	138.67
	180°	-1398.92587799 (2.0537)	-1398.92523053 (1.0318)	139.05
IN–(≡)$_3$–IA	140°	-1475.07531934 (2.0580)	-1475.07484840 (1.0368)	101.21
	150°	-1475.07751310 (2.0579)	-1475.07703792 (1.0367)	102.12
	160°	-1475.07900518 (2.0579)	-1475.07853017 (1.0367)	102.08
	170°	-1475.08310716 (2.0576)	-1475.08263164 (1.0366)	102.21
	180°	-1475.08357597 (2.0578)	-1475.08309786 (1.0367)	102.76

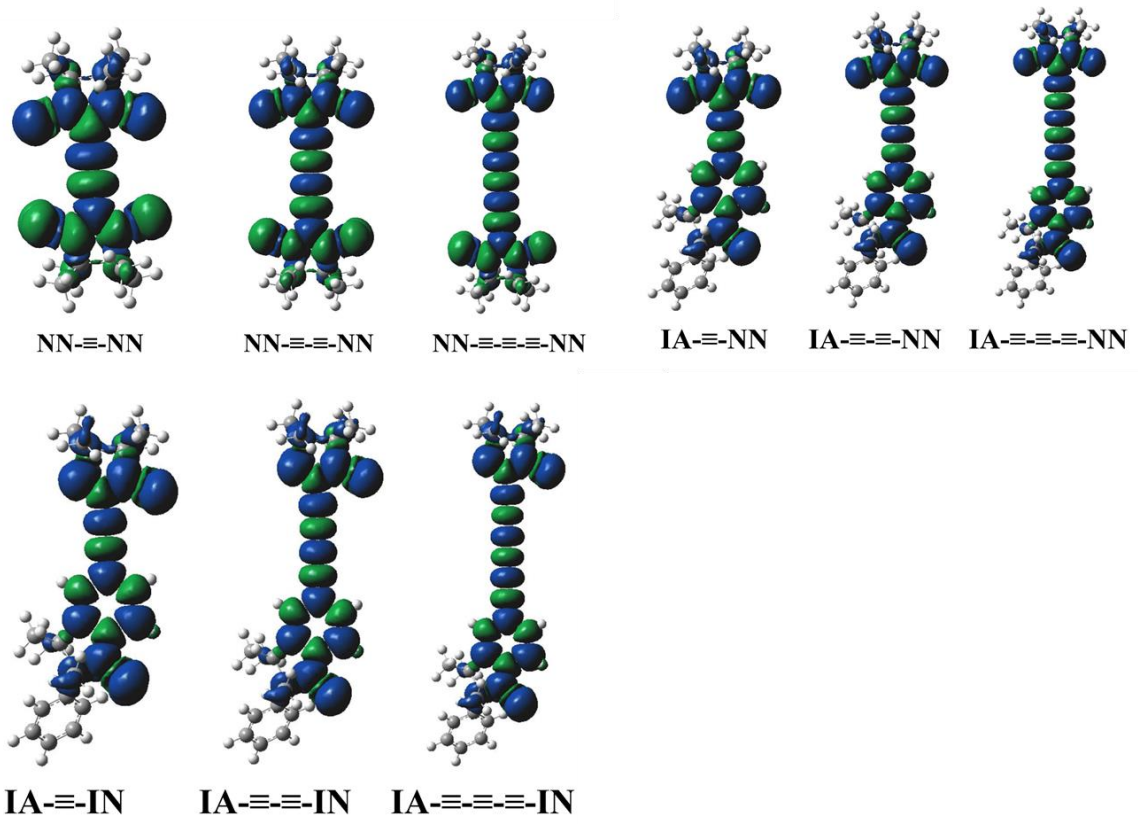

Fig. S1 Calculated spin densities of NN-(≡)$_n$-NN, IA-(≡)$_n$-NN and IA-(≡)$_n$-IN in the ground spin state.

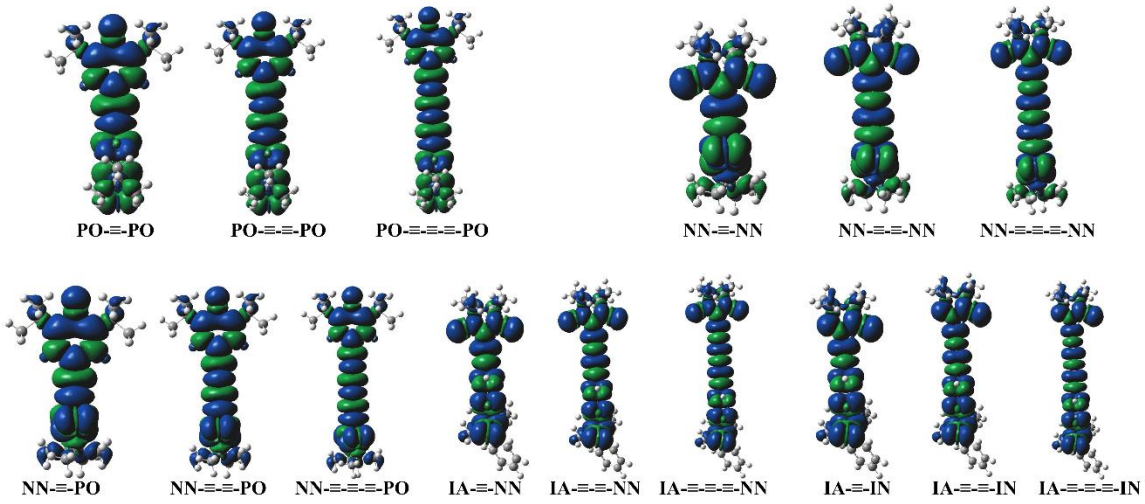

Fig. S2 Calculated spin densities of all OMMs in the ground spin state at perpendicular configurations.