Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.


# Supplementary Information

# for

# Proton, muon and $^{13}C$ hyperfine coupling constants of $C_{60}X$ and $C_{70}X$ (X=H, Mu)

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Optimized geometries, hyperfine constants, and Mulliken spin densities:

**Table S1**. $C_{60}H$

UB3LYP/6-311G(d,p)

| | Cartesian coordinates /Å | | | hfc /MHz | Mulliken spin density (%) |
|---|---|---|---|---|---|
| C(13) | -3.63606 | -0.39043 | -1.04260 | -15.7 | -5.1 |
| C(13) | -2.68786 | -0.61559 | -2.23190 | 18.6 | 6.4 |
| C(13) | -2.05787 | -1.89747 | -2.11760 | -2.7 | -1.2 |
| C(13) | -2.37183 | -2.45589 | -0.81287 | -2.7 | -1.2 |
| C(13) | -3.20015 | -1.52690 | -0.10311 | 18.6 | 6.4 |
| C(13) | -3.00895 | -1.34246 | 1.25016 | -11.2 | -4.8 |
| C(13) | -3.01941 | 0.00756 | 1.83671 | 23.8 | 23.4 |
| C(13) | -3.18589 | 1.11659 | 1.00016 | -27.7 | -13.8 |
| C(13) | -3.39604 | 0.96383 | -0.40511 | 50.1 | 45.8 |
| C(13) | -2.68576 | 2.00597 | -1.07721 | -27.7 | -13.8 |
| C(13) | -2.02620 | 1.77403 | -2.28909 | 23.9 | 23.4 |
| C(13) | -2.02002 | 0.41621 | -2.85755 | -11.2 | -4.8 |
| C(13) | -0.71752 | 0.19293 | -3.44187 | 2.7 | 3.6 |
| C(13) | -0.12210 | -1.06730 | -3.37545 | -2.5 | -1.0 |
| C(13) | -0.80726 | -2.13855 | -2.69756 | 2.1 | 2.5 |
| C(13) | 0.18036 | -2.92948 | -1.98597 | -0.3 | 0.8 |
| C(13) | -0.12044 | -3.46413 | -0.73671 | -0.3 | 0.8 |
| C(13) | -1.42335 | -3.23399 | -0.13941 | 2.1 | 2.5 |
| C(13) | -1.24326 | -3.06079 | 1.27997 | -2.5 | -1.0 |
| C(13) | -2.01813 | -2.11949 | 1.95898 | 2.7 | 3.6 |
| C(13) | -1.41272 | -1.27039 | 2.96633 | 2.6 | 2.8 |
| C(13) | -2.03223 | 0.04173 | 2.88520 | -13.6 | -6.0 |
| C(13) | -1.25587 | 1.18747 | 3.09620 | 10.9 | 11.0 |
| C(13) | -1.43799 | 2.34503 | 2.24045 | -6.1 | -3.5 |
| C(13) | -2.38144 | 2.31227 | 1.22082 | 5.2 | 5.1 |
| C(13) | -2.06924 | 2.86691 | -0.07519 | 5.2 | 5.1 |
| C(13) | -0.82384 | 3.43669 | -0.30989 | -6.1 | -3.5 |
| C(13) | -0.13443 | 3.18124 | -1.56080 | 10.9 | 11.0 |
| C(13) | -0.72986 | 2.35709 | -2.52310 | -13.6 | -6.0 |
| C(13) | 0.08116 | 1.38561 | -3.23755 | 2.6 | 2.8 |
| C(13) | 1.44157 | 1.27529 | -2.96928 | -3.6 | -2.1 |
| C(13) | 2.06037 | -0.03731 | -2.88758 | -1.2 | -0.7 |

| | | | | | |
|---|---|---|---|---|---|
| C(13) | 1.29477 | -1.18362 | -3.08765 | 1.0 | 0.9 |
| C(13) | 1.48312 | -2.33873 | -2.22859 | -0.9 | -0.5 |
| C(13) | 2.43109 | -2.30163 | -1.20883 | 0.5 | 0.2 |
| C(13) | 2.11718 | -2.85941 | 0.09444 | 0.5 | 0.2 |
| C(13) | 0.86834 | -3.43152 | 0.32446 | -0.9 | -0.5 |
| C(13) | 0.17240 | -3.17903 | 1.57328 | 1.0 | 0.9 |
| C(13) | 0.75374 | -2.36031 | 2.53857 | -1.2 | -0.7 |
| C(13) | -0.05696 | -1.38898 | 3.25393 | -3.6 | -2.1 |
| C(13) | 0.74657 | -0.19879 | 3.46448 | 6.8 | 6.7 |
| C(13) | 0.15557 | 1.06240 | 3.38309 | -7.6 | -3.8 |
| C(13) | 0.85123 | 2.14533 | 2.70805 | 1.8 | 1.1 |
| C(13) | -0.13614 | 2.94044 | 2.00173 | 0.1 | -0.3 |
| C(13) | 0.16486 | 3.47543 | 0.75174 | 0.1 | -0.3 |
| C(13) | 1.46623 | 3.23877 | 0.15471 | 1.8 | 1.1 |
| C(13) | 1.27744 | 3.05718 | -1.27510 | -7.6 | -3.8 |
| C(13) | 2.05413 | 2.12603 | -1.96548 | 6.8 | 6.7 |
| C(13) | 3.04141 | 1.33772 | -1.25684 | -4.7 | -2.4 |
| C(13) | 3.21947 | 1.50864 | 0.11386 | 2.9 | 2.2 |
| C(13) | 2.41365 | 2.47880 | 0.83347 | -1.5 | -1.4 |
| C(13) | 2.09987 | 1.92085 | 2.13691 | -1.5 | -1.4 |
| C(13) | 2.71251 | 0.60694 | 2.22027 | 2.9 | 2.2 |
| C(13) | 3.40349 | 0.35377 | 0.97099 | -3.5 | -2.3 |
| C(13) | 3.41103 | -0.92896 | 0.42365 | 4.2 | 3.8 |
| C(13) | 2.72247 | -2.01010 | 1.10210 | -3.4 | -1.9 |
| C(13) | 2.05269 | -1.76650 | 2.30024 | 3.6 | 3.5 |
| C(13) | 2.04764 | -0.42956 | 2.87104 | -4.7 | -2.4 |
| C(13) | 3.04646 | 0.00103 | -1.82804 | 3.6 | 3.5 |
| C(13) | 3.22942 | -1.10838 | -1.00396 | -3.4 | -1.9 |
| H | -4.68844 | -0.50459 | -1.34463 | 94.3 | 3.9 |

**Table S2**. $C_{70}H$ Isomer **A**

UB3LYP/6-311G(d,p)

SCF Energy -2668.3654296 a.u.

| | | Cartesian coordinates /Å | | | hfc /MHz | Mulliken spin density (%) |
|---|---|---|---|---|---|---|
| C(13) | A1 | -4.23690 | -1.23541 | -0.00011 | -15.6 | -5.3 |
| C(13) | A2 | -3.97256 | -0.29010 | -1.18193 | 19.3 | 6.9 |
| C(13) | A3 | -3.91518 | 1.06578 | -0.72393 | -3.6 | -1.5 |
| C(13) | A4 | -3.91517 | 1.06563 | 0.72416 | -3.6 | -1.5 |
| C(13) | A5 | -3.97254 | -0.29033 | 1.18189 | 19.3 | 6.9 |
| C(13) | B1 | -3.24933 | -2.38425 | -0.00024 | 50.5 | 44.0 |
| C(13) | B2 | -3.22006 | -0.66647 | -2.27585 | -9.9 | -4.2 |
| C(13) | B3 | -3.16051 | 2.02295 | -1.41906 | 3.6 | 4.5 |
| C(13) | B4 | -3.16049 | 2.02267 | 1.41948 | 3.6 | 4.5 |
| C(13) | B5 | -3.22004 | -0.66692 | 2.27574 | -9.9 | -4.2 |
| C(13) | C1 | -2.47496 | -2.72931 | 1.14530 | -25.9 | -12.7 |
| C(13) | C2 | -2.47498 | -2.72907 | -1.14580 | -26.0 | -12.7 |
| C(13) | C3 | -2.46055 | -1.91789 | -2.27593 | 21.5 | 22.0 |
| C(13) | C4 | -2.41700 | 0.30260 | -2.97978 | -0.1 | 1.4 |
| C(13) | C5 | -2.39576 | 1.62701 | -2.56792 | -1.0 | 0.0 |
| C(13) | C6 | -2.36454 | 2.98579 | -0.69246 | -1.9 | 0.0 |
| C(13) | C7 | -2.36454 | 2.98566 | 0.69307 | -1.9 | 0.0 |
| C(13) | C8 | -2.39574 | 1.62650 | 2.56825 | -1.0 | 0.0 |
| C(13) | C9 | -2.41698 | 0.30200 | 2.97985 | -0.1 | 1.4 |
| C(13) | C10 | -2.46052 | -1.91834 | 2.27555 | 21.5 | 22.0 |
| C(13) | D1 | -1.24152 | -3.36649 | 0.71116 | 3.3 | 2.9 |
| C(13) | D2 | -1.24153 | -3.36634 | -0.71181 | 3.3 | 2.9 |
| C(13) | D3 | -1.22701 | -1.70262 | -2.98517 | -12.1 | -5.2 |
| C(13) | D4 | -1.19803 | -0.33666 | -3.41998 | 5.8 | 6.7 |
| C(13) | D5 | -1.14398 | 2.35664 | -2.56752 | -1.2 | -0.8 |
| C(13) | D6 | -1.12639 | 3.19989 | -1.41184 | 1.6 | 2.1 |
| C(13) | D7 | -1.12638 | 3.19961 | 1.41249 | 1.6 | 2.1 |
| C(13) | D8 | -1.14396 | 2.35613 | 2.56799 | -1.2 | -0.8 |
| C(13) | D9 | -1.19800 | -0.33734 | 3.41992 | 5.7 | 6.7 |
| C(13) | D10 | -1.22698 | -1.70321 | 2.98485 | -12.1 | -5.2 |
| C(13) | E1 | -0.03845 | -3.25017 | 1.44919 | -2.5 | -1.5 |
| C(13) | E2 | -0.03846 | -3.24988 | -1.44982 | -2.5 | -1.5 |
| C(13) | E3 | -0.02695 | -2.38404 | -2.63475 | 5.5 | 5.7 |

| | | | | | | |
|---|---|---|---|---|---|---|
| C(13) | E4 | 0.02024 | 0.38248 | -3.52428 | -5.1 | -2.8 |
| C(13) | E5 | 0.04815 | 1.78177 | -3.07649 | 1.1 | 0.5 |
| C(13) | E6 | 0.08579 | 3.49472 | -0.73426 | -1.1 | -0.5 |
| C(13) | E7 | 0.08579 | 3.49458 | 0.73495 | -1.1 | -0.5 |
| C(13) | E8 | 0.04817 | 1.78116 | 3.07684 | 1.1 | 0.5 |
| C(13) | E9 | 0.02027 | 0.38178 | 3.52436 | -5.1 | -2.8 |
| C(13) | E10 | -0.02693 | -2.38457 | 2.63429 | 5.5 | 5.7 |
| C(13) | A1' | 3.95780 | -1.30511 | -0.00014 | 0.9 | 0.7 |
| C(13) | A2' | 3.97104 | -0.45241 | -1.17195 | -1.3 | -0.6 |
| C(13) | A3' | 3.99882 | 0.92725 | -0.72387 | 1.3 | 2.2 |
| C(13) | A4' | 3.99882 | 0.92710 | 0.72402 | 1.3 | 2.2 |
| C(13) | A5' | 3.97105 | -0.45265 | 1.17183 | -1.3 | -0.6 |
| C(13) | B1' | 3.19212 | -2.47028 | -0.00025 | -0.6 | -0.5 |
| C(13) | B2' | 3.21608 | -0.80180 | -2.29015 | 0.7 | 0.4 |
| C(13) | B3' | 3.26953 | 1.89413 | -1.41536 | -1.7 | -0.4 |
| C(13) | B4' | 3.26954 | 1.89385 | 1.41571 | -1.6 | -0.4 |
| C(13) | B5' | 3.21610 | -0.80225 | 2.28997 | 0.7 | 0.4 |
| C(13) | C1' | 2.40826 | -2.82518 | 1.16201 | 0.4 | 0.0 |
| C(13) | C2' | 2.40825 | -2.82496 | -1.16258 | 0.4 | 0.0 |
| C(13) | C3' | 2.41769 | -2.00769 | -2.28144 | 0.5 | -0.1 |
| C(13) | C4' | 2.45577 | 0.20492 | -2.99766 | -3.6 | -1.7 |
| C(13) | C5' | 2.48219 | 1.52279 | -2.56959 | 2.6 | 2.8 |
| C(13) | C6' | 2.51341 | 2.89195 | -0.69229 | -0.2 | 0.0 |
| C(13) | C7' | 2.51341 | 2.89181 | 0.69284 | -0.2 | 0.0 |
| C(13) | C8' | 2.48221 | 1.52228 | 2.56987 | 2.6 | 2.8 |
| C(13) | C9' | 2.45580 | 0.20432 | 2.99768 | -3.6 | -1.7 |
| C(13) | C10' | 2.41772 | -2.00814 | 2.28103 | 0.5 | -0.1 |
| C(13) | D1' | 1.16374 | -3.41462 | 0.71571 | -0.1 | -0.9 |
| C(13) | D2' | 1.16373 | -3.41448 | -0.71638 | -0.1 | -0.9 |
| C(13) | D3' | 1.18489 | -1.74975 | -2.99635 | -5.2 | -2.9 |
| C(13) | D4' | 1.20949 | -0.38393 | -3.43659 | 6.1 | 6.1 |
| C(13) | D5' | 1.26349 | 2.30437 | -2.56670 | -1.6 | -1.0 |
| C(13) | D6' | 1.28237 | 3.15037 | -1.41130 | 0.9 | 1.0 |
| C(13) | D7' | 1.28238 | 3.15009 | 1.41191 | 0.9 | 1.0 |
| C(13) | D8' | 1.26351 | 2.30386 | 2.56714 | -1.6 | -1.0 |
| C(13) | D9' | 1.20952 | -0.38461 | 3.43650 | 6.1 | 6.1 |
| C(13) | D10' | 1.18492 | -1.75035 | 2.99600 | -5.2 | -2.9 |
| H | A1-H | -5.27509 | -1.60099 | -0.00013 | 103.3 | 4.3 |

**Table S3** $C_{70}H$ Isomer **B**

UB3LYP/6-311G(d,p)

SCF Energy -2668.3654007 a.u.

| | | Cartesian coordinates /Å | | | hfc /MHz | Mulliken spin density (%) |
|---|---|---|---|---|---|---|
| C(13) | A1 | 4.02933 | -1.08404 | 0.00000 | 55.2 | 46.5 |
| C(13) | A2 | 3.95322 | -0.24483 | 1.15431 | -25.6 | -12.3 |
| C(13) | A3 | 3.91246 | 1.14560 | 0.71944 | 5.6 | 7.4 |
| C(13) | A4 | 3.91246 | 1.14560 | -0.71944 | 5.6 | 7.4 |
| C(13) | A5 | 3.95322 | -0.24483 | -1.15431 | -25.6 | -12.3 |
| C(13) | B1 | 3.47080 | -2.49341 | 0.00000 | -16.2 | -5.3 |
| C(13) | B2 | 3.23058 | -0.63160 | 2.28806 | 18.2 | 17.5 |
| C(13) | B3 | 3.13937 | 2.07235 | 1.41691 | -6.1 | -1.7 |
| C(13) | B4 | 3.13937 | 2.07234 | -1.41691 | -6.1 | -1.7 |
| C(13) | B5 | 3.23058 | -0.63160 | -2.28806 | 18.2 | 17.5 |
| C(13) | C1 | 2.51307 | -2.71348 | -1.17525 | 21.4 | 6.3 |
| C(13) | C2 | 2.51307 | -2.71347 | 1.17525 | 21.4 | 6.3 |
| C(13) | C3 | 2.48227 | -1.88734 | 2.27196 | -8.5 | -3.7 |
| C(13) | C4 | 2.42082 | 0.32931 | 2.98583 | -11.6 | -5.2 |
| C(13) | C5 | 2.37884 | 1.65896 | 2.57023 | 10.8 | 11.8 |
| C(13) | C6 | 2.33089 | 3.02864 | 0.69292 | -1.3 | -0.8 |
| C(13) | C7 | 2.33089 | 3.02864 | -0.69293 | -1.3 | -0.8 |
| C(13) | C8 | 2.37884 | 1.65895 | -2.57023 | 10.8 | 11.8 |
| C(13) | C9 | 2.42082 | 0.32930 | -2.98583 | -11.6 | -5.2 |
| C(13) | C10 | 2.48227 | -1.88735 | -2.27196 | -8.5 | -3.7 |
| C(13) | D1 | 1.30278 | -3.31868 | -0.71797 | -1.6 | -0.6 |
| C(13) | D2 | 1.30278 | -3.31867 | 0.71798 | -1.6 | -0.6 |
| C(13) | D3 | 1.24291 | -1.67948 | 2.98483 | 0.6 | 0.7 |
| C(13) | D4 | 1.20390 | -0.31931 | 3.42488 | 1.7 | 1.1 |
| C(13) | D5 | 1.11918 | 2.36745 | 2.56686 | -6.8 | -3.0 |
| C(13) | D6 | 1.08913 | 3.21677 | 1.41185 | 4.1 | 4.2 |
| C(13) | D7 | 1.08913 | 3.21677 | -1.41185 | 4.1 | 4.2 |
| C(13) | D8 | 1.11918 | 2.36745 | -2.56687 | -6.8 | -3.0 |
| C(13) | D9 | 1.20390 | -0.31932 | -3.42488 | 1.7 | 1.1 |
| C(13) | D10 | 1.24291 | -1.67949 | -2.98483 | 0.6 | 0.7 |
| C(13) | E1 | 0.08518 | -3.22983 | -1.45330 | 1.3 | 1.8 |
| C(13) | E2 | 0.08518 | -3.22982 | 1.45331 | 1.3 | 1.8 |
| C(13) | E3 | 0.05794 | -2.37537 | 2.63775 | -0.4 | -0.3 |

| | | | | | | |
|---|---|---|---|---|---|---|
| C(13) | E4 | -0.02346 | 0.38459 | 3.52879 | -1.7 | -1.2 |
| C(13) | E5 | -0.06797 | 1.78201 | 3.07850 | 2.9 | 3.0 |
| C(13) | E6 | -0.12527 | 3.49444 | 0.73484 | -3.0 | -1.8 |
| C(13) | E7 | -0.12527 | 3.49444 | -0.73485 | -3.0 | -1.8 |
| C(13) | E8 | -0.06797 | 1.78200 | -3.07851 | 2.9 | 3.0 |
| C(13) | E9 | -0.02346 | 0.38459 | -3.52879 | -1.7 | -1.2 |
| C(13) | E10 | 0.05794 | -2.37537 | -2.63774 | -0.4 | -0.3 |
| C(13) | A1' | -3.93492 | -1.35230 | 0.00000 | -0.1 | 0.0 |
| C(13) | A2' | -3.96088 | -0.49893 | 1.17273 | -0.1 | 0.0 |
| C(13) | A3' | -4.00682 | 0.87867 | 0.72401 | 0.0 | 0.5 |
| C(13) | A4' | -4.00682 | 0.87867 | -0.72401 | 0.0 | 0.5 |
| C(13) | A5' | -3.96088 | -0.49893 | -1.17273 | -0.1 | 0.0 |
| C(13) | B1' | -3.15162 | -2.50373 | 0.00000 | 0.5 | 0.3 |
| C(13) | B2' | -3.20590 | -0.83828 | 2.29353 | 0.0 | -0.1 |
| C(13) | B3' | -3.29007 | 1.85507 | 1.41627 | 0.3 | 0.8 |
| C(13) | B4' | -3.29007 | 1.85507 | -1.41627 | 0.3 | 0.8 |
| C(13) | B5' | -3.20590 | -0.83829 | -2.29353 | 0.0 | -0.1 |
| C(13) | C1' | -2.36365 | -2.84861 | -1.16385 | -0.5 | -0.4 |
| C(13) | C2' | -2.36365 | -2.84861 | 1.16385 | -0.5 | -0.4 |
| C(13) | C3' | -2.38986 | -2.03259 | 2.28500 | 0.6 | 0.3 |
| C(13) | C4' | -2.45834 | 0.17816 | 3.00154 | -0.6 | -0.6 |
| C(13) | C5' | -2.49944 | 1.49431 | 2.57099 | 0.5 | 0.2 |
| C(13) | C6' | -2.54506 | 2.86122 | 0.69285 | -1.3 | 0.2 |
| C(13) | C7' | -2.54506 | 2.86122 | -0.69286 | -1.3 | 0.2 |
| C(13) | C8' | -2.49944 | 1.49430 | -2.57099 | 0.5 | 0.2 |
| C(13) | C9' | -2.45834 | 0.17816 | -3.00154 | -0.6 | -0.6 |
| C(13) | C10' | -2.38986 | -2.03259 | -2.28499 | 0.6 | 0.3 |
| C(13) | D1' | -1.11041 | -3.41398 | -0.71778 | -0.1 | 0.6 |
| C(13) | D2' | -1.11041 | -3.41398 | 0.71779 | -0.1 | 0.6 |
| C(13) | D3' | -1.16562 | -1.75766 | 3.00325 | -0.8 | -0.9 |
| C(13) | D4' | -1.20508 | -0.39537 | 3.44314 | 0.9 | 0.5 |
| C(13) | D5' | -1.28914 | 2.28994 | 2.56779 | -2.8 | -1.3 |
| C(13) | D6' | -1.31864 | 3.13506 | 1.41221 | 3.8 | 4.1 |
| C(13) | D7' | -1.31864 | 3.13506 | -1.41222 | 3.8 | 4.1 |
| C(13) | D8' | -1.28914 | 2.28993 | -2.56779 | -2.8 | -1.3 |
| C(13) | D9' | -1.20508 | -0.39537 | -3.44314 | 0.9 | 0.5 |
| C(13) | D10' | -1.16562 | -1.75767 | -3.00324 | -0.8 | -0.9 |
| H | B1-H | 4.27536 | -3.24530 | 0.00000 | 104.5 | 4.3 |

**Table S4** $C_{70}H$ Isomer **C**

UB3LYP/6-311G(d,p)

SCF Energy -2668.3641965 a.u.

| | | Cartesian coordinates /Å | | | hfc /MHz | Mulliken spin density (%) |
|---|---|---|---|---|---|---|
| C(13) | A1 | 3.98972 | 1.01668 | 0.42506 | -9.3 | -3.7 |
| C(13) | A2 | 3.98602 | 0.57156 | -0.97685 | 18.3 | 19.0 |
| C(13) | A3 | 3.92457 | -0.86840 | -0.98408 | -10.2 | -3.2 |
| C(13) | A4 | 3.89219 | -1.32527 | 0.39144 | 1.1 | 1.4 |
| C(13) | A5 | 3.93273 | -0.16388 | 1.25559 | 1.9 | 2.4 |
| C(13) | B1 | 3.30922 | 2.15842 | 0.79906 | 18.0 | 6.0 |
| C(13) | B2 | 3.25431 | 1.29802 | -1.91836 | -23.1 | -10.9 |
| C(13) | B3 | 3.16175 | -1.53849 | -1.94976 | 8.2 | 10.0 |
| C(13) | B4 | 3.10972 | -2.42417 | 0.74289 | -1.6 | -1.2 |
| C(13) | B5 | 3.18889 | -0.15270 | 2.43742 | -1.6 | -0.8 |
| C(13) | C1 | 2.50281 | 2.14408 | 1.97672 | -2.0 | -0.6 |
| C(13) | C2 | 2.69577 | 3.13395 | -0.21039 | -17.3 | -4.9 |
| C(13) | C3 | 2.53899 | 2.48151 | -1.55632 | 44.1 | 43.0 |
| C(13) | C4 | 2.46185 | 0.60051 | -2.91533 | -1.8 | 1.6 |
| C(13) | C5 | 2.41523 | -0.78264 | -2.92825 | -0.2 | 2.4 |
| C(13) | C6 | 2.33944 | -2.66263 | -1.57368 | -6.7 | -2.5 |
| C(13) | C7 | 2.31389 | -3.09831 | -0.25773 | 2.5 | 2.1 |
| C(13) | C8 | 2.32637 | -2.39831 | 1.95905 | 0.0 | -0.1 |
| C(13) | C9 | 2.36533 | -1.28993 | 2.78866 | 0.1 | 0.0 |
| C(13) | C10 | 2.45807 | 1.02668 | 2.80498 | 1.8 | 2.3 |
| C(13) | D1 | 1.29347 | 2.91063 | 1.73343 | -1.8 | -1.2 |
| C(13) | D2 | 1.32411 | 3.40089 | 0.41343 | 16.6 | 4.8 |
| C(13) | D3 | 1.31769 | 2.51674 | -2.27507 | -25.5 | -12.9 |
| C(13) | D4 | 1.26489 | 1.36330 | -3.14338 | 14.8 | 16.1 |
| C(13) | D5 | 1.15594 | -1.45344 | -3.17356 | -5.3 | -3.3 |
| C(13) | D6 | 1.10819 | -2.61507 | -2.33901 | 6.9 | 7.5 |
| C(13) | D7 | 1.06012 | -3.49782 | 0.34389 | -1.7 | -1.0 |
| C(13) | D8 | 1.06787 | -3.06768 | 1.70995 | 0.8 | 0.6 |
| C(13) | D9 | 1.14424 | -0.80414 | 3.39528 | -0.2 | 0.3 |
| C(13) | D10 | 1.19906 | 0.62457 | 3.39832 | 0.0 | 1.1 |
| C(13) | E1 | 0.07060 | 2.59595 | 2.40082 | 1.8 | 1.6 |
| C(13) | E2 | 0.13349 | 3.53271 | -0.33667 | -6.4 | -2.8 |
| C(13) | E3 | 0.12095 | 3.08795 | -1.74465 | 11.8 | 11.7 |

| | | | | | | |
|---|---|---|---|---|---|---|
| C(13) | E4 | 0.03754 | 0.74155 | -3.47427 | -12.3 | -6.7 |
| C(13) | E5 | -0.01974 | -0.72602 | -3.49226 | 3.5 | 1.8 |
| C(13) | E6 | -0.11837 | -3.08246 | -1.80124 | -5.5 | -3.2 |
| C(13) | E7 | -0.14339 | -3.54353 | -0.40656 | 1.9 | 1.4 |
| C(13) | E8 | -0.12681 | -2.67012 | 2.36401 | -0.9 | -0.7 |
| C(13) | E9 | -0.08818 | -1.49209 | 3.23959 | 0.3 | 0.4 |
| C(13) | E10 | 0.02398 | 1.40608 | 3.25643 | -0.6 | 0.1 |
| C(13) | A1' | -3.92622 | 1.30891 | 0.37159 | 0.3 | 0.1 |
| C(13) | A2' | -3.93074 | 0.87324 | -1.01140 | -2.1 | -1.4 |
| C(13) | A3' | -3.99094 | -0.57548 | -1.02591 | 3.2 | 3.3 |
| C(13) | A4' | -4.01742 | -1.03472 | 0.34869 | -1.7 | -0.7 |
| C(13) | A5' | -3.97852 | 0.12997 | 1.21222 | 0.5 | 0.4 |
| C(13) | B1' | -3.13928 | 2.39580 | 0.75093 | 0.1 | -0.2 |
| C(13) | B2' | -3.14944 | 1.54327 | -1.95097 | 1.7 | 1.1 |
| C(13) | B3' | -3.26594 | -1.28796 | -1.97986 | -4.7 | -2.1 |
| C(13) | B4' | -3.31686 | -2.18487 | 0.70752 | 0.5 | 1.1 |
| C(13) | B5' | -3.24099 | 0.08936 | 2.39451 | -0.5 | -0.4 |
| C(13) | C1' | -2.37422 | 2.34572 | 1.97634 | 0.6 | 0.2 |
| C(13) | C2' | -2.32478 | 3.08039 | -0.22809 | -1.4 | -1.5 |
| C(13) | C3' | -2.32915 | 2.66420 | -1.54940 | 2.4 | 1.3 |
| C(13) | C4' | -2.39531 | 0.79790 | -2.93524 | -10.3 | -5.4 |
| C(13) | C5' | -2.45586 | -0.59001 | -2.95188 | 8.8 | 9.1 |
| C(13) | C6' | -2.53681 | -2.47856 | -1.59933 | -1.1 | 0.2 |
| C(13) | C7' | -2.56100 | -2.91641 | -0.28606 | 0.8 | 1.4 |
| C(13) | C8' | -2.54775 | -2.21787 | 1.93121 | -0.8 | -0.2 |
| C(13) | C9' | -2.51000 | -1.10462 | 2.75633 | 0.6 | 0.7 |
| C(13) | C10' | -2.42128 | 1.21754 | 2.77815 | -0.4 | -0.5 |
| C(13) | D1' | -1.10889 | 3.01672 | 1.75555 | -1.7 | -1.2 |
| C(13) | D2' | -1.07896 | 3.46672 | 0.39504 | 2.1 | 1.6 |
| C(13) | D3' | -1.09090 | 2.62580 | -2.29698 | -12.2 | -6.5 |
| C(13) | D4' | -1.13838 | 1.46731 | -3.15858 | 16.0 | 16.2 |
| C(13) | D5' | -1.25123 | -1.35727 | -3.18346 | -5.9 | -3.5 |
| C(13) | D6' | -1.29947 | -2.52164 | -2.35074 | 4.7 | 4.8 |
| C(13) | D7' | -1.34688 | -3.40966 | 0.33051 | -1.5 | -1.0 |
| C(13) | D8' | -1.33968 | -2.97975 | 1.69660 | 0.9 | 0.8 |
| C(13) | D9' | -1.26434 | -0.71379 | 3.38085 | -0.1 | 0.2 |
| C(13) | D10' | -1.20853 | 0.71786 | 3.39011 | 0.0 | 0.1 |
| H | C2-H | 3.28346 | 4.06332 | -0.28114 | 99.5 | 4.2 |

**Table S5** $C_{70}H$ Isomer **D**

UB3LYP/6-311G(d,p)

SCF Energy -2668.363796 a.u.

| | | Cartesian coordinates /Å | | | hfc /MHz | Mulliken spin density (%) |
|---|---|---|---|---|---|---|
| C(13) | A1 | -3.97533 | 1.07149 | -0.37336 | -2.4 | -0.6 |
| C(13) | A2 | -3.96237 | 0.67672 | 1.01101 | 1.3 | 1.4 |
| C(13) | A3 | -3.93585 | -0.77318 | 1.06428 | -1.3 | -0.7 |
| C(13) | A4 | -3.94212 | -1.27323 | -0.29648 | -0.7 | 1.4 |
| C(13) | A5 | -3.96160 | -0.13211 | -1.18710 | 3.5 | 4.6 |
| C(13) | B1 | -3.23647 | 2.19011 | -0.77297 | 1.7 | 1.6 |
| C(13) | B2 | -3.21012 | 1.41725 | 1.92361 | -1.1 | -0.3 |
| C(13) | B3 | -3.16417 | -1.41320 | 2.03160 | 0.7 | 0.4 |
| C(13) | B4 | -3.17916 | -2.39474 | -0.62665 | -0.3 | 1.9 |
| C(13) | B5 | -3.22780 | -0.16273 | -2.37027 | -6.3 | -2.7 |
| C(13) | C1 | -2.46200 | 2.14712 | -1.99951 | -14.4 | -7.9 |
| C(13) | C2 | -2.51006 | 2.97550 | 0.17601 | 10.0 | 2.5 |
| C(13) | C3 | -2.46946 | 2.57807 | 1.49136 | -3.2 | -1.3 |
| C(13) | C4 | -2.40879 | 0.74497 | 2.91792 | 2.6 | 2.3 |
| C(13) | C5 | -2.38697 | -0.63809 | 2.97491 | -2.6 | -1.9 |
| C(13) | C6 | -2.37125 | -2.56980 | 1.68259 | 0.8 | 0.5 |
| C(13) | C7 | -2.37852 | -3.04938 | 0.38199 | -2.5 | -0.9 |
| C(13) | C8 | -2.41596 | -2.42160 | -1.85459 | 4.9 | 6.2 |
| C(13) | C9 | -2.44098 | -1.32935 | -2.71104 | -5.1 | -2.0 |
| C(13) | C10 | -2.47402 | 0.99376 | -2.79329 | 13.4 | 13.7 |
| C(13) | D1 | -1.28210 | 2.91268 | -1.84600 | 33.5 | 24.1 |
| C(13) | D2 | -1.33656 | 3.67277 | -0.50493 | -13.5 | -4.0 |
| C(13) | D3 | -1.21423 | 2.61961 | 2.24241 | 7.6 | 8.0 |
| C(13) | D4 | -1.18467 | 1.49800 | 3.11325 | -5.4 | -2.7 |
| C(13) | D5 | -1.13825 | -1.32897 | 3.22024 | 2.7 | 2.1 |
| C(13) | D6 | -1.12833 | -2.51713 | 2.42319 | -2.4 | -1.8 |
| C(13) | D7 | -1.14246 | -3.49607 | -0.22720 | 4.5 | 4.5 |
| C(13) | D8 | -1.16640 | -3.10589 | -1.60626 | -5.3 | -3.3 |
| C(13) | D9 | -1.22671 | -0.88950 | -3.35424 | 10.4 | 10.4 |
| C(13) | D10 | -1.25214 | 0.55116 | -3.39651 | -11.0 | -6.4 |
| C(13) | E1 | -0.08091 | 2.53703 | -2.47223 | -20.2 | -10.8 |
| C(13) | E2 | -0.02874 | 3.55052 | 0.27378 | 19.7 | 16.8 |
| C(13) | E3 | -0.01842 | 3.12731 | 1.66070 | -10.0 | -4.5 |

| | | | | | | |
|---|---|---|---|---|---|---|
| C(13) | E4 | 0.03708 | 0.84407 | 3.45296 | 3.8 | 3.7 |
| C(13) | E5 | 0.05830 | -0.62173 | 3.50396 | -2.8 | -1.5 |
| C(13) | E6 | 0.07889 | -3.03113 | 1.88232 | 2.6 | 1.7 |
| C(13) | E7 | 0.07022 | -3.53724 | 0.50377 | -5.2 | -2.9 |
| C(13) | E8 | 0.02404 | -2.74660 | -2.28960 | 3.5 | 2.1 |
| C(13) | E9 | -0.00798 | -1.59230 | -3.19679 | -10.2 | -5.7 |
| C(13) | E10 | -0.06272 | 1.31237 | -3.28393 | 7.0 | 4.3 |
| C(13) | A1' | 3.92852 | 1.21536 | -0.46093 | -4.2 | -2.7 |
| C(13) | A2' | 3.96184 | 0.81537 | 0.93213 | 1.2 | 0.6 |
| C(13) | A3' | 3.98849 | -0.63344 | 0.97834 | -0.5 | -0.3 |
| C(13) | A4' | 3.98077 | -1.12783 | -0.38497 | -2.3 | 0.0 |
| C(13) | A5' | 3.94479 | 0.01235 | -1.27515 | 5.0 | 5.5 |
| C(13) | B1' | 3.15247 | 2.30347 | -0.84992 | 3.6 | 2.8 |
| C(13) | B2' | 3.22248 | 1.52531 | 1.87577 | -0.7 | -0.8 |
| C(13) | B3' | 3.26852 | -1.30665 | 1.96397 | 0.2 | 0.1 |
| C(13) | B4' | 3.25226 | -2.27590 | -0.70033 | 1.4 | 3.0 |
| C(13) | B5' | 3.18408 | -0.04100 | -2.44224 | -6.9 | -2.8 |
| C(13) | C1' | 2.35830 | 2.24794 | -2.06369 | -18.3 | -9.2 |
| C(13) | C2' | 2.38288 | 3.02988 | 0.13006 | 2.3 | 1.7 |
| C(13) | C3' | 2.41905 | 2.65534 | 1.46458 | -1.8 | -1.5 |
| C(13) | C4' | 2.46966 | 0.82079 | 2.89016 | 1.7 | 0.9 |
| C(13) | C5' | 2.49103 | -0.56443 | 2.93183 | -1.5 | -1.3 |
| C(13) | C6' | 2.50657 | -2.48882 | 1.62714 | 1.3 | 0.9 |
| C(13) | C7' | 2.49906 | -2.96147 | 0.32480 | -3.6 | -1.4 |
| C(13) | C8' | 2.46212 | -2.32637 | -1.90957 | 3.2 | 4.2 |
| C(13) | C9' | 2.42863 | -1.23437 | -2.76265 | -5.0 | -1.6 |
| C(13) | C10' | 2.38015 | 1.08899 | -2.84412 | 14.9 | 15.4 |
| C(13) | D1' | 1.13710 | 2.95948 | -1.85578 | 29.8 | 28.9 |
| C(13) | D2' | 1.13626 | 3.44255 | -0.47170 | -17.2 | -8.4 |
| C(13) | D3' | 1.19814 | 2.65550 | 2.22482 | 4.2 | 3.5 |
| C(13) | D4' | 1.22531 | 1.52624 | 3.11041 | -3.8 | -2.6 |
| C(13) | D5' | 1.27062 | -1.29369 | 3.19862 | 2.2 | 1.6 |
| C(13) | D6' | 1.28013 | -2.48009 | 2.39617 | -2.3 | -2.0 |
| C(13) | D7' | 1.26591 | -3.45053 | -0.25764 | 6.5 | 7.1 |
| C(13) | D8' | 1.24280 | -3.05836 | -1.63277 | -5.4 | -3.2 |
| C(13) | D9' | 1.18254 | -0.83829 | -3.37757 | 12.7 | 13.5 |
| C(13) | D10' | 1.15379 | 0.59619 | -3.41709 | -12.7 | -7.5 |
| H | D2-H | -1.55522 | 4.73954 | -0.66963 | 81.3 | 3.5 |

**Table S6** $C_{70}H$ Isomer **E**

UB3LYP/6-311G(d,p)

SCF Energy -2668.3454838 a.u.

| | | Cartesian coordinates /Å | | | hfc /MHz | Mulliken spin density (%) |
|---|---|---|---|---|---|---|
| C(13) | A1 | -3.96846 | 1.11059 | -0.49306 | -3.5 | -1.8 |
| C(13) | A2 | -3.96926 | 0.79834 | 0.92116 | 6.3 | 6.5 |
| C(13) | A3 | -3.96540 | -0.64392 | 1.06172 | -4.0 | -1.9 |
| C(13) | A4 | -3.96651 | -1.22423 | -0.26789 | 1.1 | 0.5 |
| C(13) | A5 | -3.96812 | -0.13817 | -1.22877 | -0.4 | -0.8 |
| C(13) | B1 | -3.22676 | 2.19215 | -0.96242 | 3.3 | 3.4 |
| C(13) | B2 | -3.21354 | 1.58302 | 1.79520 | -8.0 | -3.6 |
| C(13) | B3 | -3.21909 | -1.24295 | 2.07547 | 3.4 | 3.3 |
| C(13) | B4 | -3.21901 | -2.37183 | -0.52299 | -0.8 | -0.9 |
| C(13) | B5 | -3.22139 | -0.25035 | -2.39981 | 0.4 | -0.1 |
| C(13) | C1 | -2.44412 | 2.06138 | -2.16352 | 5.7 | 4.5 |
| C(13) | C2 | -2.44845 | 3.01136 | -0.05037 | -18.6 | -8.7 |
| C(13) | C3 | -2.44548 | 2.70085 | 1.30438 | 14.8 | 14.0 |
| C(13) | C4 | -2.43401 | 0.95090 | 2.83822 | 0.9 | 1.2 |
| C(13) | C5 | -2.43887 | -0.42734 | 2.98003 | -0.8 | -0.5 |
| C(13) | C6 | -2.44020 | -2.43026 | 1.80248 | -2.6 | -1.4 |
| C(13) | C7 | -2.43993 | -2.98327 | 0.53173 | 2.3 | 1.6 |
| C(13) | C8 | -2.44003 | -2.47886 | -1.73773 | -2.3 | -2.0 |
| C(13) | C9 | -2.44065 | -1.44076 | -2.65480 | 2.7 | 1.8 |
| C(13) | C10 | -2.44035 | 0.87217 | -2.86871 | -5.4 | -3.7 |
| C(13) | D1 | -1.20976 | 2.80721 | -1.98220 | -24.1 | -13.5 |
| C(13) | D2 | -1.23821 | 3.42104 | -0.71215 | 38.0 | 33.3 |
| C(13) | D3 | -1.19343 | 2.75789 | 2.05270 | -9.1 | -3.8 |
| C(13) | D4 | -1.19979 | 1.68296 | 2.99809 | 2.3 | 2.1 |
| C(13) | D5 | -1.20543 | -1.12241 | 3.28082 | -1.2 | -0.9 |
| C(13) | D6 | -1.20568 | -2.35567 | 2.55470 | 1.7 | 1.4 |
| C(13) | D7 | -1.20548 | -3.47932 | -0.03570 | -3.9 | -2.7 |
| C(13) | D8 | -1.20515 | -3.17003 | -1.43332 | 3.1 | 2.4 |
| C(13) | D9 | -1.20395 | -1.05125 | -3.29996 | -6.9 | -4.7 |
| C(13) | D10 | -1.20383 | 0.37268 | -3.43083 | 8.4 | 8.0 |
| C(13) | E1 | 0.00001 | 2.38434 | -2.61117 | 23.0 | 24.4 |
| C(13) | E2 | 0.00000 | 3.85637 | 0.05536 | -22.8 | -7.5 |
| C(13) | E3 | 0.00000 | 3.23054 | 1.48890 | 38.7 | 10.2 |

| | | | | | | |
|---|---|---|---|---|---|---|
| C(13) | E4 | -0.00001 | 1.03983 | 3.38879 | -1.6 | -1.0 |
| C(13) | E5 | -0.00001 | -0.41826 | 3.53474 | 1.5 | 1.2 |
| C(13) | E6 | 0.00000 | -2.91804 | 2.05947 | -2.7 | -1.4 |
| C(13) | E7 | 0.00000 | -3.50208 | 0.71422 | 4.5 | 4.5 |
| C(13) | E8 | 0.00001 | -2.87242 | -2.12091 | -3.2 | -1.9 |
| C(13) | E9 | 0.00001 | -1.77278 | -3.09170 | 5.5 | 4.9 |
| C(13) | E10 | 0.00001 | 1.12270 | -3.35047 | -13.8 | -6.0 |
| C(13) | A1' | 3.96846 | 1.11059 | -0.49305 | -3.5 | -1.8 |
| C(13) | A2' | 3.96925 | 0.79834 | 0.92117 | 6.3 | 6.5 |
| C(13) | A3' | 3.96539 | -0.64391 | 1.06174 | -4.0 | -1.9 |
| C(13) | A4' | 3.96651 | -1.22423 | -0.26787 | 1.1 | 0.5 |
| C(13) | A5' | 3.96812 | -0.13817 | -1.22875 | -0.4 | -0.8 |
| C(13) | B1' | 3.22675 | 2.19215 | -0.96241 | 3.3 | 3.4 |
| C(13) | B2' | 3.21353 | 1.58302 | 1.79522 | -8.0 | -3.6 |
| C(13) | B3' | 3.21908 | -1.24295 | 2.07548 | 3.4 | 3.3 |
| C(13) | B4' | 3.21902 | -2.37183 | -0.52298 | -0.8 | -0.9 |
| C(13) | B5' | 3.22140 | -0.25035 | -2.39980 | 0.4 | -0.1 |
| C(13) | C1' | 2.44412 | 2.06138 | -2.16351 | 5.7 | 4.5 |
| C(13) | C2' | 2.44844 | 3.01136 | -0.05036 | -18.6 | -8.7 |
| C(13) | C3' | 2.44548 | 2.70085 | 1.30440 | 14.8 | 14.0 |
| C(13) | C4' | 2.43399 | 0.95091 | 2.83823 | 0.9 | 1.2 |
| C(13) | C5' | 2.43886 | -0.42733 | 2.98004 | -0.8 | -0.5 |
| C(13) | C6' | 2.44020 | -2.43026 | 1.80249 | -2.6 | -1.4 |
| C(13) | C7' | 2.43993 | -2.98327 | 0.53175 | 2.3 | 1.6 |
| C(13) | C8' | 2.44004 | -2.47886 | -1.73772 | -2.3 | -2.0 |
| C(13) | C9' | 2.44066 | -1.44076 | -2.65480 | 2.7 | 1.8 |
| C(13) | C10' | 2.44036 | 0.87216 | -2.86871 | -5.4 | -3.7 |
| C(13) | D1' | 1.20977 | 2.80721 | -1.98221 | -24.1 | -13.5 |
| C(13) | D2' | 1.23822 | 3.42105 | -0.71214 | 38.0 | 33.3 |
| C(13) | D3' | 1.19342 | 2.75789 | 2.05271 | -9.1 | -3.8 |
| C(13) | D4' | 1.19977 | 1.68296 | 2.99809 | 2.3 | 2.1 |
| C(13) | D5' | 1.20542 | -1.12241 | 3.28082 | -1.2 | -0.9 |
| C(13) | D6' | 1.20567 | -2.35567 | 2.55471 | 1.7 | 1.4 |
| C(13) | D7' | 1.20548 | -3.47932 | -0.03570 | -3.9 | -2.7 |
| C(13) | D8' | 1.20516 | -3.17003 | -1.43331 | 3.1 | 2.4 |
| C(13) | D9' | 1.20397 | -1.05126 | -3.29995 | -6.9 | -4.7 |
| C(13) | D10' | 1.20385 | 0.37268 | -3.43083 | 8.4 | 8.0 |
| H | E2-H | -0.00001 | 4.95555 | 0.16536 | 158.3 | 6.6 |