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

Spreading out spin density in polyphenalenyl radicals

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Supplementary Information

I. Breaking down of π spin density per phenalene unit along L15 (Figure S1).

II. Plot of π spin densities in S10 and S'10 systems (Figure S2), with atom labeling defined in Figure S3.

III. List of optimized CC bond lengths in neutral and ionized P19 (Table S1), with bond labeling defined in Figure S4, and further ranking and visualization in Figure S5.

IV. Selected harmonic vibrational frequencies in **8** (Figure S6).

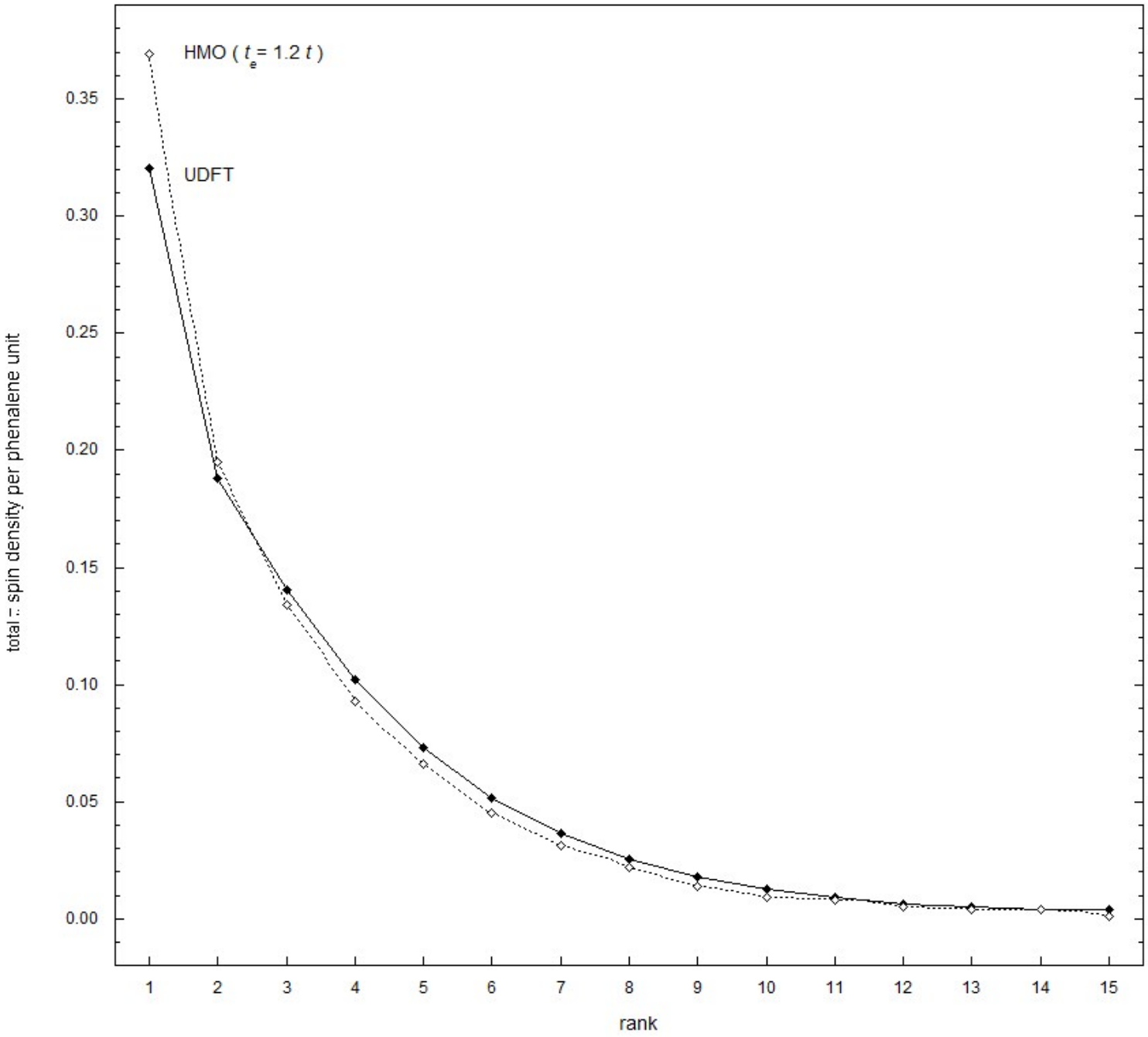

Figure S1. π spin density per phenalene unit in L15. Comparison between UDFT values (solid line) and Hückel SOMO results, using t_e=1.2 t for the shorter exterior bonds (dashed line).

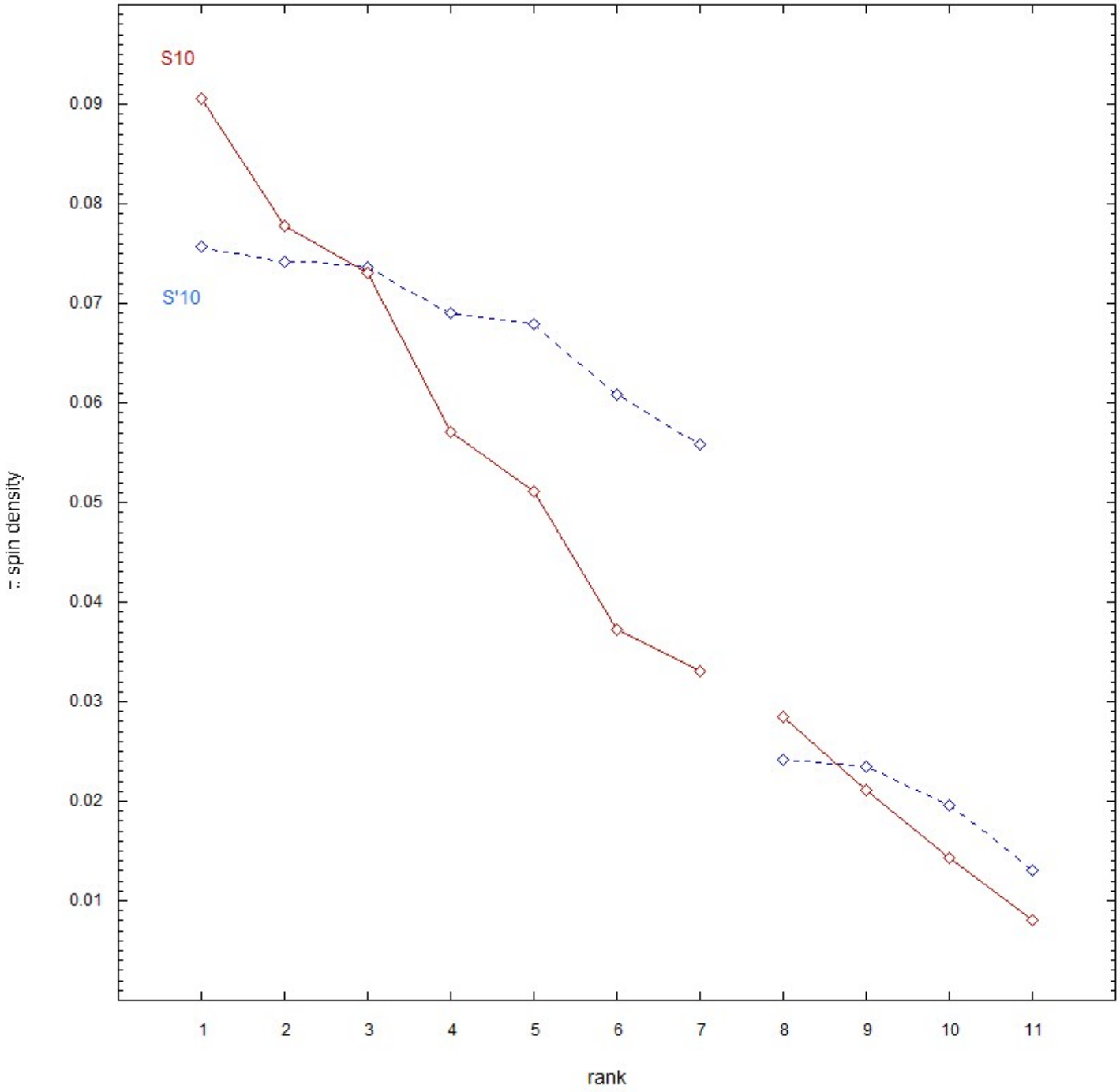

Figure S2. Ranked π spin densities on major conjugated sites in S10 and S'10 systems.

See labeling in Figure S3.

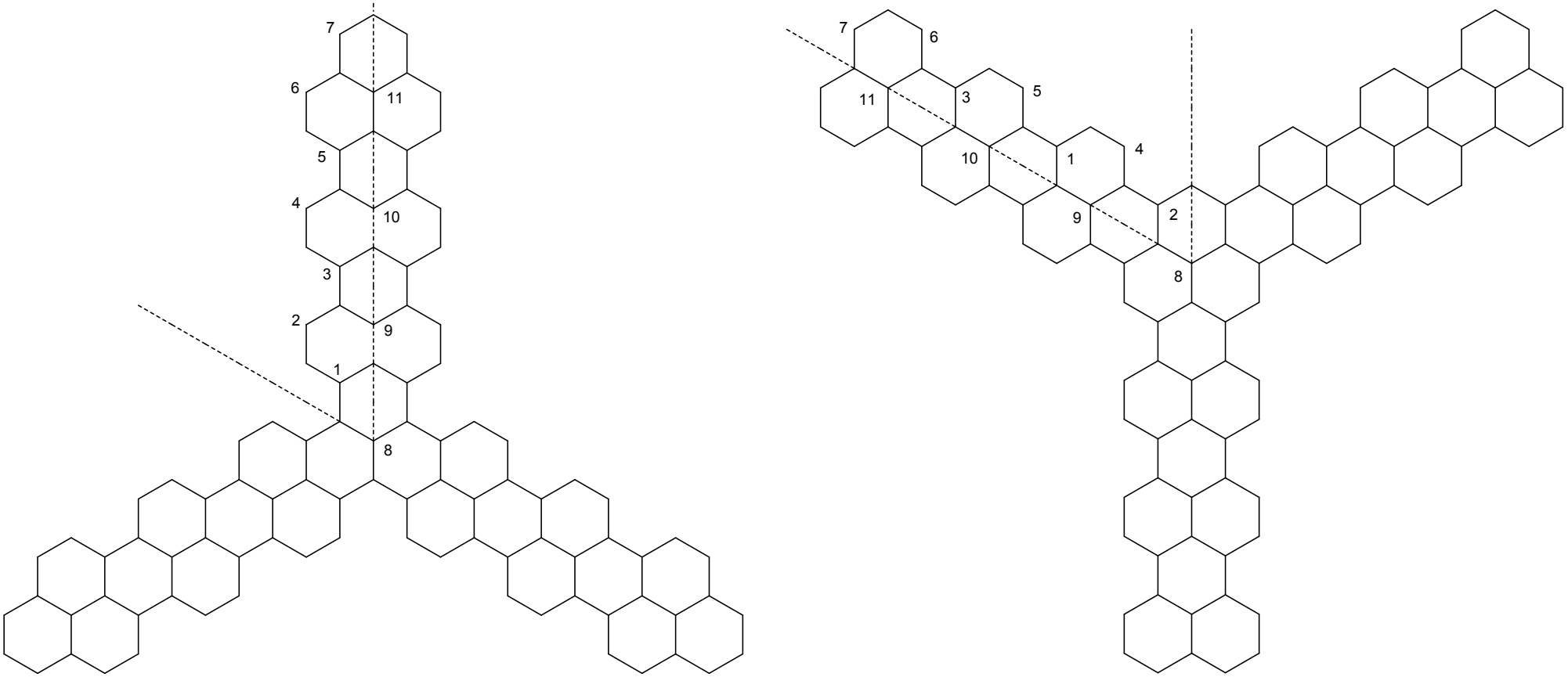

Figure S3. Labeling of major-spin sites in S10 (left) and S'10 (right). Like S4, to ensure planarity, the actual molecular system for S10 includes three additional non-conjugated -CH_2- bridges.

Table S1. Calculated CC bond lengths (Å) in P19. [a]

	radical	cation	anion
a	1.425	1.424	1.426
b	1.422	1.421	1.423
c	1.420	1.419	1.421
d	1.428	1.427	1.427
e	1.433	1.432	1.432
f	1.425	1.424	1.424
g	1.420	1.420	1.420
h	1.423	1.422	1.424
i	1.419	1.418	1.420
j	1.436	1.435	1.435
k	1.408	1.408	1.408
l	1.423	1.422	1.425
m	1.429	1.428	1.430
n	1.437	1.436	1.439
o	1.425	1.425	1.425
p	1.421	1.422	1.422
q	1.403	1.402	1.403
r	1.421	1.421	1.421
s	1.415	1.415	1.415
t	1.432	1.431	1.433
u	1.426	1.426	1.426
v	1.410	1.409	1.410
w	1.414	1.415	1.415
x	1.414	1.414	1.414
y	1.426	1.425	1.428
z	1.426	1.425	1.427
aa	1.423	1.423	1.423
bb	1.417	1.417	1.417
cc	1.365	1.365	1.365
dd	1.431	1.431	1.431
ee	1.440	1.440	1.440
ff	1.429	1.428	1.430
gg	1.421	1.421	1.421
hh	1.367	1.367	1.367
ii	1.449	1.449	1.449
jj	1.385	1.385	1.385

[a] See Figure S4 for labeling.

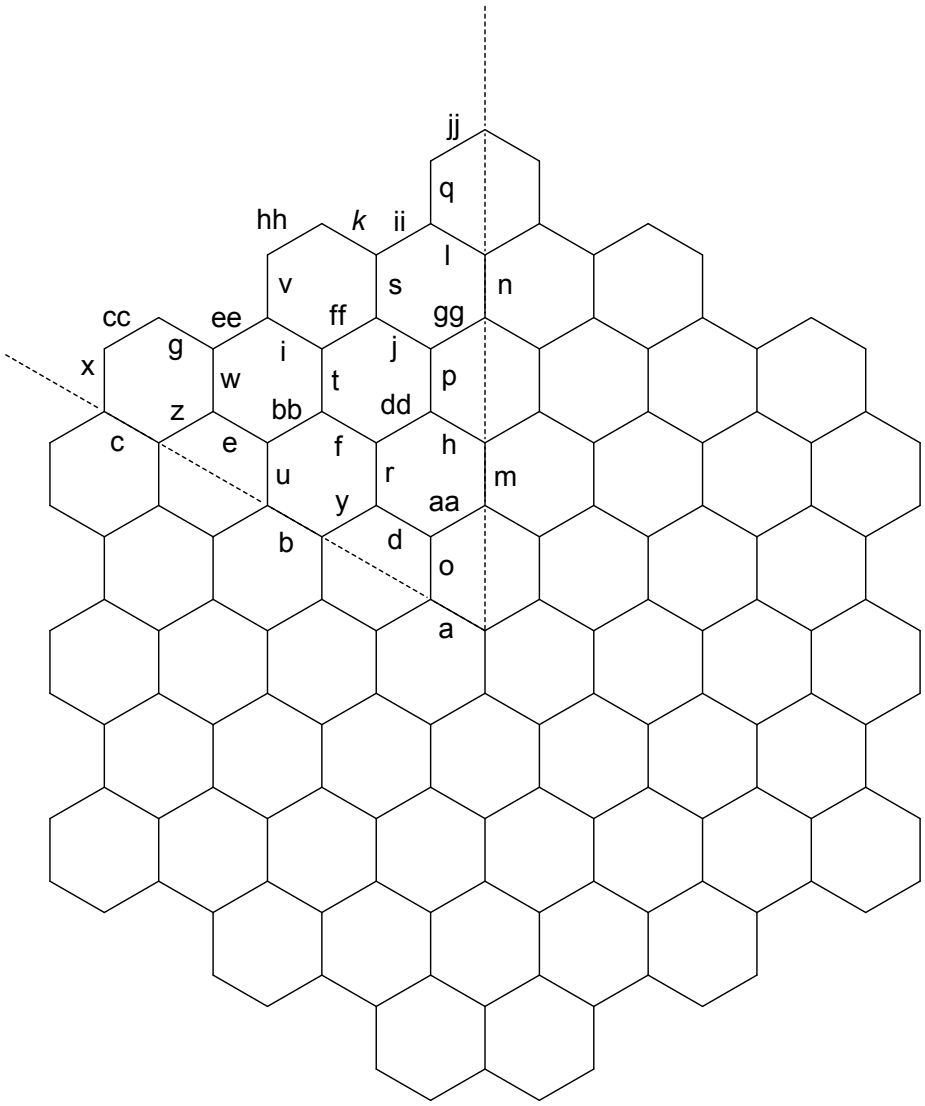

Figure S4. Bond-length labeling for Table S1.

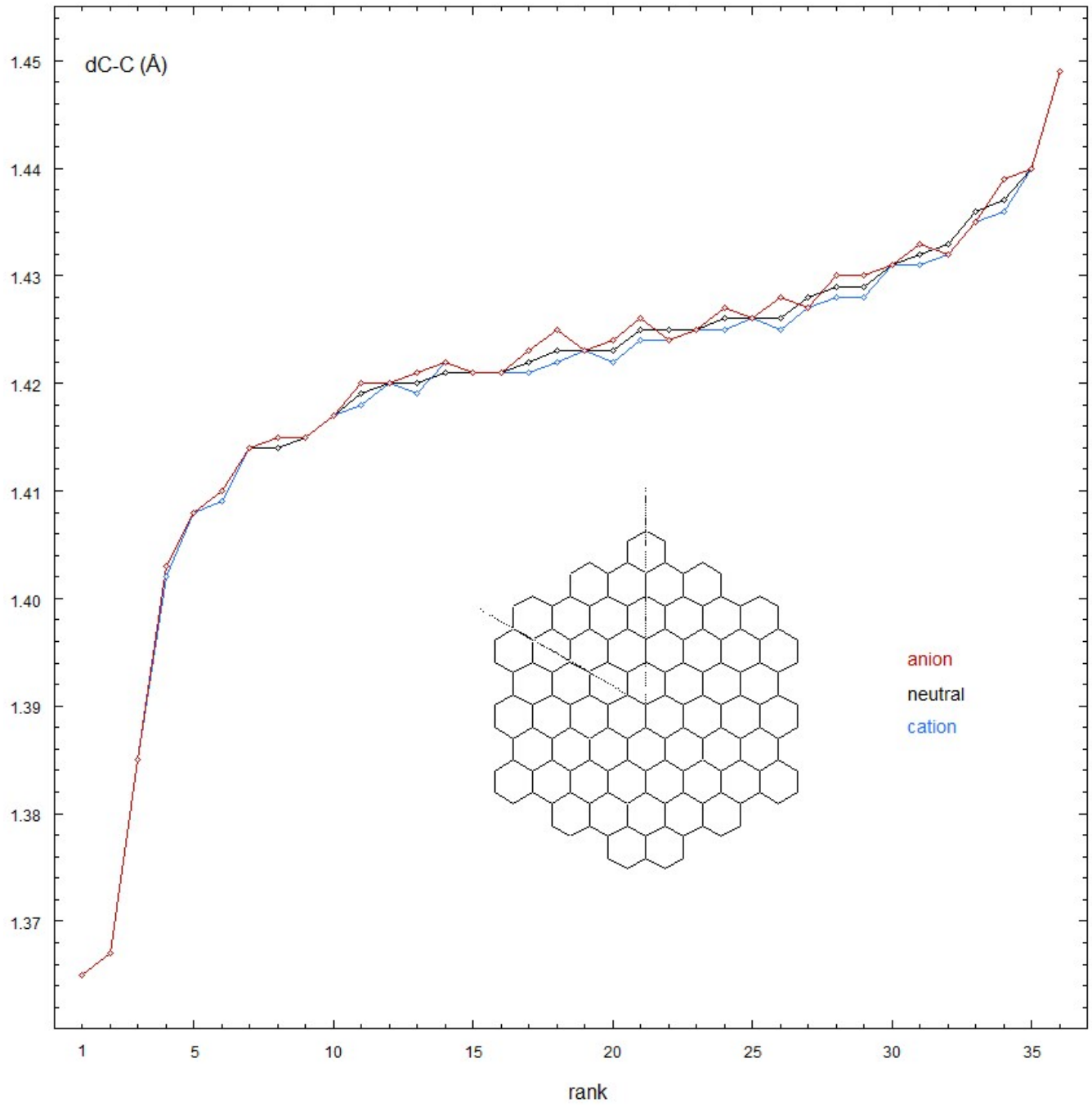

Figure S5. Ranking and comparing the CC bond lengths of Table S1.

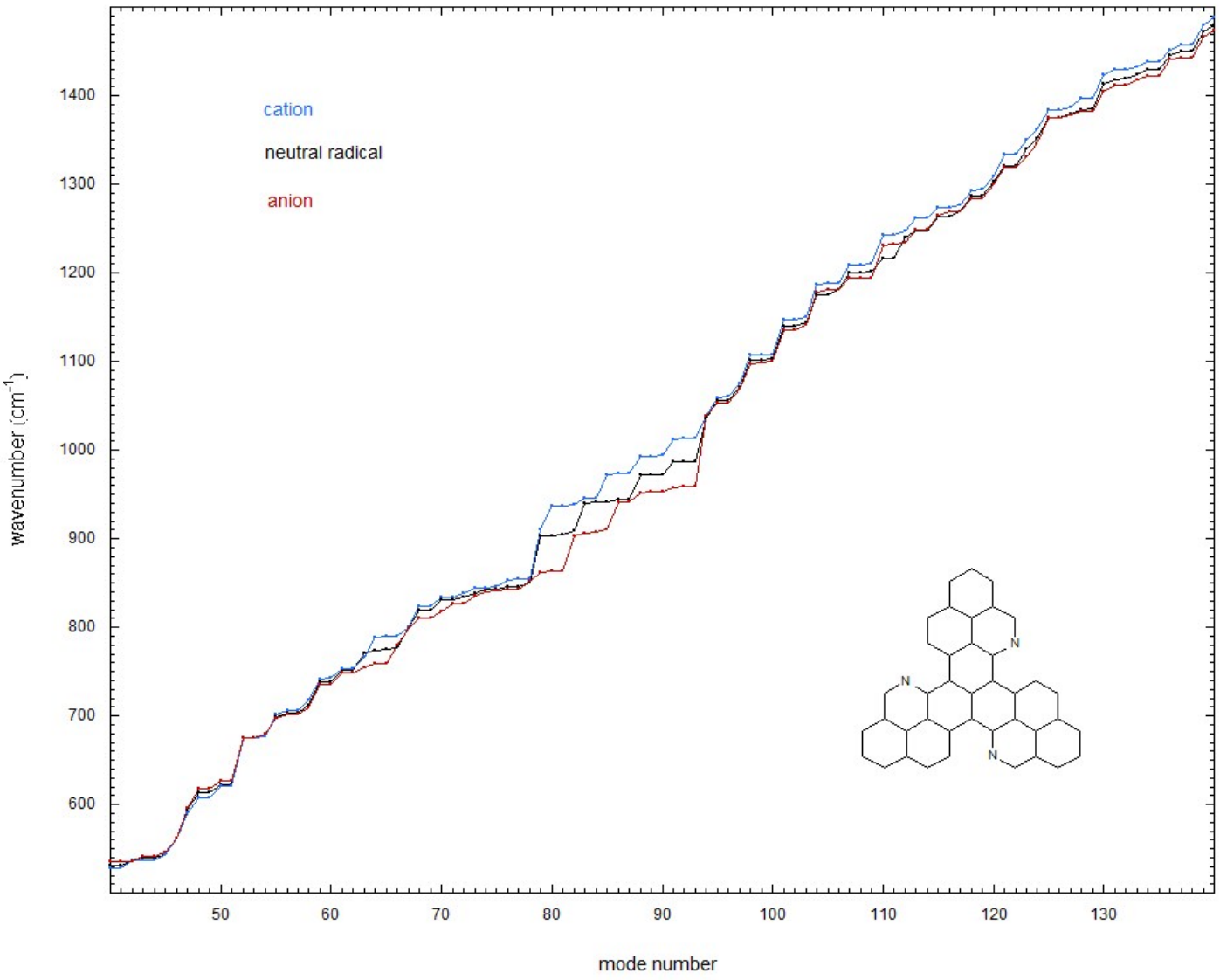

Figure S6. Some harmonic vibrational frequencies for $C_{40}H_{18}N_3$, a non-planar variant of S4 in neutral (black), cationic (blue) and anionic (red) states.