Direct enantiomeric discrimination through antisymmetric hyperfine coupling: Electronic Supporting Information

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Derivation of the chiral dipole moment contributions

The conversion of $\boldsymbol{\mu}_e^c$ from the molecule-fixed frame [MF, (α,β,γ)] to the laboratory frame [LF, (a,b,c)] is $\mu_{e,a} = l_{a\alpha}\mu_{e,\alpha}$ ($l_{a\alpha} = \cos\theta_{a\alpha}$ are the direction cosines) and the conversions of the experimentally controlled variables $\boldsymbol{B}_0$, $\hat{\boldsymbol{S}}$, and $\hat{\boldsymbol{I}}$ from LF to MF are $B_{0,\beta} = l_{\beta b}B_{0,b}$ etc. The terms of $\hat{\mathcal{H}}$ [Eq. (1)] can now be expressed as

$$\begin{aligned}
\hat{\mathcal{H}}_A &= h\hat{S}_\beta A_{\beta\gamma}\hat{I}_\gamma = h l_{\beta b} l_{\gamma c} A_{\beta\gamma}\hat{S}_b\hat{I}_c, \\
\hat{\mathcal{H}}_S &= -\mu_B B_{0,\beta} g_{S,\beta\gamma}\hat{S}_\gamma = -\mu_B l_{\beta b} l_{\gamma c} g_{S,\beta\gamma} B_{0,b}\hat{S}_c, \\
\hat{\mathcal{H}}_I &= -\mu_N B_{0,\beta} g_{I,\beta\gamma}\hat{I}_\gamma = -\mu_N l_{\beta b} l_{\gamma c} g_{I,\beta\gamma} B_{0,b}\hat{I}_c.
\end{aligned} \tag{S1}$$

The a-component $\langle\mu_{e,a}^c\rangle$ [Eq. (5)] can now be expressed in LF as

$$\langle\mu_{e,a}^c\rangle = -\frac{1}{k_B T}\langle l_{a\alpha} l_{\beta b} l_{\gamma c}\rangle_{\rm iso}\mathcal{V}_{\alpha\beta\gamma}\mathcal{W}_{bc} = -\frac{1}{6}\frac{\epsilon_{\alpha\beta\gamma}\mathcal{V}_{\alpha\beta\gamma}}{k_B T}\left(\epsilon_{abc}\mathcal{W}_{bc}\right), \tag{S2}$$

with contributions from 3- and 2-index tensors defined in MF and LF as

$$\mathcal{V}_{\alpha\beta\gamma} \equiv \mu_{e,\alpha}A_{\beta\gamma}, \qquad \mu_{e,\alpha}g_{S,\beta\gamma}, \qquad \mu_{e,\alpha}g_{I,\beta\gamma} \tag{S3}$$

$$\mathcal{W}_{bc} \equiv h\hat{S}_b\hat{I}_c, \qquad -\mu_B B_{0,b}\hat{S}_c, \qquad -\mu_N B_{0,b}\hat{I}_c, \tag{S4}$$

correspondingly, using $\langle l_{\alpha a} l_{\beta b} l_{\gamma c}\rangle_{\rm iso} = \frac{1}{6}\epsilon_{\alpha\beta\gamma}\epsilon_{abc}$, where $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita tensor.[1] From Eq. (S4) one now obtains

$$\epsilon_{abc}\mathcal{W}_{bc} = h\left(\hat{\boldsymbol{S}}\times\hat{\boldsymbol{I}}\right)_a,\ -\mu_B\left(\hat{\boldsymbol{S}}\times\boldsymbol{B}_0\right)_a,\ -\mu_N\left(\hat{\boldsymbol{I}}\times\boldsymbol{B}_0\right)_a. \tag{S5}$$

leading, hence, to Eqs. (6)-(8) in the main paper.

Details of hyperfine coupling calculations

To compute the antisymmetric part of the hyperfine coupling tensor, $\boldsymbol{A}_{\mathrm{anti}}$ (Ref. 2) one has to employ a relativistic quantum-chemical method that includes spin-orbit coupling (SOC).[3–6] Adopting the viewpoint of nonrelativistic theory supplemented by perturbationally applied SOC, $\boldsymbol{A}_{\mathrm{anti}}$ appears from the second-order cross-term of SOC with the orbital hyperfine operator, an energy contribution that includes the necessary bilinear dependence on both $\hat{\boldsymbol{S}}$ and $\hat{\boldsymbol{I}}$.[6] The perturbational method involves a distinct response calculation for each Cartesian component of the nuclear spin, which becomes computationally heavy for large molecular systems. The well-known Fermi contact and spin-dipole hyperfine operators contribute to A_{iso} and $\boldsymbol{A}_{\mathrm{sym}}$, respectively, already to first order, but do not give rise to $\boldsymbol{A}_{\mathrm{anti}}$.[7] Instead, in the four-component, fully relativistic framework, the hyperfine tensor is obtained as an expectation value of a single relativistic hyperfine operator,[8] and $\boldsymbol{A}_{\mathrm{anti}}$ can be simply extracted from the resulting, full hyperfine tensor.

Calculations of $\boldsymbol{A}$ are generally demanding.[9] One aspect of that arises due to the properties of the hyperfine operators: one has to include high-exponent functions in the basis set to be able to have sufficient flexibility in the region of the electron cloud close to the atomic core. To be used in the context of density-functional theory (DFT) calculations, Jakobsen and Jensen[10] have very recently published a series of basis sets for hyperfine coupling constants for elements from H to Ar, and which shows a rapid and systematic basis-set convergence. Another issue in hyperfine coupling calculations is the electron correlation treatment, which needs to be combined with means of allowing for the spin polarization down to the atomic core, for reliable results. In the DFT framework this means using the unrestricted Kohn-Sham formalism and within *ab initio* wave function methods correlating all electrons. While advances in large-scale correlated wave function theory calculation of hyperfine couplings have been made,[11,12] fully relativistic, unrestricted DFT calculation remains currently the method of choice of extracting $\boldsymbol{A}_{\mathrm{anti}}$ computationally for large molecular systems. It is well-known that prediction of properties involving hyperfine operators is sensitive to the used

exchange-correlation functional, particularly the choice of the exact-exchange admixture in hybrid DFT calculations.[13,14] Investigating results with multiple choices of this parameter produces a range of results that can be used for a rough estimate of the methodological error.

Structure optimizations on the Turbomole software[15] using unrestricted DFT with the PBE0 hybrid functional,[16] the empirical D3 BJ dispersion correction,[17,18] and the def2-QZVPP basis set[19] (Tab. S1-S8).

In calculating the $\boldsymbol{A}$ tensors, three related exchange-correlation functionals with varying exact-exchange parameter were employed: the generalized gradient functional PBE (0% exact exchange),[20] the one-parameter hybrid functional PBE0 (25%),[16] and PBE0-40, in which the exact-exchange admixture was raised to 40% as recommended in Ref. 21 in the context of transition-metal complexes. Very tight numerical convergence thresholds were applied in the three self-consistent field optimizations for the spin quantization axis in x, y, and z directions, in turn, as appropriate for the method.[22] Gaussian nuclear charge distribution model and "large" atomic integration grids were applied.

Entirely uncontracted large-component basis sets were employed and the small-component basis was always created by restricted kinetic balance.[23,24] The large-component basis was selected by performing calculations on the verdazyl radical **3a** using the series of polarization-consistent pcH-n ($n = 1, 2, 3$) basis sets optimized for hyperfine coupling calculations.[10] Apart from a couple of very small ^{1}H and ^{13}C HFC constants, the results at the pcH-2 level were converged for this quantity to within 1 % of the results obtained with the larger pcH-3 basis. Hence, pcH-2 was adopted in all the production calculations for all the systems.

Table S1: Optimized atomic coordinates of the radical **1a**.

	atom	X / Å	Y / Å	Z / Å
1	O	0.0012059	2.1219133	0.4703791
2	N	0.0004044	0.9479470	−0.0007456
3	C	−1.7083106	−0.0803340	1.4315369
4	C	−1.3074474	0.2484589	−0.0052515
5	C	−2.3302706	1.1995165	−0.6050515
6	C	−1.2312772	−1.0118421	−0.8602101
7	C	−0.0012504	−1.8493326	−0.5844174
8	C	1.2300867	−1.0136579	−0.8598909
9	C	1.3076326	0.2472203	−0.0058620
10	C	2.3307683	1.1970562	−0.6071062
11	C	1.7092176	−0.0808649	1.4308864
12	H	−2.7456695	−0.4157453	1.4537954
13	H	−1.6170051	0.8119269	2.0492562
14	H	−1.0911401	−0.8667180	1.8634046
15	H	−2.4469019	2.0865372	0.0121266
16	H	−2.0214461	1.5117360	−1.6026548
17	H	−3.2906337	0.6887993	−0.6821306
18	H	−2.1473799	−1.5815720	−0.6933337
19	H	−1.2285808	−0.7174932	−1.9138870
20	H	−0.0017808	−2.7384693	−1.2176353
21	H	−0.0017197	−2.2075281	0.4481214
22	H	1.2285363	−0.7201703	−1.9138085
23	H	2.1452823	−1.5845327	−0.6919577
24	H	2.4483581	2.0845757	0.0091771
25	H	3.2907534	0.6856508	−0.6843301
26	H	2.0215114	1.5085386	−1.6048106
27	H	1.0919416	−0.8667209	1.8635856
28	H	2.7464382	−0.4167266	1.4527272
29	H	1.6186768	0.8118315	2.048087

Table S2: Optimized atomic coordinates of the radical *syn*-(*R*)-**1b**.

	atom	X / Å	Y / Å	Z / Å
1	H	−6.0408974	−0.1699459	0.3052629
2	H	−4.718144	0.1538162	−2.1315081
3	H	−4.8445565	−1.3842465	0.7803631
4	H	−4.0976577	−1.2224284	−1.5381359
5	H	−4.9707884	0.117628	1.6948486
6	H	−4.2478672	1.3971521	−0.3176354
7	H	−1.8752949	−0.0961012	−1.3652404
8	H	−0.2710148	0.1297392	1.1031851
9	H	−0.0576983	−2.0951527	−0.0029189
10	H	0.013792	2.1618355	−0.2948371
11	H	0.4394575	−1.2992566	−1.4912205
12	H	0.4879518	1.1556732	−1.6585362
13	H	1.3859554	−0.9263879	2.1554707
14	H	1.6703319	−2.6154982	1.7435337
15	H	1.4079566	1.2352614	2.0130663
16	H	2.1060316	−3.3299526	−0.5614443
17	H	1.7469127	2.8444515	1.3864167
18	H	2.6444645	−2.1577826	−1.7751637
19	H	2.2181217	3.2421784	−0.9850995
20	H	3.0385616	−1.500836	1.8881052
21	H	2.7285233	1.9079553	−2.0320022
22	H	3.0798233	1.7182644	1.6879752
23	H	3.6595655	−2.4946587	−0.3688561
24	H	3.7428849	2.3920688	−0.6692338
25	C	−5.0333662	−0.3111639	0.6971914
26	C	−4.0104714	0.3335132	−0.2212439
27	C	−2.629607	0.3175135	0.4386379
28	C	−0.238075	0.048555	0.0167871
29	C	0.4674787	−1.2244779	−0.399006
30	C	0.5115461	1.229694	−0.5666031
31	C	1.916609	−1.2926457	0.0640598
32	C	2.0045277	−1.5946173	1.5588393
33	C	1.9594525	1.3102786	−0.104542
34	C	2.0507141	1.7989977	1.3396304
35	C	2.6329909	−2.387262	−0.709714
36	C	2.7160344	2.2727706	−1.0050223
37	N	−1.6154107	0.0370213	−0.3987738
38	N	2.6110921	−0.0155462	−0.2224894
39	N	−3.9670406	−0.2192108	−1.5697336
40	O	−2.4930948	0.5628009	1.6235194
41	O	3.8702052	−0.0299975	−0.1079324

Table S3: Optimized atomic coordinates of the radical *anti*-(*R*)-**1b**.

	atom	X / Å	Y / Å	Z / Å
1	H	−5.6729826	−1.0297889	−0.4774862
2	H	−4.0199815	2.0553925	−0.9058703
3	H	−4.0627896	−1.5901536	−0.9617144
4	H	−4.8719331	0.8881746	−1.6551804
5	H	−4.5758215	−1.7162099	0.7297987
6	H	−4.6402726	0.7374155	0.9175591
7	H	−1.9485810	0.6689468	−1.0912909
8	H	−0.2435020	0.0963661	1.2463088
9	H	−0.2965109	−1.9193800	−0.1932134
10	H	0.1674672	2.3072184	0.1626481
11	H	0.2036783	−0.9476660	−1.5734660
12	H	0.4670391	1.4918181	−1.3678104
13	H	1.3442753	−1.2234922	2.0208076
14	H	1.4620193	−2.8460587	1.3506137
15	H	1.5927744	0.9066232	2.1985669
16	H	1.7168016	−3.2453893	−1.0444228
17	H	2.0521125	2.5626148	1.8069948
18	H	2.3006580	−1.9599144	−2.1136933
19	H	2.4172295	3.2879973	−0.5105991
20	H	2.9284183	−1.8802609	1.5801924
21	H	2.7436025	2.0947627	−1.7782220
22	H	3.2810850	1.2876350	1.8332535
23	H	3.3483321	−2.5810741	−0.8342416
24	H	3.8715609	2.2750663	−0.4308465
25	C	−4.6290690	−1.1021622	−0.1664604
26	C	−4.0562791	0.2756977	0.1140574
27	C	−2.6539195	0.0994963	0.6904762
28	C	−0.2700047	0.2005600	0.1614846
29	C	0.2924785	−1.0493596	−0.4871853
30	C	0.5552969	1.3888800	−0.2808331
31	C	1.7478048	−1.3130077	−0.1285413
32	C	1.8767758	−1.8396997	1.2992337
33	C	2.0287655	1.2705768	0.0854971
34	C	2.2472766	1.5149746	1.5773239
35	C	2.3206048	−2.3392146	−1.0920636
36	C	2.8212096	2.2953168	−0.7095757
37	N	−1.6585165	0.3885881	−0.1657916
38	N	2.5437074	−0.0711239	−0.2745549
39	N	−4.0412373	1.0658325	−1.1091268
40	O	−2.4902909	−0.2983521	1.8293483
41	O	3.8007178	−0.2076462	−0.2519749

Table S4: Optimized atomic coordinates of the radical **2a**.

	atom	X / Å	Y / Å	Z / Å
1	O	−2.5155594	−0.5736613	0.0509528
2	N	0.9756999	−1.1369791	0.0238937
3	N	−0.3721080	−1.3038254	0.0517832
4	N	−0.8280551	0.9652495	−0.0107053
5	C	2.8783823	0.3213768	−0.0435411
6	C	1.3986319	0.1232922	−0.0208532
7	C	−0.8381690	−2.6704841	0.0924284
8	C	−1.3280226	−0.3182646	0.0316778
9	C	−1.7456532	2.0760503	−0.0480189
10	C	0.5321625	1.1849935	−0.0413391
11	H	3.3310218	−0.0656647	0.8702022
12	H	3.3224442	−0.2162128	−0.8816137
13	H	3.1297554	1.3767055	−0.1343508
14	H	0.0435563	−3.3016068	0.1221093
15	H	−1.4558164	−2.8384156	0.9740183
16	H	−1.4376205	−2.8990232	−0.7887284
17	H	−1.6535992	2.6175695	−0.9918411
18	H	−1.5393863	2.7611888	0.7757944
19	H	−2.7533796	1.6841608	0.0462386
20	H	0.8557148	2.2135507	−0.0781071

Table S5: Optimized atomic coordinates of the radical (*R*)-**2b**.

	atom	X / Å	Y / Å	Z / Å
1	O	6.2568533	10.5608126	0.2521889
2	O	3.9525655	5.3819053	1.0866562
3	O	2.5442070	4.6826080	−0.5578684
4	N	7.0793583	7.1420316	0.3457689
5	N	7.1242313	8.4726928	0.5456443
6	N	5.3263275	8.7912606	−0.8476019
7	N	5.2514775	7.4733083	−1.1004363
8	N	4.7011205	4.8158516	−0.9746295
9	C	6.2351127	9.3738591	0.0077803
10	C	6.1339897	6.7223314	−0.4668347
11	C	4.3178280	9.6049299	−1.4780240
12	C	8.1201183	8.9313195	1.4812221
13	C	6.0591195	5.2322485	−0.7429544
14	C	6.7616314	4.4105718	0.3423875
15	C	3.7395923	5.0003245	−0.0427256
16	C	1.3531075	4.7605048	0.2650922
17	C	1.4653704	3.8104870	1.4429488
18	C	1.1139005	6.1920263	0.7062125
19	C	0.2551728	4.3055888	−0.6742768
20	C	6.6957012	2.9427399	0.0623934
21	C	5.6818093	2.1651674	0.6094101
22	C	5.5939543	0.8123036	0.3263301
23	C	6.5223682	0.2150834	−0.5116140
24	C	7.5400167	0.9791619	−1.0604537
25	C	7.6233053	2.3319957	−0.7736137
26	H	4.3944641	4.6277270	−1.9095290
27	H	3.3420073	9.3967923	−1.0368212
28	H	4.2857813	9.3644966	−2.5387341
29	H	4.5762728	10.6470176	−1.3253568
30	H	7.8230982	8.6834874	2.5014700
31	H	8.2143464	10.0071796	1.3808425
32	H	9.0614085	8.4359316	1.2547137
33	H	6.5815451	5.0536242	−1.6857562
34	H	6.2907952	4.6407099	1.2959163
35	H	7.7952697	4.7479296	0.3971658
36	H	1.6929930	2.8038058	1.0907687
37	H	0.5134537	3.7798837	1.9725017
38	H	2.2424787	4.1315807	2.1307464
39	H	0.1540598	6.2554094	1.2184848
40	H	1.0815669	6.8517623	−0.1617382
41	H	1.8973472	6.5280914	1.3794463
42	H	0.1920435	4.9650757	−1.5394032
43	H	−0.7024004	4.3215383	−0.1550404
44	H	0.4448054	3.2904136	−1.0220992
45	H	4.9577853	2.6337037	1.2654553
46	H	4.7981995	0.2219259	0.7630102

47	H	6.4563418	−0.8425488	−0.7326379
48	H	8.2745219	0.5192538	−1.7094315
49	H	8.4257264	2.9256744	−1.1978064

Table S6: Optimized atomic coordinates of the radical **3a**.

	atom	X / Å	Y / Å	Z / Å
1	C	0.0000000	0.0000000	0.0004479
2	C	−0.1601750	4.2586443	−0.0003615
3	C	−1.1339608	3.5203584	−0.6592321
4	C	−1.0860808	2.1398106	−0.6563051
5	C	−0.0545490	1.4499189	0.0002846
6	C	−1.2283921	−0.7722003	0.0002846
7	C	−1.3100899	−2.0104789	−0.6563051
8	C	−2.4817394	−2.7422181	−0.6592321
9	C	−3.6080067	−2.2680378	−0.0003615
10	C	−3.5471705	−1.0478448	0.6591278
11	C	−2.3799320	−0.3091339	0.6567329
12	C	1.2829411	−0.6777186	0.0002846
13	C	2.3961708	−0.1293317	−0.6563051
14	C	3.6157002	−0.7781403	−0.6592321
15	C	3.7681817	−1.9906065	−0.0003615
16	C	2.6810455	−2.5480173	0.6591278
17	C	1.4576838	−1.9065146	0.6567329
18	C	0.9222482	2.2156485	0.6567329
19	C	0.8661250	3.5958621	0.6591278
20	H	−0.2006627	5.3402816	−0.0009563
21	H	−1.9333876	4.0259672	−1.1863551
22	H	−1.8417164	1.5741633	−1.1852852
23	H	−0.4424072	−2.3820549	−1.1852852
24	H	−2.5198961	−3.6873464	−1.1863551
25	H	−4.5244881	−2.8439198	−0.0009563
26	H	−4.4156516	−0.6727552	1.1860567
27	H	−2.3386706	0.6335808	1.1861441
28	H	2.2841236	0.8078916	−1.1852852
29	H	4.4532837	−0.3386208	−1.1863551
30	H	4.7251509	−2.4963618	−0.0009563
31	H	2.7904489	−3.4876889	1.1860567
32	H	0.6206382	−2.3421385	1.1861441
33	H	1.7180324	1.7085577	1.1861441
34	H	1.6252027	4.1604441	1.1860567

Table S7: Optimized atomic coordinates of the radical **3b**.

	atom	X / Å	Y / Å	Z / Å
1	C	0.0045860	−1.1440730	0.0042273
2	C	−1.2549878	−0.5612634	0.1563179
3	C	−1.6289025	0.7307020	0.7052361
4	C	−0.8657641	1.7446134	1.2687239
5	C	−1.5061783	2.8526276	1.7960413
6	C	−2.8885327	2.9518848	1.7826444
7	C	−3.6652514	1.9217801	1.2562285
8	C	−3.0359415	0.8243790	0.7326459
9	C	−3.5831205	−0.4445633	0.1989358
10	C	−4.8702281	−0.8699254	0.0054836
11	C	−5.0823220	−2.1504284	−0.5037948
12	C	−4.0113131	−2.9741571	−0.8138702
13	C	−2.7077351	−2.5433127	−0.6278524
14	C	−2.4907744	−1.2741377	−0.1108734
15	C	1.2593595	−0.5523216	−0.1522173
16	C	1.6230435	0.7387992	−0.7099830
17	C	0.8519107	1.7428532	−1.2801631
18	C	1.4835223	2.8522904	−1.8150394
19	C	2.8650527	2.9624859	−1.8025801
20	C	3.6498895	1.9420731	−1.2694142
21	C	3.0292861	0.8433169	−0.7383113
22	C	3.5865103	−0.4175966	−0.1960719
23	C	4.8769633	−0.8314952	−0.0000240
24	C	5.0992256	−2.1068731	0.5176561
25	C	4.0347876	−2.9369341	0.8334172
26	C	2.7278369	−2.5175993	0.6448614
27	C	2.5008105	−1.2536433	0.1195148
28	H	0.0089543	−2.2160671	0.0083831
29	H	0.1974840	1.6784495	1.2915858
30	H	−0.9242300	3.6418836	2.2227044
31	H	−3.3628540	3.8170179	2.1948647
32	H	−4.7331339	1.9840717	1.2780391
33	H	−5.7010783	−0.2380282	0.2399166
34	H	−6.0809526	−2.4996301	−0.6594576
35	H	−4.1946315	−3.9517077	−1.2070224
36	H	−1.8875844	−3.1788316	−0.8860399
37	H	−0.2107830	1.6681984	−1.3024384
38	H	0.8953198	3.6340408	−2.2469064
39	H	3.3325093	3.8285587	−2.2206241
40	H	4.7172479	2.0125807	−1.2918117
41	H	5.7027461	−0.1945790	−0.2387247
42	H	6.1005987	−2.4471727	0.6753932
43	H	4.2258601	−3.9104139	1.2329230
44	H	1.9127945	−3.1578525	0.9074759

Table S8: Optimized atomic coordinates of the radical (*R*)-**3c**.

	atom	*X* / Å	*Y* / Å	*Z* / Å
1	N	9.2717496	2.8112891	−5.5981533
2	O	9.9534495	0.4894779	−4.5117908
3	O	11.7888911	0.3183532	−5.7777427
4	O	9.5866296	4.7453136	−6.7143763
5	C	6.3202901	5.1429265	−0.3375579
6	C	6.3346205	4.3761866	0.8326674
7	C	7.1614716	4.8777049	−1.4269804
8	C	3.6208784	8.4284447	−0.6153619
9	C	3.1667004	7.1916058	−0.1817993
10	C	4.0442094	6.1264900	−0.0857012
11	C	5.3845435	6.2815467	−0.4298549
12	C	5.8300853	7.5256834	−0.8678775
13	C	4.9553397	8.5936566	−0.9556999
14	C	6.0150235	4.8194721	2.1839900
15	C	6.1513748	3.7219374	3.0620577
16	C	6.4968975	2.5477204	2.2702439
17	C	6.6191782	2.9495817	0.9249392
18	C	6.8886659	5.1202483	−2.8354927
19	C	7.9984109	4.6757509	−3.5935497
20	C	9.0471995	4.2689440	−2.6578287
21	C	8.5198140	4.3652031	−1.3530399
22	C	5.7331888	6.0811366	2.6981440
23	C	5.5645739	6.2286521	4.0661902
24	C	5.6823973	5.1396494	4.9229781
25	C	5.9841404	3.8785671	4.4242179
26	C	6.6548906	1.2229154	2.6292113
27	C	6.9218629	0.2874715	1.6371288
28	C	7.0089045	0.6735821	0.3037065
29	C	6.8536820	2.0014357	−0.0624034
30	C	5.7477163	5.5867247	−3.4776006
31	C	5.7294739	5.6474661	−4.8623658
32	C	6.8152068	5.2102021	−5.6052875
33	C	7.9440947	4.6883596	−4.9795842
34	C	10.3796433	3.9441624	−2.8466555
35	C	11.1760568	3.6931511	−1.7379260
36	C	10.6561100	3.7854042	−0.4518337
37	C	9.3313458	4.1324375	−0.2499353
38	C	10.3472081	2.1127778	−6.2406831
39	C	10.0166493	1.7149918	−7.6746914
40	C	10.6545867	0.8975175	−5.3996645
41	C	9.0194008	4.1134448	−5.8425127
42	C	12.1347945	−0.8608033	−5.0568620
43	H	2.9351120	9.2625589	−0.6885992
44	H	2.1257486	7.0565753	0.0822888
45	H	3.7017529	5.1575189	0.2537607
46	H	6.8720381	7.6419110	−1.1379044

47	H	5.3152761	9.5567290	−1.2932430
48	H	5.6409982	6.9375145	2.0453960
49	H	5.3389015	7.2064272	4.4721800
50	H	5.5462057	5.2777301	5.9879463
51	H	6.0959763	3.0351036	5.0941423
52	H	6.5578266	0.9151495	3.6629384
53	H	7.0484930	−0.7541818	1.9020102
54	H	7.1987553	−0.0704106	−0.4592525
55	H	6.9115052	2.2917526	−1.1024888
56	H	4.8890652	5.9130336	−2.9084798
57	H	4.8535617	6.0291913	−5.3702116
58	H	6.7965197	5.2465642	−6.6867468
59	H	10.8084101	3.9142337	−3.8380145
60	H	12.2193202	3.4409017	−1.8770765
61	H	11.2966093	3.5999725	0.4005582
62	H	8.9362738	4.2345723	0.7512427
63	H	11.2329457	2.7538615	−6.2588481
64	H	8.8300312	2.3423654	−4.8243000
65	H	9.1429839	1.0635491	−7.6946029
66	H	10.8596351	1.2002884	−8.1310325
67	H	9.7990796	2.6175952	−8.2410407
68	H	12.2581403	−0.6356812	−3.9986676
69	H	13.0691291	−1.2067936	−5.4863526
70	H	11.3579753	−1.6153654	−5.1680746

Table S9: Comparison between the isotropic part of the hyperfine coupling tensor $\boldsymbol{A}$ (in MHz) derived from mDKS computations with literature experimental data

Radical	Nucleus	Position	Computations			Experiment[a]	Ref.
			PBE	PBE0	PBE0-40		
1a	^{14}N		−40.36	−51.29	−57.76	−48.3	[25]
	^{13}C	CH_3	13.29	12.42	11.98	15.1	[26]
2a	^{14}N	N_α	−10.61	−15.94	−18.63	−14.4	[27]
	^{14}N	N_β	−13.24	−22.63	−29.35	−18.0	[27]
	^{1}H	o	18.71	16.98	15.82	15.1	[27]
	^{1}H	p	−5.61	−10.75	−14.17	−6.6	[27]
3a	^{13}C		40.58	61.23	75.19	66.9	[28]
	^{1}H	o	−6.37	−9.73	−12.11	−7.3	[29]
	^{1}H	m	2.79	5.27	7.79	3.2	[29]
	^{1}H	p	−7.54	−10.27	−12.91	−8.0	[29]
3b	^{13}C	C_α	23.43	40.42	53.38	38.8	[30]
	^{13}C	C_β	−21.29	−38.32	−51.69	−33.7	[30]
	^{1}H	H_α	32.56	42.77	50.17	35.5	[30]
	^{1}H	H_1	−4.85	−7.25	−9.50	−5.7	[30]
	^{1}H	H_2	1.00	2.80	4.65	1.4	[30]
	^{1}H	H_3	−5.25	−7.49	−9.48	−5.5	[30]
	^{1}H	H_4	0.64	2.39	4.18	1.0	[30]

[a] The sign of the hyperfine coupling tensor component is inferred based on the result of the computations.

Table S10: The electron spin g-tensor of studied radicals

radical	g-tensor		
	−2.007206	−0.000003	0.000007
1a	−0.000001	−2.010603	−0.002933
	−0.000007	−0.002433	−2.002918
	−2.011503	0.000044	−0.000411
syn-(R)-**1b**	0.000085	−2.007234	0.000340
	0.000185	0.000315	−2.002081
	−2.011451	0.000449	0.000345
anti-(R)-**1b**	0.000396	−2.007258	−0.000500
	0.000672	−0.000390	−2.002125
	−2.004410	−0.000339	0.000030
2a	−0.000149	−2.004735	0.000041
	−0.000005	0.000117	−2.002116
	−2.004080	0.000487	−0.001508
(R)-**2b**	0.000238	−2.005126	−0.000202
	−0.001638	−0.000489	−2.003554
	−2.002870	−0.000014	0.000011
3a	0.000015	−2.002870	−0.000020
	−0.000014	0.000016	−2.002330
	−2.002819	0.000002	0.000027
3b	−0.000002	−2.002896	0.000010
	−0.000132	−0.000009	−2.002338
	−2.002631	0.000078	−0.000057
(R)-**3c**	0.000146	−2.002687	−0.000042
	−0.000130	−0.000085	−2.002865

Table S11: Antisymmetric parts of the electron spin g-tensors of studied radicals

radical	$g^\star$ / ppm		
	PBE	PBE0	PBE0-40
1a	425.5	250.1	279.0
syn-(*R*)-**1b**	331.0	299.0	226.4
anti-(*R*)-**1b**	493.0	174.5	310.6
2a	150.0	195.3	149.6
(*R*)-**2b**	301.0	200.8	170.3
3a	28.4	26.3	5.8
3b	104.0	80.1	24.2
(*R*)-**3c**	85.1	77.6	54.3

Table S12: Hyperfine coupling tensors of the radical **1a** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	56.74	0.01	−0.02	41.92	0.01	−0.02	31.91	0.01	−0.02
	0.01	32.25	74.49	0.01	17.35	79.94	0.01	7.41	83.01
	−0.02	74.61	−158.04	−0.02	80.08	−186.28	−0.02	83.16	−203.81
2	−4.00	0.00	−0.01	−14.44	0.00	−0.01	−20.90	0.00	−0.01
	0.00	−12.67	27.97	0.00	−22.81	28.23	0.00	−28.99	28.17
	−0.01	28.30	−104.42	−0.01	28.55	−116.63	−0.01	28.50	−123.40
3	16.14	0.85	−3.87	15.66	0.83	−3.61	15.37	0.82	−3.47
	0.82	14.71	−0.79	0.80	14.32	−0.80	0.79	14.08	−0.80
	−3.86	−0.79	22.71	−3.59	−0.80	21.61	−3.46	−0.80	21.03
4	−3.48	2.39	−0.53	−7.72	2.16	−0.49	−10.19	1.99	−0.47
	2.52	−5.74	−0.39	2.28	−9.50	−0.10	2.12	−11.68	0.03
	−0.89	−0.52	−6.70	−0.86	−0.24	−11.07	−0.83	−0.11	−13.48
5	11.09	−1.14	1.05	9.93	−1.00	1.06	9.40	−0.93	1.07
	−1.12	7.98	−0.13	−0.99	6.89	−0.12	−0.92	6.39	−0.11
	0.97	−0.16	7.11	0.99	−0.15	6.07	1.00	−0.15	5.60
6	1.74	0.75	0.29	1.53	0.80	0.29	1.46	0.83	0.29
	0.72	3.20	0.42	0.78	2.93	0.43	0.81	2.83	0.43
	0.36	0.48	1.42	0.35	0.48	1.20	0.35	0.48	1.12
7	−1.09	0.00	0.00	−1.34	0.00	0.00	−1.47	0.00	0.00
	0.00	0.45	0.32	0.00	0.22	0.32	0.00	0.10	0.32
	0.00	0.32	−0.94	0.00	0.32	−1.17	0.00	0.32	−1.29
8	1.71	−0.74	−0.29	1.51	−0.80	−0.29	1.44	−0.83	−0.29
	−0.72	3.18	0.42	−0.78	2.92	0.43	−0.81	2.81	0.43
	−0.35	0.48	1.40	−0.35	0.48	1.18	−0.35	0.48	1.11
9	−3.48	−2.39	0.52	−7.73	−2.16	0.49	−10.20	−2.00	0.46
	−2.52	−5.74	−0.39	−2.29	−9.50	−0.10	−2.12	−11.67	0.03
	0.89	−0.52	−6.71	0.85	−0.24	−11.07	0.83	−0.11	−13.48

10	11.10	1.14	−1.05	9.95	1.00	−1.06	9.41	0.93	−1.07
	1.12	7.99	−0.13	0.99	6.90	−0.12	0.92	6.40	−0.11
	−0.98	−0.16	7.12	−0.99	−0.15	6.08	−1.00	−0.15	5.61
11	16.11	−0.85	3.87	15.63	−0.83	3.61	15.35	−0.82	3.48
	−0.82	14.68	−0.78	−0.80	14.29	−0.79	−0.79	14.05	−0.80
	3.86	−0.79	22.68	3.60	−0.79	21.58	3.46	−0.79	21.00
12	9.06	2.45	−0.92	7.45	2.42	−0.91	6.64	2.41	−0.90
	2.47	5.32	−0.48	2.44	3.95	−0.49	2.43	3.25	−0.50
	−1.05	−0.53	6.04	−1.02	−0.55	4.47	−1.01	−0.55	3.67
13	−2.55	2.92	−4.94	−3.05	2.89	−5.06	−3.30	2.88	−5.11
	2.91	−3.98	−1.15	2.88	−4.66	−1.31	2.88	−4.99	−1.40
	−4.95	−1.16	−1.44	−5.06	−1.32	−2.01	−5.12	−1.41	−2.30
14	−3.23	0.98	−1.00	−3.44	1.09	−1.07	−3.54	1.13	−1.09
	0.98	−0.61	−3.76	1.09	−0.68	−3.73	1.13	−0.72	−3.72
	−1.02	−3.77	−1.16	−1.08	−3.75	−1.40	−1.11	−3.73	−1.53
15	5.24	−1.77	0.78	5.31	−1.72	0.85	5.34	−1.69	0.89
	−1.77	−3.55	0.26	−1.72	−3.87	0.25	−1.69	−4.04	0.25
	0.78	0.27	−4.39	0.85	0.26	−4.61	0.89	0.25	−4.73
16	1.86	−0.71	4.51	1.66	−0.68	4.61	1.55	−0.65	4.65
	−0.71	−3.97	−0.59	−0.68	−4.24	−0.49	−0.66	−4.37	−0.43
	4.50	−0.59	−0.51	4.61	−0.49	−0.74	4.65	−0.43	−0.85
17	1.64	0.88	1.07	0.91	0.94	1.07	0.50	0.97	1.07
	0.88	−2.67	0.14	0.94	−3.39	0.17	0.97	−3.79	0.18
	1.06	0.16	−2.88	1.06	0.18	−3.58	1.06	0.20	−3.98
18	−1.34	1.99	0.55	−1.63	1.99	0.55	−1.80	1.99	0.55
	1.97	−0.15	0.74	1.97	−0.49	0.72	1.97	−0.69	0.71
	0.58	0.76	−2.32	0.58	0.74	−2.67	0.58	0.73	−2.87
19	−1.72	2.21	1.76	−1.87	2.20	1.83	−1.94	2.19	1.86
	2.20	−0.09	2.98	2.19	−0.20	2.99	2.19	−0.25	2.99
	1.76	2.99	−0.17	1.83	3.00	−0.34	1.86	3.00	−0.43

20	−0.13	0.00	0.00	−0.20	0.00	0.00	−0.21	0.00	0.00
	0.00	2.44	0.81	0.00	2.37	0.80	0.00	2.35	0.79
	0.00	0.82	0.20	0.00	0.81	0.13	0.00	0.80	0.12
21	−1.40	0.00	0.00	−1.42	0.00	0.00	−1.42	0.00	0.00
	0.00	3.24	−0.38	0.00	3.28	−0.38	0.00	3.30	−0.38
	0.00	−0.39	−1.51	0.00	−0.39	−1.48	0.00	−0.39	−1.47
22	−1.72	−2.20	−1.75	−1.87	−2.19	−1.82	−1.94	−2.19	−1.85
	−2.20	−0.09	2.98	−2.19	−0.19	2.99	−2.18	−0.24	2.99
	−1.75	2.98	−0.18	−1.82	3.00	−0.35	−1.85	2.99	−0.43
23	−1.35	−1.99	−0.54	−1.63	−1.99	−0.55	−1.81	−1.98	−0.55
	−1.97	−0.15	0.74	−1.97	−0.48	0.72	−1.97	−0.68	0.71
	−0.58	0.76	−2.33	−0.58	0.74	−2.68	−0.58	0.72	−2.88
24	5.24	1.76	−0.79	5.32	1.71	−0.86	5.35	1.68	−0.90
	1.76	−3.56	0.26	1.72	−3.87	0.25	1.68	−4.05	0.25
	−0.79	0.27	−4.39	−0.86	0.26	−4.61	−0.90	0.25	−4.73
25	1.64	−0.89	−1.07	0.91	−0.95	−1.07	0.50	−0.98	−1.07
	−0.88	−2.67	0.14	−0.95	−3.38	0.17	−0.98	−3.79	0.18
	−1.07	0.16	−2.87	−1.06	0.18	−3.58	−1.06	0.20	−3.98
26	1.85	0.70	−4.51	1.65	0.67	−4.61	1.54	0.64	−4.66
	0.70	−3.97	−0.59	0.67	−4.24	−0.48	0.65	−4.37	−0.42
	−4.51	−0.59	−0.50	−4.61	−0.48	−0.73	−4.65	−0.42	−0.85
27	−3.24	−0.98	1.00	−3.44	−1.09	1.07	−3.54	−1.13	1.09
	−0.98	−0.61	−3.76	−1.09	−0.68	−3.73	−1.13	−0.72	−3.72
	1.01	−3.77	−1.16	1.08	−3.75	−1.40	1.11	−3.73	−1.53
28	9.04	−2.45	0.92	7.43	−2.42	0.90	6.63	−2.41	0.90
	−2.47	5.31	−0.47	−2.44	3.94	−0.49	−2.43	3.24	−0.50
	1.05	−0.53	6.03	1.02	−0.54	4.45	1.01	−0.55	3.66
29	−2.55	−2.92	4.94	−3.04	−2.89	5.06	−3.29	−2.89	5.12
	−2.92	−3.97	−1.15	−2.89	−4.66	−1.31	−2.88	−4.99	−1.40
	4.95	−1.16	−1.45	5.07	−1.33	−2.01	5.12	−1.41	−2.31

Table S13: Hyperfine coupling tensors of the radical *syn*-(*R*)-**1b** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	0.21	0.01	−0.02	0.21	0.01	−0.02	0.20	0.01	−0.02
	0.01	−0.10	0.00	0.01	−0.10	0.00	0.01	−0.10	0.00
	−0.02	0.00	−0.10	−0.02	0.00	−0.10	−0.02	0.00	−0.10
2	0.28	−0.01	0.11	0.27	−0.01	0.11	0.27	−0.01	0.10
	−0.01	−0.15	0.00	−0.01	−0.15	0.00	−0.01	−0.15	0.00
	0.11	0.00	−0.12	0.11	0.00	−0.12	0.10	0.00	−0.12
3	0.28	0.07	−0.05	0.27	0.07	−0.05	0.27	0.07	−0.05
	0.07	−0.14	−0.01	0.07	−0.13	−0.01	0.07	−0.13	−0.01
	−0.05	−0.01	−0.14	−0.05	−0.01	−0.14	−0.05	−0.01	−0.14
4	0.35	0.09	0.10	0.35	0.09	0.10	0.35	0.09	0.10
	0.09	−0.18	0.02	0.09	−0.18	0.02	0.09	−0.18	0.02
	0.10	0.02	−0.17	0.10	0.02	−0.17	0.10	0.02	−0.17
5	0.26	−0.01	−0.09	0.26	−0.01	−0.09	0.25	−0.01	−0.09
	−0.01	−0.14	0.00	−0.01	−0.14	0.00	−0.01	−0.14	0.00
	−0.09	0.00	−0.12	−0.09	0.00	−0.12	−0.09	0.00	−0.12
6	0.35	−0.11	0.01	0.35	−0.11	0.01	0.35	−0.11	0.01
	−0.11	−0.17	0.00	−0.11	−0.17	0.00	−0.11	−0.17	0.00
	0.01	0.00	−0.19	0.01	0.00	−0.19	0.01	0.00	−0.19
7	1.09	0.03	0.40	1.08	0.03	0.39	1.08	0.03	0.39
	0.03	−0.64	0.01	0.03	−0.64	0.01	0.03	−0.64	0.01
	0.40	0.01	−0.51	0.39	0.01	−0.50	0.39	0.01	−0.50
8	2.63	−0.19	−1.65	2.67	−0.19	−1.65	2.69	−0.19	−1.64
	−0.19	−1.44	0.07	−0.19	−1.45	0.07	−0.19	−1.44	0.07
	−1.65	0.07	−0.94	−1.65	0.07	−0.91	−1.65	0.07	−0.89
9	0.13	2.01	−0.13	−0.21	2.01	−0.15	−0.41	2.01	−0.16
	2.02	−1.50	−0.13	2.02	−1.77	−0.13	2.02	−1.94	−0.13
	−0.12	−0.09	−2.59	−0.14	−0.10	−2.92	−0.15	−0.10	−3.10

10	0.01	−2.06	0.15	−0.33	−2.06	0.13	−0.54	−2.06	0.12
	−2.07	−1.33	−0.03	−2.07	−1.61	−0.04	−2.07	−1.78	−0.04
	0.16	−0.07	−2.59	0.14	−0.07	−2.92	0.13	−0.07	−3.11
11	1.63	2.71	2.29	1.53	2.71	2.28	1.48	2.71	2.26
	2.71	−1.64	0.98	2.72	−1.79	1.05	2.72	−1.86	1.08
	2.29	0.98	−1.92	2.28	1.05	−2.11	2.27	1.08	−2.19
12	1.45	−2.50	2.63	1.35	−2.51	2.62	1.29	−2.52	2.60
	−2.51	−1.77	−1.08	−2.52	−1.94	−1.16	−2.52	−2.01	−1.19
	2.63	−1.09	−1.65	2.62	−1.16	−1.82	2.61	−1.19	−1.89
13	−2.73	0.40	−3.34	−2.80	0.47	−3.37	−2.83	0.51	−3.38
	0.40	−3.43	−0.87	0.48	−3.64	−0.97	0.51	−3.76	−1.01
	−3.35	−0.89	1.05	−3.38	−0.98	0.83	−3.40	−1.02	0.69
14	5.32	2.13	−0.37	3.89	2.10	−0.43	3.17	2.08	−0.46
	2.14	8.88	−1.73	2.11	7.24	−1.71	2.10	6.43	−1.69
	−0.43	−1.87	6.62	−0.49	−1.83	5.04	−0.52	−1.81	4.24
15	−2.75	−0.83	−3.24	−2.82	−0.92	−3.25	−2.86	−0.96	−3.26
	−0.84	−3.08	1.51	−0.93	−3.27	1.60	−0.96	−3.37	1.64
	−3.25	1.53	0.69	−3.26	1.62	0.44	−3.27	1.65	0.30
16	−2.61	1.23	−0.02	−3.29	1.29	0.00	−3.67	1.32	0.01
	1.24	1.64	0.43	1.29	0.92	0.42	1.32	0.52	0.41
	0.00	0.43	−3.07	0.02	0.41	−3.77	0.03	0.40	−4.17
17	5.27	−2.04	−0.05	3.82	−2.02	−0.12	3.10	−2.02	−0.15
	−2.06	9.51	1.36	−2.04	7.85	1.34	−2.04	7.02	1.34
	−0.11	1.49	6.27	−0.17	1.47	4.69	−0.20	1.45	3.89
18	−3.95	0.77	0.50	−4.15	0.84	0.60	−4.24	0.88	0.65
	0.78	2.44	4.27	0.85	2.25	4.34	0.88	2.15	4.37
	0.50	4.26	−1.07	0.60	4.34	−1.37	0.65	4.37	−1.52
19	−2.70	−1.08	0.11	−3.38	−1.13	0.14	−3.76	−1.16	0.15
	−1.08	1.51	−1.03	−1.14	0.81	−1.01	−1.17	0.41	−1.01
	0.13	−1.02	−2.89	0.15	−1.00	−3.59	0.17	−0.99	−3.99

20	−4.34	1.38	−0.46	−5.11	1.32	−0.57	−5.48	1.31	−0.63
	1.39	−3.36	−5.39	1.33	−3.87	−5.49	1.31	−4.13	−5.54
	−0.48	−5.40	−0.28	−0.58	−5.50	−0.75	−0.64	−5.55	−0.99
21	−3.97	−0.48	0.41	−4.18	−0.54	0.51	−4.28	−0.57	0.57
	−0.49	1.39	−4.61	−0.54	1.18	−4.70	−0.58	1.07	−4.74
	0.41	−4.61	0.08	0.51	−4.70	−0.21	0.57	−4.74	−0.35
22	−4.40	−1.36	−0.09	−5.15	−1.31	−0.20	−5.51	−1.29	−0.27
	−1.36	−1.87	5.55	−1.31	−2.34	5.65	−1.30	−2.58	5.71
	−0.10	5.56	−1.69	−0.21	5.66	−2.18	−0.28	5.71	−2.43
23	−3.52	−1.30	0.10	−3.84	−1.23	0.12	−4.01	−1.18	0.12
	−1.30	5.45	0.59	−1.22	5.53	0.62	−1.18	5.56	0.64
	0.11	0.59	−4.59	0.12	0.62	−4.82	0.13	0.64	−4.95
24	−3.41	1.60	−0.12	−3.73	1.54	−0.10	−3.90	1.50	−0.08
	1.60	5.05	−1.78	1.54	5.12	−1.86	1.50	5.15	−1.90
	−0.11	−1.79	−4.27	−0.09	−1.86	−4.48	−0.08	−1.89	−4.60
25	0.07	0.00	−0.01	0.07	0.00	−0.01	0.07	0.00	−0.01
	0.00	−0.04	0.00	0.00	−0.04	0.00	0.00	−0.04	0.00
	−0.01	0.00	−0.04	−0.01	0.00	−0.04	−0.01	0.00	−0.04
26	0.15	−0.01	0.01	0.14	−0.01	0.00	0.13	−0.01	0.00
	−0.01	−0.02	0.00	−0.01	−0.03	0.00	−0.01	−0.04	0.00
	0.01	0.00	−0.02	0.00	0.00	−0.03	0.00	0.00	−0.03
27	0.21	−0.02	−0.04	0.21	−0.02	−0.04	0.21	−0.02	−0.04
	−0.02	−0.10	0.01	−0.02	−0.10	0.00	−0.02	−0.10	0.00
	−0.04	0.01	−0.09	−0.04	0.00	−0.09	−0.04	0.00	−0.09
28	0.48	−0.03	−0.09	0.30	−0.03	−0.09	0.20	−0.03	−0.09
	−0.03	−1.13	0.01	−0.03	−1.32	0.01	−0.03	−1.42	0.01
	−0.09	0.01	−1.03	−0.09	0.01	−1.21	−0.09	0.01	−1.29
29	3.56	0.77	−0.19	3.24	0.83	−0.18	3.10	0.85	−0.17
	0.78	2.01	0.05	0.83	1.73	0.02	0.86	1.63	0.01
	−0.13	0.11	1.60	−0.12	0.09	1.30	−0.11	0.08	1.19

30	3.66	−0.85	−0.09	3.31	−0.90	−0.07	3.16	−0.92	−0.06
	−0.85	2.16	−0.09	−0.90	1.87	−0.08	−0.92	1.76	−0.07
	−0.03	−0.16	1.75	−0.01	−0.14	1.43	0.00	−0.13	1.31
31	−6.29	2.26	−0.82	−9.87	2.04	−0.72	−11.96	1.89	−0.65
	2.23	−4.05	−1.41	2.01	−8.18	−1.33	1.86	−10.60	−1.25
	−0.95	−1.78	−6.55	−0.86	−1.70	−10.95	−0.80	−1.64	−13.39
32	14.66	−0.21	1.48	14.22	−0.17	1.28	13.96	−0.14	1.18
	−0.18	15.41	−3.56	−0.14	14.96	−3.33	−0.11	14.69	−3.21
	1.48	−3.54	22.77	1.29	−3.31	21.70	1.19	−3.19	21.14
33	−6.41	−2.26	−0.49	−9.95	−2.03	−0.42	−12.02	−1.88	−0.38
	−2.24	−3.50	1.07	−2.02	−7.65	0.96	−1.88	−10.07	0.89
	−0.62	1.46	−6.86	−0.55	1.34	−11.21	−0.52	1.27	−13.61
34	14.78	0.42	1.48	14.35	0.35	1.28	14.09	0.31	1.19
	0.39	16.60	4.38	0.32	16.08	4.07	0.28	15.78	3.92
	1.48	4.36	21.90	1.29	4.06	20.91	1.20	3.90	20.38
35	7.76	−0.76	−0.27	6.70	−0.63	−0.25	6.20	−0.55	−0.25
	−0.75	11.24	1.06	−0.62	10.09	1.04	−0.55	9.54	1.03
	−0.30	0.98	7.06	−0.29	0.96	6.03	−0.28	0.96	5.55
36	7.66	0.81	−0.41	6.61	0.68	−0.37	6.12	0.61	−0.36
	0.80	10.71	−1.53	0.67	9.59	−1.49	0.59	9.07	−1.48
	−0.44	−1.45	7.24	−0.40	−1.42	6.22	−0.39	−1.41	5.75
37	−0.40	0.00	−0.01	−0.38	0.00	−0.01	−0.37	0.00	−0.01
	0.00	−0.13	0.00	0.00	−0.11	0.00	0.00	−0.11	0.00
	−0.01	0.00	−0.13	−0.01	0.00	−0.11	−0.01	0.00	−0.11
38	−5.21	−0.13	−2.23	−15.28	−0.16	−2.60	−21.46	−0.18	−2.82
	−0.11	−4.85	−6.89	−0.14	−15.20	−7.00	−0.16	−21.57	−7.01
	−1.89	−6.89	−111.60	−2.27	−7.00	−123.59	−2.49	−7.01	−130.14
39	−0.04	0.00	−0.01	−0.04	0.00	−0.01	−0.04	0.00	−0.01
	0.00	0.02	0.00	0.00	0.02	0.00	0.00	0.02	0.00
	−0.01	0.00	0.02	−0.01	0.00	0.02	−0.01	0.00	0.02

40	−0.12	0.02	0.04	−0.10	0.02	0.04	−0.10	0.02	0.04
	0.02	0.06	−0.01	0.02	0.05	−0.01	0.02	0.05	−0.01
	0.04	−0.01	0.04	0.04	−0.01	0.04	0.04	−0.01	0.04
41	57.92	0.61	9.65	44.91	0.64	10.47	36.04	0.64	10.92
	0.62	56.05	−15.73	0.65	41.15	−16.76	0.65	31.11	−17.33
	9.75	−15.74	−183.07	10.60	−16.76	−213.31	11.05	−17.33	−231.99

Table S14: Hyperfine coupling tensors of the radical *anti*-(*R*)-**1b** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	0.23	0.04	0.01	0.23	0.04	0.01	0.23	0.04	0.01
	0.04	−0.11	0.00	0.04	−0.11	0.00	0.04	−0.11	0.00
	0.01	0.00	−0.12	0.01	0.00	−0.11	0.01	0.00	−0.11
2	0.33	−0.17	0.05	0.33	−0.16	0.05	0.33	−0.16	0.05
	−0.17	−0.14	−0.02	−0.16	−0.14	−0.01	−0.16	−0.14	−0.01
	0.05	−0.02	−0.19	0.05	−0.01	−0.19	0.05	−0.01	−0.19
3	0.38	0.12	0.06	0.37	0.12	0.06	0.37	0.12	0.06
	0.12	−0.18	0.01	0.12	−0.18	0.01	0.12	−0.18	0.01
	0.06	0.01	−0.20	0.06	0.01	−0.20	0.06	0.01	−0.19
4	0.28	−0.06	0.08	0.28	−0.05	0.07	0.28	−0.05	0.07
	−0.06	−0.14	−0.01	−0.05	−0.14	−0.01	−0.05	−0.14	−0.01
	0.08	−0.01	−0.14	0.07	−0.01	−0.13	0.07	−0.01	−0.13
5	0.30	0.10	−0.06	0.30	0.10	−0.06	0.30	0.10	−0.06
	0.10	−0.14	−0.01	0.10	−0.14	−0.01	0.10	−0.14	−0.01
	−0.06	−0.01	−0.16	−0.06	−0.01	−0.15	−0.06	−0.01	−0.15
6	0.31	−0.06	−0.07	0.31	−0.05	−0.07	0.31	−0.05	−0.07
	−0.06	−0.17	0.01	−0.05	−0.16	0.01	−0.05	−0.16	0.01
	−0.07	0.01	−0.16	−0.07	0.01	−0.15	−0.07	0.01	−0.15
7	1.10	−0.28	0.27	1.09	−0.28	0.27	1.09	−0.28	0.27
	−0.28	−0.60	−0.05	−0.28	−0.60	−0.05	−0.28	−0.60	−0.05
	0.28	−0.05	−0.56	0.27	−0.05	−0.56	0.27	−0.05	−0.55
8	2.36	−0.25	−1.86	2.41	−0.25	−1.86	2.43	−0.26	−1.86
	−0.25	−1.44	0.14	−0.25	−1.44	0.13	−0.26	−1.44	0.13
	−1.87	0.14	−0.70	−1.87	0.13	−0.67	−1.87	0.13	−0.65
9	0.50	1.84	−0.02	0.16	1.85	−0.04	−0.05	1.85	−0.04
	1.85	−1.79	−0.10	1.85	−2.08	−0.09	1.86	−2.25	−0.08
	0.00	−0.06	−2.59	−0.02	−0.05	−2.92	−0.03	−0.05	−3.11

10	−0.39	−2.13	−0.32	−0.73	−2.12	−0.33	−0.92	−2.12	−0.34
	−2.14	−0.99	0.33	−2.13	−1.26	0.33	−2.12	−1.42	0.33
	−0.31	0.29	−2.54	−0.32	0.30	−2.86	−0.33	0.30	−3.04
11	2.36	2.04	2.52	2.26	2.05	2.51	2.21	2.05	2.50
	2.04	−2.36	0.61	2.05	−2.53	0.68	2.05	−2.60	0.72
	2.52	0.61	−1.98	2.52	0.69	−2.14	2.51	0.72	−2.22
12	1.20	−3.16	1.87	1.09	−3.18	1.85	1.04	−3.18	1.83
	−3.17	−0.91	−1.02	−3.19	−1.05	−1.08	−3.19	−1.11	−1.11
	1.88	−1.02	−2.26	1.86	−1.09	−2.45	1.84	−1.12	−2.53
13	−2.98	0.75	−3.14	−3.04	0.82	−3.17	−3.07	0.84	−3.18
	0.75	−3.23	−1.26	0.82	−3.43	−1.35	0.85	−3.54	−1.38
	−3.15	−1.27	1.04	−3.18	−1.36	0.79	−3.20	−1.40	0.64
14	5.84	2.37	−0.13	4.38	2.33	−0.21	3.65	2.32	−0.24
	2.40	9.14	−1.43	2.36	7.48	−1.41	2.34	6.66	−1.40
	−0.20	−1.56	6.33	−0.27	−1.53	4.76	−0.30	−1.51	3.96
15	−3.23	−0.29	−2.94	−3.32	−0.38	−2.97	−3.37	−0.42	−2.99
	−0.30	−3.39	1.15	−0.38	−3.59	1.25	−0.42	−3.69	1.29
	−2.96	1.16	1.53	−2.99	1.26	1.32	−3.00	1.31	1.19
16	−2.37	1.60	0.23	−3.02	1.65	0.25	−3.39	1.68	0.26
	1.60	1.15	0.98	1.65	0.44	0.96	1.68	0.04	0.94
	0.24	0.97	−2.88	0.26	0.94	−3.58	0.27	0.93	−3.98
17	4.97	−1.51	−0.08	3.52	−1.50	−0.16	2.80	−1.50	−0.20
	−1.53	9.35	1.94	−1.52	7.70	1.92	−1.52	6.88	1.91
	−0.13	2.08	6.88	−0.21	2.05	5.30	−0.24	2.03	4.51
18	−3.57	1.39	1.25	−3.76	1.45	1.35	−3.84	1.48	1.41
	1.40	1.04	4.44	1.46	0.81	4.52	1.49	0.69	4.54
	1.25	4.44	0.13	1.35	4.51	−0.17	1.41	4.54	−0.32
19	−2.89	−0.71	−0.10	−3.56	−0.77	−0.08	−3.94	−0.80	−0.07
	−0.72	1.85	−0.25	−0.78	1.16	−0.23	−0.81	0.77	−0.22
	−0.08	−0.24	−3.10	−0.07	−0.22	−3.79	−0.06	−0.21	−4.18

20	−4.13	1.27	−0.56	−4.90	1.24	−0.66	−5.28	1.24	−0.73
	1.27	−2.03	−5.61	1.25	−2.49	−5.69	1.25	−2.73	−5.74
	−0.57	−5.61	−1.78	−0.67	−5.70	−2.26	−0.74	−5.74	−2.50
21	−3.98	−0.25	0.16	−4.20	−0.33	0.24	−4.29	−0.37	0.29
	−0.26	2.84	−4.18	−0.33	2.67	−4.26	−0.37	2.58	−4.30
	0.16	−4.18	−1.32	0.24	−4.26	−1.65	0.29	−4.30	−1.81
22	−4.60	−0.89	0.44	−5.36	−0.79	0.38	−5.74	−0.75	0.33
	−0.89	−3.39	5.38	−0.79	−3.92	5.48	−0.76	−4.18	5.53
	0.44	5.38	0.04	0.37	5.49	−0.42	0.32	5.54	−0.65
23	−3.68	−0.43	0.04	−3.97	−0.32	0.08	−4.12	−0.25	0.10
	−0.43	5.24	2.04	−0.32	5.29	2.10	−0.25	5.31	2.14
	0.04	2.04	−4.20	0.08	2.11	−4.42	0.10	2.14	−4.53
24	−3.07	2.29	0.00	−3.38	2.26	0.02	−3.56	2.23	0.03
	2.29	5.05	−0.42	2.26	5.14	−0.45	2.23	5.19	−0.46
	0.01	−0.43	−4.58	0.02	−0.45	−4.81	0.03	−0.46	−4.93
25	0.08	0.02	0.00	0.08	0.02	0.00	0.08	0.02	0.00
	0.02	−0.04	0.00	0.02	−0.04	0.00	0.02	−0.04	0.00
	0.00	0.00	−0.05	0.00	0.00	−0.04	0.00	0.00	−0.04
26	0.15	−0.01	0.00	0.14	−0.01	−0.01	0.13	−0.01	−0.01
	−0.01	−0.02	0.00	−0.01	−0.03	0.00	−0.01	−0.04	0.00
	0.00	0.00	−0.02	−0.01	0.00	−0.03	−0.01	0.00	−0.03
27	0.21	−0.01	−0.05	0.20	−0.01	−0.05	0.20	−0.01	−0.05
	−0.01	−0.10	0.00	−0.01	−0.10	0.00	−0.01	−0.10	0.00
	−0.05	0.00	−0.09	−0.05	0.00	−0.08	−0.05	0.00	−0.08
28	0.44	−0.16	−0.20	0.26	−0.16	−0.20	0.17	−0.16	−0.20
	−0.16	−1.12	0.01	−0.16	−1.31	0.01	−0.16	−1.40	0.01
	−0.19	0.01	−1.01	−0.20	0.01	−1.18	−0.20	0.01	−1.27
29	3.87	0.65	−0.21	3.53	0.71	−0.19	3.39	0.74	−0.18
	0.65	2.00	0.07	0.70	1.70	0.04	0.73	1.59	0.03
	−0.14	0.13	1.80	−0.12	0.10	1.47	−0.11	0.09	1.35

30	3.51	−0.93	−0.34	3.14	−0.98	−0.33	2.99	−1.00	−0.32
	−0.94	2.37	0.06	−0.98	2.08	0.08	−1.00	1.97	0.10
	−0.29	−0.01	1.78	−0.27	0.01	1.46	−0.26	0.02	1.34
31	−5.81	2.46	−0.64	−9.39	2.18	−0.62	−11.48	1.99	−0.59
	2.47	−3.87	−1.14	2.19	−7.94	−1.01	2.01	−10.32	−0.93
	−0.80	−1.50	−6.68	−0.79	−1.37	−11.02	−0.77	−1.29	−13.42
32	14.80	−0.51	1.39	14.38	−0.43	1.20	14.12	−0.38	1.11
	−0.49	16.79	−4.61	−0.41	16.25	−4.27	−0.36	15.94	−4.11
	1.39	−4.59	21.52	1.21	−4.26	20.58	1.12	−4.09	20.07
33	−6.76	−1.79	−0.67	−10.29	−1.64	−0.63	−12.35	−1.54	−0.60
	−1.78	−3.50	1.71	−1.64	−7.67	1.62	−1.54	−10.11	1.53
	−0.76	2.12	−6.24	−0.73	2.02	−10.65	−0.70	1.93	−13.10
34	15.07	0.43	2.41	14.59	0.38	2.14	14.30	0.35	2.01
	0.41	15.23	3.11	0.36	14.80	2.92	0.33	14.54	2.82
	2.41	3.09	22.78	2.15	2.90	21.73	2.02	2.81	21.17
35	7.32	−0.30	−0.24	6.33	−0.19	−0.21	5.87	−0.12	−0.19
	−0.28	10.61	1.66	−0.17	9.49	1.60	−0.10	8.96	1.58
	−0.28	1.58	7.20	−0.25	1.53	6.19	−0.23	1.51	5.72
36	7.73	1.06	−0.41	6.65	0.92	−0.39	6.15	0.85	−0.38
	1.05	10.87	−0.97	0.91	9.76	−0.94	0.84	9.25	−0.93
	−0.43	−0.88	6.89	−0.42	−0.86	5.87	−0.41	−0.85	5.40
37	−0.40	0.03	0.01	−0.38	0.03	0.01	−0.37	0.03	0.01
	0.03	−0.13	0.00	0.03	−0.12	0.00	0.03	−0.11	0.00
	0.01	0.00	−0.13	0.00	0.00	−0.11	0.00	0.00	−0.11
38	−6.21	0.90	−8.95	−16.29	0.92	−9.44	−22.47	0.93	−9.69
	0.88	−5.63	9.75	0.90	−15.94	9.94	0.90	−22.28	9.98
	−8.59	9.71	−110.66	−9.11	9.91	−122.53	−9.34	9.94	−129.01
39	−0.04	0.01	−0.01	−0.04	0.01	−0.01	−0.04	0.01	−0.01
	0.01	0.02	0.00	0.01	0.02	0.00	0.01	0.02	0.00
	−0.01	0.00	0.02	−0.01	0.00	0.02	−0.01	0.00	0.02

40	−0.12	0.00	0.05	−0.10	0.00	0.05	−0.09	0.00	0.05
	0.00	0.06	0.00	0.00	0.06	0.00	−0.01	0.06	0.00
	0.05	0.00	0.04	0.05	0.00	0.03	0.05	0.00	0.03
41	58.17	0.36	−5.75	45.20	0.19	−6.02	36.34	0.08	−6.21
	0.35	55.36	20.50	0.18	40.46	21.81	0.07	30.41	22.56
	−5.62	20.49	−182.72	−5.92	21.81	−212.97	−6.07	22.53	−231.65

Table S15: Hyperfine coupling tensors of the radical **2a** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	−1.29	−0.36	0.07	−0.78	−0.40	0.08	−0.34	−0.42	0.07
	−0.37	0.07	0.04	−0.41	0.76	0.04	−0.43	1.26	0.02
	0.07	0.04	3.10	0.08	0.04	3.57	0.07	0.02	3.00
2	−1.29	−0.36	0.07	−0.78	−0.40	0.08	−0.34	−0.42	0.07
	−0.37	0.07	0.04	−0.41	0.76	0.04	−0.43	1.26	0.02
	0.07	0.04	3.10	0.08	0.04	3.57	0.07	0.02	3.00
3	−1.29	−0.36	0.07	−0.78	−0.40	0.08	−0.34	−0.42	0.07
	−0.37	0.07	0.04	−0.41	0.76	0.04	−0.43	1.26	0.02
	0.07	0.04	3.10	0.08	0.04	3.57	0.07	0.02	3.00
4	7.02	0.18	−0.44	3.29	0.18	−0.42	1.30	0.19	−0.41
	0.13	7.04	−1.63	0.14	3.16	−1.59	0.15	1.06	−1.54
	−0.43	−1.64	−37.16	−0.42	−1.59	−40.48	−0.41	−1.54	−41.44
5	0.46	0.18	−0.04	3.18	0.20	−0.05	5.45	0.21	−0.05
	0.18	−0.50	−0.02	0.20	2.22	−0.03	0.21	4.43	−0.04
	−0.04	−0.02	−1.25	−0.05	−0.03	1.31	−0.05	−0.04	3.45
6	−18.68	−0.03	−0.21	−26.32	0.10	−0.46	−32.94	0.17	−0.62
	−0.03	−13.82	−0.69	0.09	−21.06	−1.30	0.17	−27.43	−1.67
	−0.21	−0.69	−35.82	−0.46	−1.28	−62.56	−0.62	−1.67	−81.45
7	−7.64	0.96	−0.05	−8.67	0.92	−0.05	−9.15	0.89	−0.06
	0.96	−6.28	−0.03	0.91	−7.55	−0.04	0.89	−8.20	−0.04
	−0.05	−0.03	−7.48	−0.05	−0.03	−8.87	−0.05	−0.03	−9.53
8	−10.72	−0.49	−0.03	−13.06	−0.45	−0.05	−14.43	−0.42	−0.06
	−0.49	−9.23	−0.13	−0.45	−11.86	−0.17	−0.42	−13.45	−0.17
	−0.04	−0.13	−14.39	−0.05	−0.17	−18.39	−0.06	−0.18	−20.10
9	−6.71	−1.17	0.02	−7.59	−1.06	0.00	−8.00	−0.98	−0.01
	−1.17	−6.65	−0.05	−1.06	−7.73	−0.06	−0.98	−8.26	−0.06
	0.02	−0.05	−8.08	0.00	−0.06	−9.34	0.00	−0.06	−9.90

10	−13.76	0.29	1.24	−5.04	0.45	1.41	1.48	0.55	1.53
	0.25	−12.92	3.04	0.41	−3.64	3.35	0.51	3.25	3.55
	1.24	3.04	85.36	1.41	3.34	104.52	1.53	3.55	118.02
11	−3.71	0.70	1.21	−11.00	0.86	1.10	−15.79	0.95	1.04
	0.70	−7.51	0.31	0.86	−14.45	0.40	0.95	−18.99	0.44
	1.19	0.31	−8.41	1.07	0.40	−15.44	1.02	0.44	−20.01
12	−2.87	0.64	−1.30	−9.39	0.84	−1.21	−13.68	0.96	−1.16
	0.64	−6.94	−0.33	0.84	−13.15	−0.43	0.96	−17.21	−0.47
	−1.29	−0.34	−7.56	−1.19	−0.43	−13.82	−1.14	−0.47	−17.89
13	1.53	1.19	−0.17	0.80	0.92	−0.16	0.22	0.73	−0.15
	1.20	−3.25	−0.06	0.92	−4.00	−0.06	0.74	−4.56	−0.06
	−0.17	−0.06	−3.53	−0.16	−0.06	−4.29	−0.15	−0.06	−4.87
14	−2.45	1.18	−0.06	−2.53	1.51	−0.07	−2.59	1.73	−0.08
	1.19	4.82	−0.29	1.52	5.30	−0.31	1.73	5.53	−0.32
	−0.06	−0.29	−2.79	−0.07	−0.31	−2.80	−0.08	−0.32	−2.82
15	18.73	2.60	−1.40	16.48	2.68	−1.32	15.05	2.71	−1.27
	2.59	20.03	−1.60	2.68	17.77	−1.60	2.71	16.31	−1.60
	−1.44	−1.66	16.94	−1.36	−1.66	14.50	−1.31	−1.66	12.95
16	19.89	2.66	1.23	17.56	2.74	1.14	16.07	2.77	1.09
	2.66	21.43	1.40	2.74	19.08	1.39	2.76	17.54	1.38
	1.26	1.46	17.92	1.18	1.44	15.38	1.12	1.43	13.76
17	25.14	−2.76	1.27	22.50	−2.86	1.37	20.89	−2.91	1.43
	−2.76	26.00	−1.60	−2.86	23.16	−1.45	−2.91	21.41	−1.37
	1.28	−1.62	22.95	1.38	−1.47	19.98	1.43	−1.38	18.19
18	21.72	−2.84	−0.99	19.46	−2.98	−1.11	18.09	−3.06	−1.17
	−2.83	23.36	1.37	−2.98	20.90	1.23	−3.05	19.39	1.15
	−1.00	1.39	19.46	−1.11	1.25	16.97	−1.17	1.17	15.47
19	3.28	−2.50	−0.10	3.51	−2.57	−0.09	3.59	−2.59	−0.08
	−2.50	−1.38	0.07	−2.57	−1.11	0.07	−2.59	−0.95	0.07
	−0.10	0.07	−2.07	−0.09	0.07	−1.78	−0.09	0.07	−1.64

20	−40.77	8.43	−0.03	−49.94	9.72	−0.04	−56.27	10.61	−0.05
	8.40	−14.19	−0.39	9.69	−20.34	−0.46	10.59	−24.62	−0.50
	−0.03	−0.39	−25.07	−0.04	−0.46	−32.50	−0.05	−0.50	−37.62

Table S16: Hyperfine coupling tensors of the radical (*R*)-**2b** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	1.43	0.38	−2.11	2.49	0.41	−2.43	2.90	0.33	−2.11
	0.38	−1.60	−0.80	0.40	−1.15	−0.90	0.32	−0.68	−0.83
	−2.11	−0.80	2.43	−2.43	−0.90	3.52	−2.11	−0.83	3.73
2	0.43	−0.05	0.01	0.95	0.02	−0.03	1.27	0.06	−0.05
	−0.05	−0.04	0.28	0.02	0.26	0.28	0.07	0.45	0.29
	0.01	0.28	0.14	−0.03	0.28	0.48	−0.06	0.29	0.70
3	0.17	−0.18	0.04	0.32	−0.20	0.06	0.40	−0.22	0.06
	−0.18	0.16	−0.02	−0.20	0.28	−0.03	−0.21	0.34	−0.04
	0.04	−0.02	0.29	0.06	−0.03	0.41	0.06	−0.04	0.48
4	−15.57	−5.31	33.53	−25.28	−6.12	38.52	−32.29	−6.66	41.93
	−5.38	10.00	6.34	−6.18	4.62	7.15	−6.72	0.59	7.68
	33.50	6.41	−32.59	38.50	7.21	−44.60	41.91	7.74	−53.16
5	−13.76	−4.12	25.04	−18.89	−4.17	25.43	−21.39	−4.10	25.13
	−4.16	6.23	4.86	−4.20	1.44	5.03	−4.13	−1.25	5.03
	25.04	4.88	−23.33	25.43	5.03	−28.80	25.14	5.03	−31.27
6	−12.91	−4.28	25.73	−18.46	−4.42	26.25	−21.19	−4.41	25.99
	−4.26	6.44	5.52	−4.41	1.53	5.64	−4.40	−1.25	5.58
	25.73	5.48	−25.03	26.24	5.61	−30.59	25.98	5.57	−33.12
7	−20.33	−5.48	36.79	−30.83	−6.10	42.03	−38.25	−6.49	45.53
	−5.40	10.36	7.00	−6.02	4.39	8.13	−6.42	−0.01	8.88
	36.81	6.93	−31.40	42.05	8.07	−43.72	45.54	8.81	−52.38
8	0.88	0.11	−0.02	1.73	0.21	0.00	2.32	0.28	0.01
	0.12	0.18	−0.08	0.21	0.98	−0.06	0.28	1.52	−0.04
	−0.02	−0.08	0.82	−0.01	−0.06	1.51	0.01	−0.04	2.00
9	−13.59	−0.61	3.24	−17.75	−0.80	4.22	−19.98	−0.82	4.41
	−0.61	−13.19	0.37	−0.79	−16.36	0.65	−0.82	−18.21	0.72
	3.22	0.37	−14.94	4.20	0.65	−19.49	4.39	0.73	−21.78

10	−18.10	−1.93	11.80	−33.09	−3.35	22.30	−44.70	−4.26	29.12
	−1.94	−14.34	0.95	−3.36	−21.70	2.55	−4.27	−28.37	3.57
	11.84	0.95	−23.29	22.34	2.55	−42.60	29.16	3.57	−57.01
11	−8.31	−0.98	0.63	−10.18	−0.93	0.75	−11.14	−0.89	0.78
	−0.98	−8.61	−0.69	−0.93	−10.08	−0.58	−0.88	−10.83	−0.52
	0.61	−0.68	−9.21	0.72	−0.57	−11.09	0.76	−0.51	−12.02
12	−8.15	0.69	0.96	−9.87	0.65	1.06	−10.75	0.62	1.07
	0.70	−8.79	0.43	0.65	−10.13	0.46	0.62	−10.80	0.47
	0.99	0.43	−7.96	1.08	0.47	−9.78	1.10	0.47	−10.70
13	−0.79	0.01	0.26	1.68	−0.01	0.25	3.80	−0.03	0.25
	0.01	0.29	0.11	−0.01	2.86	0.10	−0.03	5.10	0.10
	0.26	0.11	−1.07	0.25	0.10	1.36	0.25	0.11	3.46
14	−1.66	0.06	−0.29	−2.66	0.17	−0.35	−3.36	0.25	−0.40
	0.07	−0.63	0.02	0.18	−1.60	0.13	0.25	−2.28	0.20
	−0.29	0.02	−1.35	−0.35	0.13	−2.37	−0.40	0.20	−3.07
15	0.36	0.48	−0.10	0.34	0.49	−0.09	0.34	0.49	−0.09
	0.48	0.65	−0.20	0.48	0.63	−0.23	0.48	0.64	−0.25
	−0.10	−0.20	0.10	−0.09	−0.23	0.13	−0.09	−0.25	0.16
16	0.10	0.14	−0.03	0.09	0.14	−0.03	0.09	0.14	−0.03
	0.14	0.00	−0.03	0.14	0.01	−0.03	0.14	0.01	−0.03
	−0.03	−0.03	−0.09	−0.03	−0.03	−0.09	−0.03	−0.03	−0.09
17	0.05	0.09	−0.04	0.04	0.09	−0.04	0.04	0.09	−0.04
	0.09	0.03	−0.04	0.09	0.03	−0.04	0.09	0.03	−0.04
	−0.04	−0.04	−0.04	−0.04	−0.04	−0.04	−0.04	−0.04	−0.04
18	0.23	0.12	−0.09	0.20	0.12	−0.09	0.19	0.12	−0.09
	0.12	−0.02	−0.05	0.12	−0.06	−0.05	0.12	−0.07	−0.05
	−0.09	−0.05	−0.04	−0.09	−0.05	−0.07	−0.10	−0.05	−0.08
19	0.06	0.08	0.00	0.06	0.08	0.00	0.05	0.09	0.00
	0.08	−0.02	0.00	0.08	−0.02	0.00	0.09	−0.02	0.00
	0.00	0.00	−0.07	0.00	0.00	−0.07	0.00	0.00	−0.07

20	0.03	−0.04	0.01	0.10	−0.03	0.02	0.16	−0.03	0.02
	−0.05	0.47	−0.01	−0.03	0.51	−0.01	−0.03	0.57	−0.01
	0.01	−0.01	0.01	0.02	−0.01	0.06	0.03	−0.01	0.12
21	0.08	0.04	−0.01	0.04	0.04	−0.01	0.02	0.04	−0.01
	0.04	0.36	−0.03	0.04	0.30	−0.02	0.04	0.27	−0.01
	−0.01	−0.02	0.07	−0.01	−0.02	0.02	−0.01	−0.01	−0.01
22	−0.04	0.02	0.00	−0.04	0.02	0.01	−0.03	0.02	0.01
	0.02	0.10	−0.01	0.02	0.10	−0.01	0.02	0.11	−0.01
	0.00	−0.01	−0.04	0.01	−0.01	−0.03	0.01	−0.01	−0.02
23	−0.04	0.00	−0.01	−0.05	0.00	−0.01	−0.05	0.00	−0.02
	0.00	0.09	0.01	0.00	0.08	0.01	0.00	0.08	0.01
	−0.01	0.01	−0.05	−0.01	0.01	−0.06	−0.02	0.01	−0.06
24	−0.02	−0.03	0.00	−0.03	−0.03	0.00	−0.03	−0.03	0.00
	−0.03	0.12	0.02	−0.03	0.11	0.02	−0.03	0.11	0.01
	0.00	0.02	−0.02	0.00	0.02	−0.03	0.00	0.01	−0.03
25	−0.06	−0.07	−0.02	−0.12	−0.06	−0.02	−0.15	−0.06	−0.03
	−0.07	0.18	0.04	−0.06	0.12	0.04	−0.06	0.08	0.04
	−0.02	0.04	−0.10	−0.02	0.04	−0.15	−0.03	0.04	−0.18
26	−0.84	0.99	0.44	−0.77	0.92	0.35	−0.72	0.87	0.29
	0.98	1.65	0.94	0.91	1.96	0.88	0.87	2.16	0.84
	0.44	0.94	−0.80	0.35	0.89	−0.74	0.29	0.85	−0.70
27	37.72	−3.07	0.92	34.24	−3.36	1.05	31.94	−3.52	1.12
	−3.06	34.21	−0.44	−3.36	31.08	−0.40	−3.52	29.02	−0.36
	0.90	−0.44	32.10	1.03	−0.39	28.78	1.10	−0.36	26.61
28	22.72	−1.95	3.08	20.75	−2.08	3.15	19.43	−2.15	3.17
	−1.95	23.04	−2.48	−2.08	21.17	−2.79	−2.15	19.93	−2.97
	3.10	−2.47	24.47	3.18	−2.79	22.20	3.20	−2.97	20.70
29	−0.86	−2.70	0.59	−0.82	−2.84	0.56	−0.83	−2.89	0.55
	−2.71	3.80	−1.61	−2.85	3.90	−1.70	−2.90	3.89	−1.73
	0.60	−1.62	−1.54	0.57	−1.71	−1.50	0.57	−1.74	−1.51

30	34.21	0.82	2.50	30.68	0.84	2.66	28.38	0.85	2.74
	0.82	33.92	1.72	0.84	30.43	1.99	0.85	28.19	2.15
	2.53	1.73	38.53	2.68	2.00	34.77	2.76	2.16	32.35
31	1.60	2.78	1.66	1.49	2.89	1.68	1.39	2.93	1.68
	2.78	3.21	2.38	2.89	3.01	2.49	2.92	2.85	2.52
	1.63	2.36	0.93	1.65	2.47	0.83	1.66	2.50	0.72
32	22.44	1.26	3.02	20.74	1.61	3.20	19.61	1.82	3.29
	1.28	17.46	0.70	1.62	15.70	0.73	1.83	14.57	0.75
	3.00	0.71	17.75	3.19	0.74	16.08	3.28	0.76	14.98
33	−8.49	−0.96	0.36	−15.69	−1.01	0.50	−20.43	−1.06	0.57
	−0.96	−5.05	1.75	−1.02	−12.64	1.57	−1.07	−17.67	1.46
	0.36	1.75	−8.13	0.50	1.56	−15.44	0.57	1.45	−20.22
34	0.71	0.57	−0.16	0.73	0.72	−0.20	0.83	0.83	−0.22
	0.57	4.75	−1.35	0.72	4.78	−1.24	0.83	4.89	−1.15
	−0.16	−1.36	1.14	−0.19	−1.24	0.90	−0.22	−1.16	0.83
35	−1.25	−1.68	0.10	−1.35	−1.66	−0.01	−1.38	−1.64	−0.09
	−1.68	3.12	−0.40	−1.65	3.49	−0.30	−1.63	3.76	−0.23
	0.10	−0.40	−1.80	−0.01	−0.30	−1.75	−0.08	−0.23	−1.69
36	0.05	0.32	−0.09	0.04	0.31	−0.09	0.04	0.31	−0.08
	0.32	0.15	−0.12	0.31	0.15	−0.12	0.31	0.15	−0.12
	−0.09	−0.12	−0.19	−0.09	−0.12	−0.19	−0.08	−0.12	−0.18
37	0.13	0.24	−0.14	0.12	0.24	−0.14	0.12	0.24	−0.14
	0.24	−0.01	−0.11	0.24	−0.01	−0.11	0.24	−0.01	−0.12
	−0.14	−0.11	−0.13	−0.14	−0.11	−0.13	−0.14	−0.12	−0.13
38	0.09	0.41	−0.27	0.07	0.40	−0.26	0.07	0.40	−0.25
	0.41	0.05	−0.29	0.40	0.06	−0.29	0.40	0.06	−0.29
	−0.27	−0.29	−0.15	−0.26	−0.29	−0.15	−0.25	−0.29	−0.15
39	0.46	0.22	−0.24	0.45	0.22	−0.24	0.44	0.23	−0.24
	0.22	−0.22	−0.08	0.22	−0.24	−0.08	0.23	−0.24	−0.08
	−0.24	−0.08	−0.21	−0.24	−0.08	−0.22	−0.25	−0.08	−0.22

40	1.13	0.41	−0.20	1.14	0.42	−0.22	1.15	0.43	−0.23
	0.41	−0.44	−0.08	0.42	−0.47	−0.08	0.43	−0.49	−0.07
	−0.20	−0.08	−0.53	−0.22	−0.08	−0.55	−0.23	−0.07	−0.57
41	0.76	0.49	−0.72	0.75	0.51	−0.74	0.74	0.52	−0.74
	0.49	−0.50	−0.29	0.51	−0.52	−0.30	0.52	−0.52	−0.31
	−0.72	−0.29	−0.24	−0.74	−0.30	−0.23	−0.75	−0.31	−0.22
42	0.35	0.33	0.10	0.35	0.34	0.10	0.35	0.34	0.10
	0.33	−0.09	0.05	0.34	−0.09	0.05	0.34	−0.09	0.05
	0.10	0.05	−0.26	0.10	0.05	−0.26	0.10	0.05	−0.26
43	0.22	0.22	−0.02	0.21	0.22	−0.02	0.21	0.22	−0.03
	0.22	−0.06	−0.02	0.22	−0.06	−0.02	0.22	−0.06	−0.02
	−0.02	−0.02	−0.18	−0.02	−0.02	−0.18	−0.03	−0.02	−0.18
44	0.14	0.29	0.03	0.14	0.29	0.03	0.13	0.29	0.03
	0.29	0.05	0.02	0.29	0.06	0.02	0.29	0.06	0.02
	0.03	0.02	−0.20	0.03	0.02	−0.20	0.03	0.02	−0.19
45	−0.31	0.28	−0.05	−0.30	0.27	−0.04	−0.28	0.27	−0.03
	0.28	0.65	−0.30	0.27	0.64	−0.28	0.27	0.63	−0.27
	−0.05	−0.30	−0.33	−0.04	−0.28	−0.33	−0.03	−0.27	−0.32
46	−0.14	0.08	−0.01	−0.14	0.08	0.00	−0.14	0.08	0.00
	0.08	0.28	−0.06	0.08	0.27	−0.06	0.08	0.26	−0.06
	−0.01	−0.06	−0.15	0.00	−0.06	−0.15	0.00	−0.06	−0.15
47	−0.10	−0.01	0.00	−0.10	−0.01	0.00	−0.09	−0.01	0.00
	−0.01	0.23	0.02	−0.01	0.22	0.02	−0.01	0.22	0.02
	0.00	0.02	−0.11	0.00	0.02	−0.10	0.00	0.02	−0.10
48	−0.10	−0.12	−0.02	−0.11	−0.11	−0.02	−0.12	−0.11	−0.02
	−0.12	0.29	0.09	−0.11	0.27	0.09	−0.11	0.26	0.09
	−0.02	0.09	−0.12	−0.02	0.09	−0.12	−0.02	0.09	−0.12
49	−0.28	−0.43	−0.09	−0.31	−0.40	−0.08	−0.32	−0.37	−0.08
	−0.43	0.62	0.27	−0.39	0.60	0.28	−0.37	0.60	0.29
	−0.09	0.27	−0.40	−0.08	0.28	−0.39	−0.08	0.29	−0.38

Table S17: Hyperfine coupling tensors of the radical **3a** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	−7.12	0.00	0.00	7.61	0.00	0.00	19.01	0.00	0.00
	0.00	−7.12	0.00	0.00	7.61	0.00	0.00	19.01	0.00
	0.00	0.00	135.99	0.00	0.00	168.48	0.00	0.00	187.53
2	5.54	0.24	−12.49	10.62	0.30	−14.87	15.46	0.35	−17.36
	0.24	−1.15	−0.46	0.30	2.52	−0.54	0.35	5.88	−0.64
	−12.49	−0.46	18.80	−14.88	−0.54	25.61	−17.36	−0.63	32.42
3	−6.35	0.05	3.27	−11.24	0.00	6.64	−16.05	−0.01	9.58
	0.06	−3.31	0.09	0.01	−5.80	0.26	−0.01	−8.53	0.40
	3.28	0.09	−9.12	6.65	0.27	−16.70	9.60	0.40	−23.94
4	17.58	−2.24	−12.40	24.24	−2.16	−15.28	30.02	−2.02	−17.92
	−2.25	5.21	1.55	−2.17	9.99	1.52	−2.02	14.03	1.43
	−12.40	1.55	16.32	−15.28	1.51	25.40	−17.93	1.42	33.53
5	−22.38	−0.17	4.60	−33.64	−0.25	8.98	−42.66	−0.31	12.34
	−0.17	−17.83	0.17	−0.25	−27.06	0.33	−0.31	−34.50	0.46
	4.60	0.17	−26.36	8.98	0.34	−42.99	12.34	0.46	−55.73
6	−19.12	2.05	−2.45	−28.91	2.97	−4.77	−36.80	3.69	−6.57
	2.05	−21.09	3.90	2.97	−31.77	7.61	3.68	−40.35	10.46
	−2.45	3.90	−26.36	−4.78	7.60	−42.98	−6.57	10.46	−55.74
7	6.36	−4.23	4.85	11.68	−5.08	6.32	16.28	−5.91	7.72
	−4.23	16.43	−11.51	−5.09	22.55	−13.99	−5.92	27.77	−16.24
	4.86	−11.51	16.32	6.34	−13.99	25.40	7.73	−16.24	33.53
8	−4.02	1.29	−1.71	−7.15	2.35	−3.55	−10.42	3.26	−5.14
	1.29	−5.63	2.79	2.36	−9.88	5.62	3.26	−14.16	8.10
	−1.72	2.80	−9.12	−3.56	5.63	−16.70	−5.15	8.11	−23.94
9	0.73	−3.02	6.64	4.80	−3.66	7.91	8.58	−4.32	9.23
	−3.02	3.66	−10.59	−3.66	8.34	−12.61	−4.32	12.76	−14.71
	6.64	−10.59	18.80	7.91	−12.61	25.61	9.23	−14.72	32.42

10	−4.33	1.44	−1.77	−7.53	2.54	−3.52	−10.91	3.50	−5.07
	1.43	−5.34	2.75	2.53	−9.51	5.63	3.49	−13.68	8.15
	−1.77	2.77	−9.12	−3.52	5.65	−16.70	−5.07	8.16	−23.93
11	11.22	−6.58	8.27	16.52	−7.42	9.86	20.97	−8.18	11.28
	−6.57	11.54	−9.36	−7.42	17.69	−11.76	−8.18	23.05	−14.00
	8.27	−9.37	16.31	9.86	−11.78	25.38	11.28	−14.01	33.50
12	−18.82	−1.88	−2.15	−28.49	−2.72	−4.20	−36.28	−3.38	−5.77
	−1.88	−21.39	−4.07	−2.72	−32.21	−7.94	−3.38	−40.89	−10.92
	−2.15	−4.07	−26.36	−4.20	−7.94	−42.99	−5.77	−10.92	−55.74
13	10.25	6.48	7.54	15.43	7.25	8.95	19.78	7.94	10.20
	6.47	12.54	9.96	7.25	18.80	12.47	7.93	24.28	14.80
	7.54	9.97	16.32	8.95	12.48	25.40	10.20	14.82	33.53
14	−4.12	−1.34	−1.56	−7.16	−2.36	−3.09	−10.41	−3.25	−4.45
	−1.34	−5.54	−2.87	−2.35	−9.87	−5.88	−3.25	−14.18	−8.50
	−1.56	−2.89	−9.12	−3.09	−5.90	−16.71	−4.45	−8.51	−23.94
15	0.31	2.78	5.85	4.28	3.36	6.97	7.97	3.97	8.13
	2.78	4.08	11.05	3.36	8.85	13.15	3.97	13.37	15.35
	5.85	11.05	18.80	6.97	13.16	25.61	8.13	15.35	32.43
16	−3.84	−1.15	−1.50	−6.82	−2.12	−3.12	−9.96	−2.94	−4.52
	−1.16	−5.82	−2.91	−2.13	−10.22	−5.86	−2.95	−14.63	−8.46
	−1.51	−2.92	−9.12	−3.13	−5.88	−16.70	−4.53	−8.47	−23.94
17	5.76	3.42	3.98	10.96	4.21	5.26	15.45	4.99	6.48
	3.43	16.99	11.84	4.22	23.23	14.42	4.99	28.58	16.77
	3.99	11.85	16.30	5.27	14.43	25.38	6.50	16.77	33.50
18	17.16	3.15	−12.24	23.82	3.20	−15.12	29.62	3.19	−17.76
	3.15	5.61	−2.48	3.21	10.39	−2.66	3.19	14.41	−2.77
	−12.25	−2.47	16.31	−15.13	−2.65	25.39	−17.77	−2.76	33.51
19	−6.32	−0.27	3.27	−11.21	−0.40	6.64	−16.02	−0.54	9.59
	−0.28	−3.34	0.16	−0.41	−5.83	0.23	−0.55	−8.58	0.32
	3.28	0.15	−9.12	6.65	0.23	−16.70	9.60	0.31	−23.93

20	−10.88	−0.30	−2.27	−14.21	−0.35	−2.80	−17.47	−0.41	−3.34
	−0.30	−2.87	−0.09	−0.35	−4.84	−0.11	−0.41	−6.69	−0.13
	−2.27	−0.09	−8.87	−2.80	−0.11	−11.75	−3.34	−0.13	−14.56
21	2.44	−0.07	0.67	4.44	0.66	0.50	6.55	1.34	0.36
	−0.07	4.03	0.06	0.66	7.30	0.59	1.35	10.52	1.07
	0.67	0.06	1.90	0.50	0.59	4.06	0.36	1.07	6.30
22	−4.45	0.83	2.03	−7.00	1.43	2.12	−9.39	2.13	2.17
	0.82	−7.32	0.80	1.43	−11.02	1.15	2.12	−14.40	1.54
	2.03	0.80	−7.33	2.12	1.15	−10.09	2.17	1.54	−12.55
23	−5.89	−1.65	−1.71	−8.78	−2.45	−2.06	−11.30	−3.23	−2.41
	−1.66	−5.88	1.36	−2.46	−9.24	1.26	−3.24	−12.48	1.11
	−1.71	1.36	−7.33	−2.06	1.26	−10.09	−2.42	1.11	−12.55
24	3.57	0.72	−0.38	7.16	0.90	−0.76	10.69	1.05	−1.11
	0.72	2.89	0.55	0.91	4.58	0.14	1.05	6.38	−0.22
	−0.39	0.56	1.90	−0.76	0.14	4.06	−1.11	−0.22	6.30
25	−5.13	3.62	1.21	−7.49	4.23	1.50	−9.74	4.87	1.78
	3.62	−8.62	−1.92	4.23	−11.56	−2.37	4.87	−14.43	−2.83
	1.21	−1.92	−8.87	1.49	−2.37	−11.75	1.78	−2.83	−14.56
26	3.59	0.72	−0.33	5.78	1.57	0.21	8.01	2.34	0.68
	0.71	2.88	0.59	1.57	5.97	0.75	2.34	9.06	0.90
	−0.33	0.59	1.90	0.21	0.75	4.06	0.68	0.90	6.30
27	−7.18	−1.04	−0.48	−11.07	−1.35	−0.24	−14.78	−1.56	0.05
	−1.03	−4.60	2.13	−1.35	−6.94	2.40	−1.55	−9.00	2.66
	−0.49	2.13	−7.32	−0.24	2.40	−10.09	0.05	2.66	−12.55
28	−7.32	0.83	−0.32	−11.25	1.03	−0.06	−14.98	1.11	0.25
	0.83	−4.46	−2.16	1.02	−6.76	−2.41	1.10	−8.80	−2.65
	−0.32	−2.16	−7.33	−0.06	−2.41	−10.09	0.25	−2.65	−12.55
29	3.69	−0.66	−0.29	6.01	−1.57	0.26	8.36	−2.39	0.75
	−0.65	2.78	−0.61	−1.57	5.73	−0.73	−2.39	8.71	−0.85
	−0.29	−0.61	1.90	0.26	−0.73	4.06	0.75	−0.85	6.30

30	−4.61	−3.32	1.06	−6.88	−3.88	1.31	−9.04	−4.46	1.56
	−3.32	−9.14	2.01	−3.88	−12.17	2.48	−4.46	−15.13	2.96
	1.06	2.01	−8.87	1.31	2.48	−11.75	1.56	2.95	−14.56
31	3.68	−0.66	−0.34	7.28	−0.70	−0.75	10.83	−0.71	−1.12
	−0.67	2.79	−0.58	−0.70	4.47	−0.19	−0.71	6.25	0.14
	−0.34	−0.58	1.90	−0.75	−0.20	4.06	−1.12	0.14	6.30
32	−6.15	1.63	−1.60	−9.15	2.46	−1.96	−11.80	3.28	−2.33
	1.64	−5.64	−1.48	2.47	−8.87	−1.41	3.29	−11.99	−1.29
	−1.60	−1.48	−7.33	−1.96	−1.41	−10.09	−2.33	−1.29	−12.55
33	−4.35	−0.60	2.09	−6.81	−1.12	2.20	−9.10	−1.73	2.28
	−0.59	−7.43	−0.64	−1.11	−11.21	−0.99	−1.72	−14.68	−1.37
	2.09	−0.64	−7.32	2.20	−0.99	−10.09	2.28	−1.37	−12.55
34	2.44	−0.05	0.67	4.56	−0.87	0.54	6.78	−1.63	0.44
	−0.05	4.03	−0.01	−0.87	7.19	−0.55	−1.63	10.30	−1.04
	0.68	−0.01	1.90	0.55	−0.55	4.06	0.44	−1.04	6.30

Table S18: Hyperfine coupling tensors of the radical **3b** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	−13.67	0.00	−2.92	−21.94	−0.01	−5.67	−29.42	−0.02	−7.61
	0.00	−16.14	−0.08	−0.01	−24.91	−0.19	−0.02	−32.64	−0.26
	−2.90	−0.09	−34.06	−5.65	−0.19	−68.10	−7.59	−0.26	−93.00
2	−2.20	−1.35	3.98	8.65	−1.02	4.02	18.02	−0.74	3.89
	−1.35	8.81	−27.87	−1.02	21.99	−33.81	−0.74	32.68	−37.22
	3.98	−27.89	63.71	4.03	−33.83	90.66	3.89	−37.25	109.49
3	−10.79	0.02	0.06	−17.09	0.27	0.25	−22.49	0.46	0.43
	0.02	−10.99	2.52	0.27	−19.04	5.62	0.45	−25.87	8.09
	0.06	2.51	−14.73	0.25	5.62	−26.97	0.43	8.09	−37.14
4	−1.34	0.70	−1.66	1.20	0.86	−1.98	3.80	0.99	−2.25
	0.70	2.34	−6.95	0.86	5.97	−9.29	1.00	9.55	−11.43
	−1.66	−6.95	12.16	−1.98	−9.29	18.97	−2.25	−11.43	25.49
5	−3.20	−0.37	0.06	−5.25	−0.53	0.24	−7.49	−0.67	0.40
	−0.37	−4.17	1.85	−0.53	−7.30	3.90	−0.67	−10.53	5.77
	0.06	1.85	−6.96	0.24	3.90	−13.12	0.40	5.77	−19.11
6	−1.70	0.01	−0.45	0.57	0.03	−0.67	2.82	0.06	−0.87
	0.01	2.47	−7.79	0.03	5.76	−9.66	0.06	9.00	−11.51
	−0.45	−7.80	14.19	−0.67	−9.67	20.34	−0.87	−11.51	26.40
7	−3.32	−0.06	0.02	−5.11	−0.10	0.09	−7.14	−0.14	0.15
	−0.06	−3.51	1.45	−0.10	−6.16	3.49	−0.14	−8.95	5.31
	0.02	1.45	−5.71	0.09	3.49	−11.59	0.15	5.31	−17.27
8	−2.02	−0.08	−0.34	−0.49	−0.13	−0.25	1.13	−0.16	−0.18
	−0.08	0.44	−5.27	−0.14	2.83	−7.14	−0.16	5.19	−8.83
	−0.34	−5.28	9.84	−0.25	−7.15	15.44	−0.17	−8.84	20.71
9	−1.94	0.33	−0.46	−0.36	0.37	−0.53	1.24	0.39	−0.59
	0.33	−0.63	−4.76	0.37	1.59	−6.46	0.39	3.81	−7.98
	−0.46	−4.76	10.29	−0.53	−6.46	16.07	−0.60	−7.99	21.50

10	−3.18	0.29	0.09	−4.83	0.32	0.19	−6.78	0.36	0.29
	0.29	−3.57	0.92	0.32	−5.95	2.76	0.36	−8.53	4.37
	0.09	0.92	−5.59	0.19	2.76	−11.92	0.29	4.37	−17.95
11	−1.55	0.20	−0.01	0.91	0.30	−0.10	3.29	0.38	−0.17
	0.20	1.01	−7.07	0.30	3.91	−8.59	0.39	6.79	−10.17
	−0.01	−7.07	15.62	−0.10	−8.60	21.88	−0.17	−10.17	28.17
12	−3.12	−0.04	0.19	−5.06	−0.09	0.24	−7.24	−0.13	0.29
	−0.04	−4.22	1.46	−0.09	−6.95	3.22	−0.14	−9.86	4.83
	0.19	1.46	−7.25	0.24	3.22	−13.74	0.29	4.83	−20.07
13	−1.05	0.55	−0.96	1.63	0.48	−0.94	4.23	0.41	−0.93
	0.55	1.31	−6.27	0.48	4.82	−8.16	0.41	8.19	−9.93
	−0.97	−6.27	15.32	−0.94	−8.16	22.47	−0.94	−9.93	29.32
14	−10.45	0.67	0.33	−16.85	0.51	0.66	−22.37	0.38	0.86
	0.67	−11.13	2.16	0.51	−18.07	4.97	0.39	−23.99	7.14
	0.32	2.16	−15.68	0.65	4.97	−28.26	0.86	7.14	−38.60
15	−2.21	1.29	3.77	8.65	0.95	3.78	18.01	0.66	3.62
	1.30	9.22	28.24	0.96	22.47	34.27	0.66	33.19	37.74
	3.78	28.27	63.26	3.78	34.30	90.12	3.62	37.76	108.91
16	−10.77	−0.02	0.08	−17.06	−0.25	0.30	−22.45	−0.43	0.49
	−0.01	−11.01	−2.54	−0.25	−19.10	−5.67	−0.42	−25.95	−8.16
	0.08	−2.54	−14.68	0.30	−5.66	−26.86	0.49	−8.16	−36.99
17	−1.33	−0.74	−1.71	1.21	−0.90	−2.05	3.81	−1.05	−2.33
	−0.74	2.42	6.99	−0.91	6.07	9.35	−1.06	9.68	11.50
	−1.70	7.00	12.05	−2.05	9.35	18.82	−2.33	11.51	25.31
18	−3.20	0.38	0.07	−5.25	0.54	0.27	−7.49	0.70	0.44
	0.38	−4.18	−1.86	0.54	−7.33	−3.93	0.70	−10.58	−5.82
	0.07	−1.86	−6.93	0.27	−3.93	−13.05	0.44	−5.82	−19.01
19	−1.70	−0.04	−0.50	0.57	−0.08	−0.74	2.82	−0.12	−0.96
	−0.04	2.57	7.86	−0.08	5.88	9.74	−0.11	9.14	11.60
	−0.50	7.86	14.07	−0.74	9.75	20.18	−0.96	11.61	26.22

20	−3.32	0.06	0.03	−5.11	0.11	0.12	−7.14	0.15	0.19
	0.06	−3.52	−1.46	0.11	−6.20	−3.52	0.15	−9.01	−5.36
	0.03	−1.47	−5.68	0.11	−3.52	−11.53	0.19	−5.36	−17.18
21	−2.02	0.06	−0.38	−0.49	0.11	−0.31	1.12	0.13	−0.24
	0.06	0.52	5.32	0.11	2.92	7.22	0.13	5.31	8.93
	−0.38	5.33	9.75	−0.30	7.23	15.32	−0.24	8.94	20.57
22	−1.93	−0.35	−0.50	−0.36	−0.39	−0.58	1.24	−0.42	−0.65
	−0.35	−0.57	4.82	−0.39	1.67	6.54	−0.42	3.90	8.09
	−0.50	4.82	10.21	−0.58	6.54	15.97	−0.65	8.09	21.37
23	−3.18	−0.29	0.10	−4.82	−0.31	0.22	−6.76	−0.34	0.32
	−0.29	−3.58	−0.93	−0.31	−5.99	−2.79	−0.34	−8.59	−4.43
	0.10	−0.93	−5.57	0.22	−2.79	−11.87	0.32	−4.43	−17.87
24	−1.55	−0.21	−0.06	0.91	−0.32	−0.16	3.29	−0.41	−0.24
	−0.22	1.10	7.16	−0.32	4.02	8.70	−0.41	6.91	10.29
	−0.06	7.16	15.50	−0.16	8.70	21.74	−0.24	10.30	28.01
25	−3.11	0.05	0.20	−5.06	0.10	0.27	−7.23	0.16	0.32
	0.05	−4.23	−1.47	0.10	−6.98	−3.26	0.16	−9.91	−4.89
	0.20	−1.47	−7.22	0.26	−3.26	−13.68	0.32	−4.89	−19.99
26	−1.04	−0.58	−1.01	1.64	−0.51	−0.99	4.23	−0.45	−1.00
	−0.57	1.38	6.35	−0.51	4.92	8.26	−0.44	8.30	10.05
	−1.01	6.35	15.22	−1.00	8.26	22.34	−1.01	10.05	29.16
27	−10.42	−0.66	0.35	−16.82	−0.50	0.70	−22.34	−0.37	0.91
	−0.66	−11.16	−2.19	−0.50	−18.12	−5.02	−0.37	−24.06	−7.22
	0.34	−2.19	−15.64	0.69	−5.03	−28.16	0.91	−7.23	−38.47
28	36.09	0.01	−0.49	50.66	0.04	−0.99	61.19	0.06	−1.34
	0.01	32.30	−0.01	0.04	38.81	0.00	0.06	43.46	0.00
	−0.45	−0.01	29.29	−0.94	0.00	38.84	−1.30	0.00	45.85
29	−3.20	−0.81	−0.06	−4.83	−0.99	−0.04	−6.28	−1.11	−0.02
	−0.81	−5.98	−0.31	−0.98	−9.07	−0.71	−1.11	−12.00	−1.14
	−0.06	−0.31	−5.70	−0.04	−0.71	−8.15	−0.02	−1.14	−10.39

30	1.24	−0.42	−0.26	3.40	−1.10	−0.58	5.61	−1.73	−0.87
	−0.42	1.44	0.78	−1.10	3.08	0.77	−1.73	4.76	0.76
	−0.26	0.78	0.46	−0.58	0.77	2.05	−0.87	0.76	3.69
31	−7.38	−2.31	−1.24	−10.27	−2.88	−1.56	−13.06	−3.43	−1.86
	−2.32	−3.39	0.85	−2.88	−5.28	0.93	−3.43	−7.09	1.02
	−1.24	0.85	−4.77	−1.56	0.94	−6.85	−1.86	1.02	−8.85
32	1.11	−0.14	−0.06	2.17	−0.07	−0.04	3.33	0.00	−0.01
	−0.14	1.04	0.66	−0.07	3.41	1.07	0.00	5.78	1.45
	−0.06	0.66	−0.01	−0.04	1.07	1.73	−0.01	1.45	3.51
33	1.19	0.09	0.05	2.93	0.84	0.34	4.68	1.52	0.60
	0.09	0.80	0.47	0.85	2.73	0.62	1.53	4.69	0.76
	0.05	0.47	−0.26	0.34	0.62	1.39	0.60	0.76	3.07
34	−2.93	2.39	0.93	−4.68	2.87	1.13	−6.37	3.37	1.33
	2.39	−7.39	−0.86	2.87	−10.15	−1.13	3.37	−12.89	−1.41
	0.94	−0.85	−5.63	1.13	−1.13	−7.76	1.34	−1.41	−9.86
35	1.67	0.35	0.15	4.32	0.12	0.07	6.97	−0.12	−0.01
	0.35	1.03	0.42	0.12	2.24	0.24	−0.12	3.54	0.08
	0.15	0.42	0.16	0.07	0.24	1.72	−0.01	0.08	3.35
36	−4.33	−2.27	−0.24	−6.78	−3.21	−0.60	−9.07	−4.09	−0.96
	−2.28	−4.26	0.14	−3.21	−6.85	−0.01	−4.10	−9.30	−0.16
	−0.24	0.14	−5.64	−0.61	0.00	−7.86	−0.96	−0.15	−9.97
37	−3.21	0.83	−0.07	−4.84	1.02	−0.05	−6.30	1.16	−0.04
	0.83	−5.96	0.31	1.02	−9.03	0.72	1.15	−11.95	1.15
	−0.07	0.31	−5.69	−0.05	0.71	−8.15	−0.04	1.14	−10.39
38	1.23	0.41	−0.26	3.38	1.10	−0.58	5.58	1.73	−0.88
	0.42	1.44	−0.78	1.10	3.08	−0.79	1.73	4.77	−0.77
	−0.26	−0.78	0.47	−0.58	−0.79	2.05	−0.88	−0.77	3.70
39	−7.41	2.27	−1.25	−10.30	2.82	−1.57	−13.10	3.37	−1.88
	2.27	−3.36	−0.87	2.82	−5.24	−0.96	3.37	−7.04	−1.04
	−1.25	−0.87	−4.76	−1.57	−0.96	−6.83	−1.88	−1.05	−8.83

40	1.10	0.14	−0.06	2.17	0.06	−0.03	3.33	−0.02	0.00
	0.14	1.03	−0.66	0.06	3.39	−1.08	−0.02	5.76	−1.46
	−0.06	−0.66	0.00	−0.03	−1.08	1.74	0.00	−1.46	3.53
41	1.19	−0.08	0.05	2.94	−0.84	0.35	4.70	−1.52	0.62
	−0.08	0.79	−0.48	−0.84	2.71	−0.62	−1.52	4.65	−0.77
	0.05	−0.48	−0.26	0.35	−0.62	1.39	0.62	−0.76	3.08
42	−2.89	−2.34	0.94	−4.63	−2.82	1.14	−6.31	−3.30	1.34
	−2.34	−7.41	0.87	−2.82	−10.17	1.16	−3.30	−12.91	1.44
	0.94	0.87	−5.64	1.14	1.15	−7.77	1.35	1.44	−9.87
43	1.67	−0.34	0.16	4.32	−0.10	0.07	6.97	0.14	−0.01
	−0.34	1.02	−0.43	−0.10	2.24	−0.24	0.14	3.54	−0.08
	0.16	−0.43	0.17	0.07	−0.24	1.72	−0.01	−0.08	3.35
44	−4.36	2.27	−0.25	−6.83	3.20	−0.62	−9.12	4.08	−0.98
	2.27	−4.22	−0.15	3.20	−6.79	0.00	4.09	−9.23	0.15
	−0.26	−0.16	−5.63	−0.63	−0.01	−7.85	−0.99	0.14	−9.96

Table S19: Hyperfine coupling tensors of the radical (*R*)-**3c** in MHz.

nucleus	PBE			PBE0			PBE0-40		
1	0.28	−0.01	−0.08	0.70	0.00	−0.04	1.06	−0.01	−0.02
	−0.01	0.29	−0.10	0.00	0.64	−0.07	−0.01	0.93	−0.06
	−0.07	−0.11	0.06	−0.04	−0.07	0.39	−0.02	−0.07	0.68
2	0.02	0.03	0.00	0.03	0.03	0.01	0.04	0.03	0.01
	0.03	0.01	−0.04	0.03	0.02	−0.04	0.03	0.03	−0.04
	0.00	−0.04	0.01	0.01	−0.04	0.02	0.01	−0.04	0.03
3	0.01	0.02	0.02	0.03	0.02	0.02	0.05	0.02	0.02
	0.02	0.01	−0.02	0.02	0.03	−0.02	0.02	0.05	−0.02
	0.02	−0.02	0.00	0.02	−0.02	0.02	0.02	−0.02	0.04
4	−0.26	0.57	0.46	−0.16	0.33	0.39	−0.03	0.38	0.49
	0.57	−0.51	−0.30	0.33	−0.15	0.22	0.38	0.05	0.49
	0.47	−0.30	0.10	0.40	0.22	0.58	0.49	0.49	1.15
5	−25.17	−7.91	−5.83	−48.62	−19.49	−12.87	−66.86	−27.54	−17.86
	−7.91	−22.92	−4.86	−19.49	−43.01	−10.92	−27.53	−58.95	−15.23
	−5.85	−4.87	−17.35	−12.89	−10.93	−30.41	−17.88	−15.24	−41.67
6	70.03	12.99	17.19	99.57	17.41	20.15	120.58	20.24	21.66
	13.02	1.01	3.30	17.44	14.22	4.33	20.27	25.51	4.93
	17.19	3.30	3.58	20.15	4.33	16.23	21.67	4.93	26.98
7	6.61	24.11	4.43	21.81	31.17	5.40	34.37	35.47	5.92
	24.08	67.10	12.42	31.14	95.72	13.98	35.44	115.88	14.70
	4.43	12.42	1.25	5.40	13.98	13.68	5.93	14.70	24.41
8	0.66	−0.23	−0.33	0.14	−0.35	−0.92	−0.78	−0.47	−1.52
	−0.23	0.66	−0.43	−0.35	0.07	−1.13	−0.47	−0.94	−1.84
	−0.33	−0.43	−0.77	−0.91	−1.12	−3.65	−1.51	−1.84	−7.04
9	−0.03	−0.11	0.09	0.41	0.04	0.43	1.29	0.20	0.87
	−0.11	−0.17	0.11	0.04	0.25	0.51	0.19	1.11	1.05
	0.09	0.10	0.23	0.43	0.50	2.14	0.87	1.05	4.96

10	−2.35	0.04	0.22	−5.96	0.11	0.42	−8.99	0.09	0.33
	0.03	−2.83	−0.64	0.10	−6.42	−1.54	0.08	−9.48	−2.39
	0.22	−0.64	−4.48	0.42	−1.54	−10.59	0.33	−2.38	−16.20
11	20.38	−2.70	0.39	27.53	−2.69	1.03	33.24	−2.58	1.75
	−2.70	21.52	0.04	−2.69	28.77	0.74	−2.58	34.57	1.54
	0.37	0.03	19.76	1.02	0.73	29.12	1.73	1.52	37.33
12	−2.74	0.03	−0.52	−6.21	−0.01	−1.15	−9.18	−0.09	−1.78
	0.04	−2.37	0.09	0.00	−5.98	0.19	−0.08	−9.04	−0.01
	−0.51	0.09	−4.62	−1.15	0.19	−10.92	−1.78	−0.01	−16.65
13	−0.24	−0.07	0.09	0.16	0.08	0.41	0.99	0.25	0.84
	−0.07	0.01	0.09	0.08	0.46	0.52	0.25	1.36	1.06
	0.08	0.09	0.22	0.40	0.52	2.11	0.84	1.06	4.91
14	−16.35	−1.25	−1.07	−30.44	−2.73	−1.63	−42.10	−3.89	−2.10
	−1.26	−10.96	0.52	−2.73	−17.63	0.52	−3.91	−23.42	0.49
	−1.08	0.52	−10.37	−1.63	0.53	−17.48	−2.10	0.50	−23.64
15	11.41	3.69	0.83	17.87	4.77	1.31	23.97	5.73	1.76
	3.69	−1.43	0.29	4.77	0.39	0.40	5.73	2.27	0.51
	0.83	0.29	−1.83	1.31	0.41	−0.15	1.77	0.51	1.56
16	9.73	2.32	1.56	15.50	3.05	2.40	21.03	3.72	3.13
	2.32	−1.45	0.13	3.06	−0.17	0.25	3.73	1.27	0.37
	1.56	0.13	−2.02	2.40	0.25	−0.74	3.13	0.37	0.71
17	−15.29	−0.86	−0.76	−28.89	−1.63	−1.78	−40.22	−2.27	−2.58
	−0.85	−9.87	0.13	−1.62	−17.12	0.32	−2.26	−23.26	0.46
	−0.76	0.13	−10.64	−1.79	0.32	−17.48	−2.59	0.46	−23.27
18	−11.95	−2.06	0.30	−19.66	−4.81	0.46	−26.23	−7.00	0.55
	−2.06	−16.40	−0.82	−4.80	−30.10	−0.93	−6.99	−41.41	−1.04
	0.31	−0.82	−10.31	0.47	−0.93	−17.60	0.56	−1.05	−23.87
19	0.57	6.03	−0.63	3.00	8.02	−0.63	5.46	9.74	−0.65
	6.03	9.23	−1.23	8.02	15.72	−1.28	9.74	21.67	−1.35
	−0.62	−1.23	−1.83	−0.62	−1.27	−0.17	−0.64	−1.35	1.59

20	−0.95	3.39	−0.01	0.63	4.59	0.10	2.34	5.68	0.19
	3.38	10.05	0.49	4.58	15.93	0.95	5.67	21.55	1.29
	−0.01	0.49	−2.27	0.10	0.94	−1.03	0.18	1.29	0.41
21	−10.39	−1.70	0.26	−18.34	−3.43	0.44	−24.99	−4.83	0.57
	−1.72	−15.21	−0.30	−3.44	−29.03	−0.64	−4.84	−40.48	−0.86
	0.26	−0.30	−10.55	0.44	−0.64	−17.45	0.57	−0.86	−23.24
22	16.81	3.28	1.20	24.70	4.11	1.74	32.31	4.92	2.28
	3.28	−0.61	0.38	4.11	2.32	0.41	4.93	5.20	0.44
	1.20	0.38	−1.03	1.75	0.41	1.74	2.28	0.44	4.47
23	−7.57	−0.75	−0.31	−14.65	−1.68	−0.83	−21.61	−2.54	−1.31
	−0.75	−3.76	−0.12	−1.68	−6.02	−0.26	−2.54	−8.53	−0.39
	−0.31	−0.12	−3.16	−0.83	−0.25	−5.24	−1.31	−0.39	−7.56
24	17.31	3.49	2.00	24.13	4.39	2.42	31.05	5.30	2.86
	3.50	−1.19	0.47	4.39	1.31	0.59	5.30	3.79	0.70
	2.00	0.47	−1.19	2.42	0.58	1.37	2.87	0.70	3.88
25	−5.40	−0.43	−0.47	−12.32	−1.51	−1.00	−19.00	−2.47	−1.48
	−0.43	−3.45	0.18	−1.51	−5.42	0.14	−2.47	−7.69	0.11
	−0.47	0.18	−3.15	−1.00	0.14	−4.88	−1.48	0.11	−6.95
26	−4.63	−0.26	−0.21	−10.90	−1.03	−0.81	−17.06	−1.73	−1.37
	−0.26	−2.75	0.04	−1.03	−4.43	−0.04	−1.73	−6.38	−0.11
	−0.21	0.04	−3.18	−0.81	−0.04	−4.93	−1.37	−0.11	−6.95
27	15.46	2.29	2.65	22.04	3.01	3.07	28.62	3.71	3.51
	2.29	−0.89	0.25	3.01	1.42	0.26	3.71	3.77	0.29
	2.65	0.25	−1.17	3.06	0.26	0.90	3.51	0.29	3.04
28	−6.45	−0.68	−0.23	−13.01	−1.31	−0.71	−19.48	−1.90	−1.16
	−0.69	−2.66	−0.34	−1.32	−4.66	−0.47	−1.91	−6.87	−0.59
	−0.23	−0.34	−2.85	−0.70	−0.47	−4.82	−1.16	−0.59	−7.01
29	14.95	2.89	−0.89	22.63	3.53	−0.31	29.97	4.13	0.28
	2.90	−0.10	−0.28	3.53	2.36	−0.27	4.14	4.88	−0.23
	−0.88	−0.29	−0.92	−0.30	−0.27	1.66	0.28	−0.23	4.29

30	1.49	6.92	0.17	4.99	8.60	0.20	8.43	10.27	0.23
	6.93	15.47	0.03	8.60	22.86	0.23	10.27	30.09	0.44
	0.18	0.03	−1.19	0.21	0.23	1.60	0.23	0.44	4.31
31	−4.45	−1.52	−0.10	−7.27	−3.39	−0.20	−10.33	−5.12	−0.31
	−1.52	−7.28	0.09	−3.38	−13.82	−0.07	−5.12	−20.32	−0.21
	−0.10	0.09	−3.31	−0.20	−0.07	−5.33	−0.31	−0.21	−7.62
32	1.26	7.45	0.32	4.50	9.21	0.38	7.68	11.01	0.44
	7.45	15.63	0.55	9.20	21.70	0.65	11.01	28.08	0.74
	0.31	0.55	−1.45	0.38	0.65	1.16	0.44	0.75	3.70
33	−3.65	−0.50	−0.13	−6.91	−2.81	0.12	−10.18	−4.83	0.27
	−0.49	−5.30	−0.59	−2.81	−11.64	−0.33	−4.82	−17.89	−0.18
	−0.13	−0.59	−3.09	0.12	−0.34	−5.02	0.26	−0.19	−7.22
34	−2.91	−0.47	0.07	−4.81	−1.61	0.03	−6.97	−2.69	0.00
	−0.46	−4.64	−0.24	−1.61	−10.96	−0.55	−2.69	−17.19	−0.85
	0.07	−0.24	−3.22	0.03	−0.55	−4.97	0.00	−0.85	−6.99
35	−0.33	3.90	0.28	2.22	5.05	0.26	4.81	6.16	0.26
	3.90	15.37	1.50	5.04	22.10	1.56	6.16	28.73	1.65
	0.29	1.50	−1.53	0.26	1.56	0.53	0.26	1.65	2.67
36	−2.97	−1.16	−0.24	−5.21	−2.32	−0.33	−7.64	−3.40	−0.41
	−1.15	−6.47	0.07	−2.32	−13.05	−0.01	−3.40	−19.54	−0.11
	−0.24	0.08	−3.04	−0.33	−0.01	−5.04	−0.42	−0.11	−7.24
37	0.81	4.74	−0.84	3.63	6.06	−0.88	6.50	7.26	−0.89
	4.74	13.96	−2.38	6.05	21.83	−2.27	7.25	29.28	−2.12
	−0.84	−2.37	−0.72	−0.88	−2.27	1.89	−0.90	−2.11	4.56
38	−0.24	−0.03	−0.04	−0.39	−0.01	−0.03	−0.52	0.01	−0.02
	−0.03	−0.23	0.05	−0.01	−0.36	0.04	0.01	−0.48	0.03
	−0.04	0.05	−0.13	−0.03	0.04	−0.26	−0.02	0.03	−0.37
39	0.18	−0.02	−0.03	0.08	−0.02	−0.03	0.05	−0.02	−0.03
	−0.02	0.20	0.07	−0.02	0.09	0.05	−0.02	0.05	0.05
	−0.03	0.07	0.27	−0.03	0.05	0.17	−0.03	0.05	0.14

40	0.05	−0.05	−0.04	0.04	−0.05	−0.04	0.04	−0.05	−0.04
	−0.05	0.09	0.06	−0.05	0.07	0.06	−0.05	0.06	0.06
	−0.04	0.06	0.09	−0.04	0.06	0.08	−0.04	0.06	0.08
41	0.11	−0.11	−0.12	1.21	−0.08	−0.04	2.38	−0.06	0.02
	−0.10	−0.04	0.05	−0.08	0.97	0.10	−0.06	2.05	0.15
	−0.12	0.06	0.23	−0.04	0.10	1.28	0.02	0.15	2.40
42	0.00	−0.03	−0.02	0.00	−0.03	−0.03	−0.01	−0.03	−0.03
	−0.03	0.01	0.03	−0.03	0.01	0.03	−0.03	0.01	0.03
	−0.02	0.03	0.00	−0.03	0.03	0.00	−0.03	0.03	−0.01
43	0.55	−0.10	−0.06	1.60	0.31	−0.20	2.82	0.75	−0.35
	−0.10	0.58	−0.08	0.31	1.44	−0.15	0.75	2.46	−0.24
	−0.06	−0.08	0.31	−0.20	−0.15	1.27	−0.36	−0.24	2.36
44	0.67	−0.39	0.02	0.05	−0.32	0.02	−0.65	−0.21	−0.02
	−0.39	0.08	−0.01	−0.32	−0.89	0.04	−0.21	−2.15	0.12
	0.02	−0.01	0.12	0.02	0.04	−0.51	−0.02	0.12	−1.32
45	2.29	−1.34	0.55	3.49	−1.77	0.92	4.88	−2.14	1.18
	−1.34	−0.76	−0.15	−1.77	0.06	0.07	−2.14	1.05	0.26
	0.55	−0.15	−0.75	0.92	0.07	0.07	1.17	0.26	1.12
46	−1.17	−0.72	0.02	−0.55	−1.05	0.32	0.27	−1.30	0.58
	−0.72	2.82	0.21	−1.05	4.23	0.46	−1.31	5.78	0.60
	0.02	0.21	−0.88	0.32	0.46	−0.09	0.57	0.60	0.97
47	−0.02	−0.26	0.00	−0.95	−0.12	0.01	−2.15	0.09	0.03
	−0.26	0.83	−0.05	−0.12	0.18	−0.05	0.09	−0.60	−0.09
	0.00	−0.05	0.14	0.01	−0.05	−0.47	0.03	−0.09	−1.26
48	−5.54	−0.56	0.40	−7.86	−0.49	0.60	−10.11	−0.42	0.79
	−0.57	−3.51	−1.82	−0.50	−5.70	−2.65	−0.42	−7.83	−3.47
	0.40	−1.82	−5.40	0.60	−2.64	−8.27	0.79	−3.46	−10.99
49	0.10	−0.23	−0.26	1.76	−0.13	−0.26	3.50	−0.03	−0.25
	−0.23	1.03	0.25	−0.13	2.38	−0.27	−0.03	3.85	−0.79
	−0.26	0.25	1.72	−0.26	−0.27	4.21	−0.25	−0.79	6.74

50	−5.27	0.42	−0.66	−7.38	0.59	−0.76	−9.48	0.75	−0.86
	0.42	−8.32	1.21	0.58	−11.37	1.45	0.75	−14.42	1.69
	−0.67	1.21	−2.14	−0.76	1.45	−3.75	−0.86	1.69	−5.32
51	−0.35	−0.30	−0.21	1.28	−0.37	−0.43	2.99	−0.44	−0.63
	−0.30	0.85	−0.06	−0.37	2.46	0.69	−0.44	4.17	1.38
	−0.21	−0.06	1.13	−0.43	0.69	3.21	−0.63	1.38	5.33
52	−0.60	−0.17	−0.19	0.80	−0.34	−0.21	2.34	−0.51	−0.23
	−0.17	0.71	−0.20	−0.34	2.92	0.13	−0.51	5.21	0.46
	−0.19	−0.20	0.79	−0.21	0.13	1.84	−0.23	0.46	3.02
53	−4.74	−0.18	0.52	−6.76	−0.22	0.66	−8.74	−0.26	0.81
	−0.18	−1.68	−1.19	−0.22	−3.23	−1.53	−0.26	−4.74	−1.86
	0.52	−1.19	−7.28	0.66	−1.53	−10.22	0.81	−1.86	−13.08
54	0.00	−0.16	−0.15	1.44	−0.12	−0.06	3.00	−0.08	0.02
	−0.16	1.98	−0.19	−0.12	3.81	−0.90	−0.08	5.69	−1.58
	−0.15	−0.19	0.77	−0.06	−0.89	2.42	0.02	−1.58	4.20
55	−5.95	−0.10	0.86	−8.64	0.14	0.85	−11.03	0.38	0.82
	−0.11	−3.70	−1.47	0.14	−6.60	−2.07	0.38	−9.47	−2.60
	0.87	−1.47	−2.60	0.86	−2.08	−4.47	0.83	−2.60	−6.14
56	−3.76	−1.16	−1.56	−5.86	−1.20	−2.26	−7.90	−1.25	−2.98
	−1.16	−5.28	0.78	−1.20	−7.61	1.06	−1.24	−9.88	1.36
	−1.56	0.77	−5.98	−2.25	1.06	−9.04	−2.97	1.36	−11.98
57	0.86	−0.39	0.20	2.37	−0.29	−0.39	3.98	−0.21	−0.96
	−0.39	0.15	−0.17	−0.29	1.83	0.03	−0.21	3.58	0.24
	0.20	−0.17	1.81	−0.39	0.03	4.23	−0.97	0.24	6.68
58	−8.30	1.19	0.47	−11.25	1.57	0.55	−14.23	1.95	0.62
	1.19	−5.95	−0.41	1.57	−8.22	−0.44	1.96	−10.50	−0.46
	0.47	−0.41	−1.87	0.55	−0.44	−3.48	0.62	−0.47	−5.03
59	0.70	−0.35	−0.16	2.81	−0.55	0.32	5.02	−0.75	0.81
	−0.35	−0.55	0.03	−0.55	0.90	−0.06	−0.75	2.49	−0.15
	−0.16	0.03	0.83	0.32	−0.06	1.98	0.81	−0.15	3.29

60	−1.66	−0.65	−0.50	−3.21	−0.79	−0.66	−4.72	−0.92	−0.84
	−0.65	−4.68	0.32	−0.79	−6.73	0.39	−0.92	−8.72	0.46
	−0.49	0.33	−7.71	−0.66	0.39	−10.82	−0.84	0.46	−13.84
61	2.03	−0.43	−0.04	4.04	−0.53	−0.71	6.10	−0.62	−1.36
	−0.43	0.13	−0.10	−0.53	1.63	0.08	−0.62	3.26	0.25
	−0.04	−0.10	0.86	−0.71	0.08	2.37	−1.36	0.25	4.01
62	−3.70	−0.75	−1.42	−6.55	−0.55	−2.17	−9.31	−0.32	−2.85
	−0.74	−5.74	1.35	−0.54	−8.59	1.54	−0.31	−11.10	1.69
	−1.42	1.36	−3.23	−2.17	1.55	−5.34	−2.85	1.70	−7.21
63	0.03	−0.16	−0.25	−0.01	−0.14	−0.25	−0.03	−0.13	−0.24
	−0.16	−0.18	0.13	−0.14	−0.22	0.13	−0.13	−0.24	0.13
	−0.25	0.13	0.18	−0.25	0.13	0.17	−0.24	0.13	0.17
64	−0.46	−0.34	−0.32	−0.51	−0.35	−0.38	−0.55	−0.34	−0.44
	−0.34	−0.05	0.68	−0.34	−0.15	0.83	−0.34	−0.22	0.95
	−0.32	0.68	0.29	−0.39	0.83	0.29	−0.44	0.95	0.31
65	−0.14	−0.07	−0.08	−0.14	−0.07	−0.09	−0.14	−0.08	−0.09
	−0.07	−0.03	0.19	−0.07	−0.04	0.18	−0.08	−0.05	0.18
	−0.08	0.19	0.12	−0.09	0.18	0.13	−0.09	0.18	0.12
66	0.01	−0.08	−0.11	−0.02	−0.07	−0.11	−0.02	−0.07	−0.10
	−0.08	0.00	0.12	−0.07	−0.03	0.11	−0.07	−0.04	0.11
	−0.11	0.12	0.15	−0.11	0.11	0.13	−0.11	0.11	0.13
67	−0.10	−0.08	−0.14	−0.08	−0.08	−0.14	−0.07	−0.08	−0.14
	−0.08	−0.11	0.13	−0.08	−0.13	0.11	−0.08	−0.13	0.10
	−0.14	0.13	0.20	−0.14	0.11	0.19	−0.14	0.10	0.18
68	−0.01	−0.18	−0.11	−0.01	−0.18	−0.11	−0.01	−0.18	−0.11
	−0.18	0.08	0.13	−0.18	0.08	0.14	−0.18	0.08	0.14
	−0.11	0.13	−0.07	−0.11	0.14	−0.07	−0.11	0.14	−0.07
69	0.00	−0.10	−0.08	0.00	−0.10	−0.08	0.00	−0.10	−0.08
	−0.10	0.02	0.08	−0.10	0.02	0.09	−0.10	0.02	0.09
	−0.08	0.08	−0.02	−0.08	0.09	−0.02	−0.08	0.09	−0.02

70	−0.05	−0.11	−0.07	−0.05	−0.11	−0.08	−0.05	−0.11	−0.08
	−0.11	0.07	0.12	−0.11	0.07	0.12	−0.11	0.07	0.13
	−0.07	0.12	−0.02	−0.08	0.12	−0.02	−0.08	0.13	−0.02

Table S20: Permanent electric dipole moments of radicals **1**-**3**.

radical	$(\mu_e)_x$ / C·m	$(\mu_e)_x$ / C·m	$(\mu_e)_x$ / C·m	μ_e / D
1a	−0.06622	−98.76426	−21.65321	3.0312
syn-(*R*)-**1b**	−127.52995	−28.90983	−95.04480	4.8464
anti-(*R*)-**1b**	−124.70403	54.47173	−84.04770	4.7950
2a	−37.44456	−0.28004	1.03410	1.1230
(*R*)-**2b**	−24.29142	−26.37531	−65.30621	2.2335
3a	0.00000	0.00000	0.00644	0.0002
3b	−0.01094	1.25090	−0.00170	0.0375
(*R*)-**3c**	−4.77187	−72.17546	53.10730	2.6902

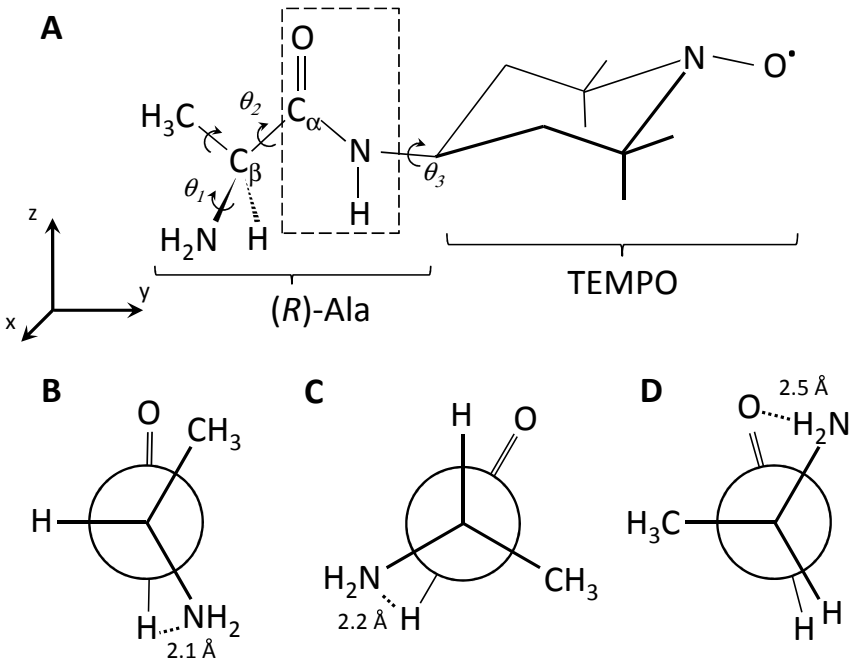

Figure S1: The structure of (*R*)-**1b** with marked degrees of freedom of internal rotation (A) and the Newman projection of its three lowest-energy conformers **1b-I** (*i.e.*, *syn*-**1b**) (B), **1b-II** (*i.e.*, *anti*-**1b**) (C), and **1b-III** (D) shown along the axis from the C_β to C_α carbon atom. The horizontal plane of the projections is approximately in the plane of the cyclohexane ring.

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