Supporting Information

Electronic Structure of Fullerene Acceptors in Organic Bulk-Heterojunctions: A Combined EPR and DFT Study

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All geometry optimizations were carried out using density functional theory (DFT) and the B3LYP functional[1-4] using the 3-21G, 6-31G, and the 6-31G+(d) basis set, as implemented in PQSMol.[5] The spectroscopic parameters were obtained via single point DFT calculations, using functionals and basis sets as implemented in the program package ORCA (v 2.9.1).[6] For details, see *Density Functional Theory (DFT) Calculations* part in the main text.

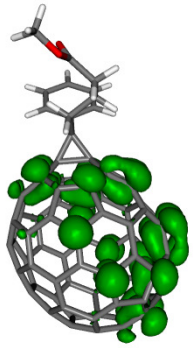
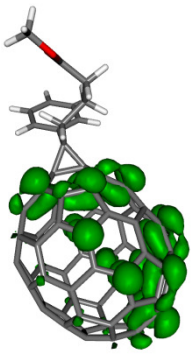
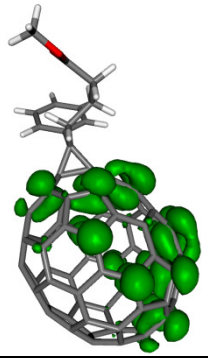
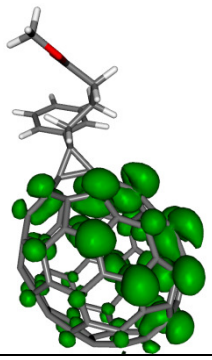
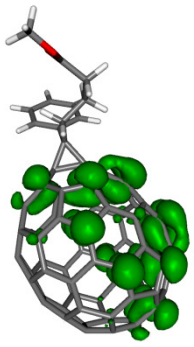
Table S1. Comparison of calculated EPR g-values for fullerene radical anions geometry optimized using different basis sets. All geometry optimizations were performed using the B3LYP functional and all EPR parameter calculations used the EPRII basis set[7-8] and the B3LYP functional.

Molecule	3-21G	6-31G	6-31G+(d)	Experimental
$PC_{61}BM$	1.9996, 2.0009, 2.0009	1.9994, 2.0008, 2.0009	1.9995, 2.0008, 2.0009	1.9985, 2.0005, 2.0006
α-$PC_{71}BM$	2.0027, 2.0036, 2.0054	2.0028, 2.0036, 2.0054	2.0026, 2.0035, 2.0054	2.0021, 2.0028, 2.0060
β_1-$PC_{71}BM$	2.0028, 2.0031, 2.0055	2.0027, 2.0030, 2.0056	2.0019, 2.0029, 2.0048	
β_2-$PC_{71}BM$	2.0027, 2.0030, 2.0054	2.0026, 2.0029, 2.0055	2.0017, 2.0029, 2.0047	

Table S2. Comparison of calculated EPR g-values for fullerene radical anions using the EPRII and def2-TZVPP basis sets[9]. All geometry optimizations were performed using the B3LYP functional and the 6-31G+(d) basis set.

Molecule	EPRII	def2-TZVPP	Experimental
$PC_{61}BM$	1.9995, 2.0008, 2.0009	1.9994, 2.0008, 2.0011	1.9985, 2.0005, 2.0006
α-$PC_{71}BM$	2.0026, 2.0035, 2.0054	2.0027, 2.0034, 2.0052	2.0021, 2.0028, 2.0060
β_1-$PC_{71}BM$	2.0019, 2.0029, 2.0048	2.0020, 2.0029, 2.0045	
β_2-$PC_{71}BM$	2.0017, 2.0029, 2.0047	2.0018, 2.0029, 2.0045	

Table S3. Comparison of calculated EPR g-values and spin density plots for α-$PC_{71}BM$ radical anion using a variety of functionals. All the geometry optimizations used the B3LYP functional and the 6-31G+(d) basis set and all the EPR parameters were calculated using the EPRII basis set. The experimental values are 2.0021, 2.0028, and 2.0060. The spin density contours are shown at the level of 0.001 e/a_0^3.

	B3LYP	BP86	B3PW91	BHandHLYP	PBE0
g_1	2.0026	2.0024	2.0026	2.0026	2.0026
g_2	2.0035	2.0034	2.0034	2.0033	2.0034
g_3	2.0054	2.0055	2.0053	2.0051	2.0053
Spin Density Plot					

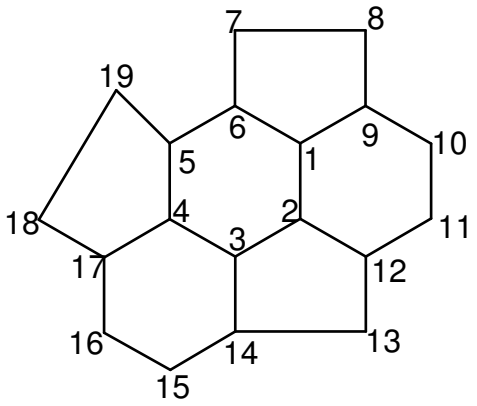


Figure S1. Numbering scheme used for $PC_{61}BM$ and α-$PC_{71}BM$ in Tables S4-S5. Bond 1-2 is where the PCBM side chain is bonded.[10]

Table S4. Selected bond lengths (Å) for $PC_{61}BM$ radical anion calculated using the B3LYP functional and a variety of basis sets. The bond numbering corresponds to that shown in Figure S1.

Basis set/ C-C bond	**3-21G (Å)**	**6-31G (Å)**	**6-31G+(d) (Å)**
1-2	1.6146	1.6184	1.6075
2-3	1.4986	1.5015	1.4989
3-4	1.3849	1.3934	1.3913
4-5	1.4767	1.4763	1.4710
5-6	1.3845	1.3931	1.3907
6-1	1.4952	1.4988	1.4970
6-7	1.4456	1.4459	1.4407
7-8	1.4513	1.4510	1.4458
8-9	1.4495	1.4490	1.4433
9-1	1.4954	1.4986	1.4960
9-10	1.3812	1.3895	1.3874
10-11	1.4788	1.4780	1.4725
11-12	1.3815	1.3898	1.3874
12-2	1.4965	1.4996	1.4969
12-13	1.4485	1.4482	1.4426
13-14	1.4514	1.4511	1.4458
14-3	1.4464	1.4465	1.4413
14-15	1.4038	1.4101	1.4095
15-16	1.4434	1.4437	1.4385
16-17	1.3973	1.4041	1.4030
17-4	1.4620	1.4617	1.4563
17-18	1.4513	1.4516	1.4466
18-19	1.4506	1.4512	1.4461
19-5	1.4621	1.4614	1.4559

Table S5. Selected bond lengths (Å) for α-$PC_{71}BM$ radical anion calculated using the B3LYP functional and a variety of basis sets. The bond numbering corresponds to that shown in Figure S1.

Basis set/ C-C bond	3-21G (Å)	6-31G (Å)	6-31G+(d) (Å)
1-2	1.6201	1.6253	1.6147
2-3	1.4959	1.4984	1.4961
3-4	1.3874	1.3954	1.3959
4-5	1.4668	1.4654	1.4582
5-6	1.3905	1.3973	1.3945
6-1	1.4915	1.4939	1.4913
6-7	1.4279	1.4274	1.4234
7-8	1.4625	1.4610	1.4551
8-9	1.4259	1.4275	1.4238
9-1	1.4930	1.4957	1.4934
9-10	1.3931	1.4003	1.3974
10-11	1.4679	1.4668	1.4593
11-12	1.3880	1.3964	1.3971
12-2	1.4950	1.4982	1.4965
12-13	1.4426	1.4427	1.4350
13-14	1.4567	1.4558	1.4519
14-3	1.4436	1.4434	1.4352
14-15	1.3989	1.4055	1.4063
15-16	1.4412	1.4406	1.4350
16-17	1.3907	1.3969	1.3949
17-4	1.4573	1.4567	1.4502
17-18	1.4465	1.4466	1.4446
18-19	1.4469	1.4481	1.4409
19-5	1.4463	1.4465	1.4442

Table S6. Atomic number, type, and coordinates of $PC_{61}BM$ radical anion structure optimized using B3LYP|6-31G+(d); Mulliken spin populations, and hyperfine coupling constants (B3LYP|EPRII). Mulliken spin populations are unitless and sum to 1. The principal axes of the hyperfine coupling tensors for ^{13}C and ^{1}H are aligned with the principal axes of the g-tensor.

Atom Number	Atom Type	X coord (Å)	Y coord (Å)	Z coord (Å)	Spin Population	A_1 (MHz)	A_2 (MHz)	A_3 (MHz)
0	C	1.509846	-1.123261	-1.49699	0.015644	-0.12	-0.19	3.98
1	C	1.1062	-2.413732	-2.032793	-0.000384	-3.69	-3.29	-0.65
2	C	0.83441	-3.482678	-1.164232	0.010323	-2.38	1.89	-2.2
3	C	0.27546	-2.193508	-3.195957	0.066546	-1.25	14.59	-1.46
4	C	-0.817371	-3.050386	-3.455066	0.069285	16	-0.98	-0.78
5	C	0.134285	-0.76585	-3.378455	0.000553	-0.53	-3.67	-3.28
6	C	-1.085823	-0.229333	-3.81983	0.010834	1.89	-2.25	-2.43
7	C	0.898982	-0.085438	-2.344357	0.015278	3.94	-0.19	-0.12
8	C	0.42549	1.097534	-1.795473	-0.010754	-3.92	-1.54	-1.77
9	C	1.63198	-0.948101	-0.126167	-0.01125	-1.76	-1.54	-3.9
10	C	1.301563	-2.032901	0.765526	0.009254	-2.2	-2.74	0.96
11	C	0.625387	-1.490894	1.922845	0.022389	-2.08	-2.46	3.7
12	C	0.934727	-3.292384	0.257305	0.057128	1.05	1.32	14.73
13	C	-0.144091	-4.043179	0.887885	-0.011368	-3.48	-5.24	-2.86
14	C	-0.300294	-4.362814	-1.424585	0.00778	-2.56	1.23	-2.4
15	C	-0.897665	-4.718454	-0.153936	0.059706	0.41	14.51	0.2
16	C	-1.106803	-4.141974	-2.547962	-0.001132	-0.91	-4	-3.45
17	C	-2.553855	-4.270582	-2.431883	0.020519	4.87	-0.4	-0.28
18	C	-2.080227	-2.493108	-3.893677	-0.000765	-3.46	-4.02	-0.93
19	C	-3.157473	-3.248123	-3.266229	0.021011	-0.39	-0.27	4.9
20	C	-2.219383	-1.111786	-4.077766	0.00784	-2.4	1.23	-2.56
21	C	-3.426347	-0.435081	-3.649707	0.059508	0.16	0.38	14.5
22	C	-1.588655	0.982718	-3.232116	0.057834	14.91	1.33	1.06
23	C	-3.036104	0.855503	-3.110149	-0.011477	-2.87	-3.49	-5.26
24	C	-0.853423	1.618518	-2.214976	0.008718	1.01	-2.23	-2.77
25	C	-1.532733	2.162426	-1.060379	0.024035	3.9	-2.06	-2.44
26	C	0.64117	1.472575	-0.363359	0.001224	-0.69	-0.82	-0.63
27	C	-0.688944	1.985187	0.093848	-0.010922	-4.07	-2.12	-2.51
28	C	1.314891	0.343313	0.561198	0.001859	-0.66	-0.85	-0.76
29	C	0.524533	-0.061435	1.768767	-0.010083	-2.49	-2.11	-3.96
30	C	-4.09488	-0.446099	2.107335	0.065021	-1.28	14.69	-1.52
31	C	-4.932285	-0.226319	0.946842	-0.00041	-3.42	-4.06	-0.88
32	C	-4.660945	0.828924	0.065381	0.019042	-1.79	4.07	-1.49
33	C	-5.325621	-1.523562	0.413912	0.011589	-0.34	-0.33	2.63
34	C	-5.426005	-1.714799	-0.967442	-0.009955	-1.61	-1.42	-3.41
35	C	-4.721254	-2.547239	1.249393	0.011506	2.6	-0.33	-0.34
36	C	-4.238175	-3.726553	0.674459	-0.009906	-3.39	-1.4	-1.59
37	C	-3.958566	-1.875527	2.292762	-0.000423	-0.89	-4.06	-3.42
38	C	-2.740906	-2.422615	2.719189	0.019401	-1.47	4.1	-1.78
39	C	-2.987144	0.399787	2.34262	0.075676	17.47	-0.83	-0.63
40	C	-2.699375	1.484552	1.430601	-0.000918	-1.02	-4.1	-3.72
41	C	-1.255179	1.625086	1.31194	0.027577	6.7	-0.39	-0.27
42	C	-3.521505	1.706036	0.315137	0.001353	-3.5	-0.35	-3.19

43	C	-2.933988	2.053465	-0.951292	0.070852	1.69	18.33	1.51
44	C	-4.770232	0.632669	-1.364647	0.049385	-0.11	0.19	12.03
45	C	-3.691376	1.379083	-1.994556	-0.012919	-3.66	-5.65	-3.03
46	C	-5.145567	-0.62011	-1.88148	0.017516	-2.25	-2.71	2.85
47	C	-4.466556	-1.1622	-3.038699	0.028593	-2.01	-2.36	5.52
48	C	-4.920206	-2.940185	-1.566184	-0.000907	0.22	0.05	-0.37
49	C	-4.320773	-2.593259	-2.845921	-0.010135	-2.25	-1.89	-3.91
50	C	-4.339222	-3.924052	-0.763151	-0.000895	-0.41	0.04	0.21
51	C	-3.131793	-4.607096	-1.202385	-0.010287	-3.87	-2.23	-1.87
52	C	-2.975907	-4.29457	1.117582	0.017274	2.85	-2.23	-2.69
53	C	-2.296185	-4.838356	-0.038537	0.02855	5.42	-2	-2.35
54	C	-2.243341	-3.646528	2.127881	0.048865	11.92	0.18	-0.1
55	C	-0.799029	-3.51903	2.003189	-0.012754	-3.01	-3.62	-5.57
56	C	-1.60094	-1.545947	2.969221	0.00166	-3.14	-0.31	-3.43
57	C	-0.407398	-2.22482	2.540454	0.069616	1.44	1.61	18.02
58	C	-1.726125	-0.162166	2.774567	-0.001356	-3.73	-4.13	-1.13
59	C	-0.641969	0.589443	2.157727	0.02672	-0.43	-0.31	6.6
60	C	1.981757	1.686532	0.323007	-0.000374	1.92	1.39	0.95
61	C	1.999247	2.689572	1.487118	0.000547	0.98	0.66	0.68
64	C	3.102403	2.47241	2.550055	0.000085	0.04	0.05	0.17
67	C	4.398174	3.281037	2.292688	-0.000011	0.09	0.09	0.17
70	C	5.397919	3.096827	3.42125	0.000029	0.01	0.01	0.07
73	C	4.485932	5.082004	4.52735	0.000003	-0.02	-0.01	0.03
78	C	3.213599	1.749788	-0.550981	0.000607	0.47	0.5	0.65
79	C	3.323759	2.758558	-1.520387	0.000419	0.15	0.03	-0.03
80	C	4.485881	2.897107	-2.281761	-0.000032	0.05	-0.03	-0.07
81	C	5.560579	2.023755	-2.085649	0.000217	0.02	-0.02	0.12
82	C	5.459887	1.012667	-1.126888	0.000102	-0.06	-0.02	0.07
83	C	4.2938	0.876187	-0.368046	0.000047	-0.01	-0.09	0.11
62	H	1.030086	2.658274	1.988955	-0.000084	0.76	-0.21	-0.35
63	H	2.085592	3.699	1.060402	0.00012	0.47	-0.05	0.01
65	H	3.353852	1.408421	2.627186	-0.000017	-0.19	-0.11	0.35
66	H	2.694933	2.755306	3.529426	-0.000013	-0.2	-0.18	0.27
68	H	4.146205	4.336632	2.144749	0.000005	-0.1	-0.09	0.18
69	H	4.886523	2.925688	1.38331	-0.000014	-0.09	-0.08	0.2
74	H	4.624639	5.519253	5.518107	0	-0.05	-0.05	0.1
75	H	3.439054	4.785292	4.409474	0	-0.08	-0.08	0.16
76	H	4.751502	5.824837	3.766608	0	-0.06	-0.06	0.11
77	H	6.287265	0.325902	-0.966643	0.000007	-0.11	-0.09	0.22
84	H	4.224388	0.082574	0.370184	-0.00003	-0.25	-0.17	0.27
85	H	2.485522	3.431445	-1.686358	-0.000074	-0.24	-0.17	0.27
86	H	4.548018	3.680418	-3.033727	0.000011	0.21	-0.08	-0.1
87	H	6.465411	2.126597	-2.679955	-0.000021	-0.1	-0.12	0.15
71	O	6.207054	2.196502	3.448205	0.000008			
72	O	5.369732	3.956631	4.485097	0.000002			

Table S7. Atomic number, type, coordinates of α-$PC_{71}BM$ radical anion structure optimized using B3LYP|6-31G+(d); Mulliken spin populations, and hyperfine coupling constants B3LYP|EPRII). Mulliken spin populations are unitless and sum to 1. The principal axes of the hyperfine coupling tensors for ^{13}C and ^{1}H are aligned with the principal axes of the g-tensor.

Atom Number	Atom Type	X coord (Å)	Y coord (Å)	Z coord (Å)	Spin Population	A_1 (MHz)	A_2 (MHz)	A_3 (MHz)
0	C	-0.830036	-2.383096	-0.651209	0.051068	-0.17	0.09	12.38
1	C	-0.366285	-1.457972	-1.731694	-0.008945	-3.35	-2.99	-4.97
2	C	-1.469914	-0.457511	-1.875021	0.050723	12.19	-0.03	-0.3
3	C	-2.60334	-0.923575	-1.128491	0.013214	2.87	-2.21	-2.19
4	C	-2.211101	-2.103379	-0.37864	0.010942	2.49	-2.21	-2.25
5	C	1.107661	-0.835016	-1.515392	-0.004683	-1.29	-0.6	-0.52
6	C	1.18978	0.655966	-1.496617	0.022051	2.99	-4.32	-4.5
7	C	0.100433	1.480343	-1.790703	0.068895	16.69	0.21	-0.11
8	C	-1.234169	0.9166	-1.965499	0.003875	-0.59	-4.25	-3.59
9	C	-2.167989	1.851903	-1.368639	-0.013241	-2.85	-5.25	-3.55
10	C	-3.30145	1.390933	-0.698688	0.009943	0.12	2.83	0.29
11	C	-3.52845	-0.020584	-0.57408	0.002963	0.13	-1.07	-0.89
12	C	-4.067433	-0.253588	0.750377	-0.00273	-0.62	-0.22	-0.38
13	C	-3.692164	-1.38384	1.467932	-0.002886	-0.2	-0.32	-0.59
14	C	-2.761294	-2.334823	0.894756	0.00207	-0.87	-1.04	-0.08
15	C	-1.914034	-2.794711	1.957791	0.009383	0.31	2.68	0.15
16	C	-0.557191	-3.009391	1.715451	-0.012251	-3.4	-2.76	-5.06
17	C	-0.001924	-2.800611	0.392081	0.003712	-4.42	-3.72	-0.98
18	C	1.335265	-2.249566	0.5779	0.070979	0.38	0.7	17.87
19	C	1.819291	-1.258108	-0.274959	0.012441	-4.81	-4.95	0.68
20	C	0.421373	-2.623752	2.705641	0.086187	3.21	2.98	23.46
21	C	1.575167	-2.13086	1.997051	-0.022914	-5.49	-4.9	-10.26
22	C	2.120654	1.047308	-0.492872	0.066502	-1.72	15.37	-2
23	C	2.51122	-0.135109	0.25987	0.074863	-1.33	17.67	-1.08
24	C	-2.326528	-2.147701	3.192012	-0.004437	-0.48	-1.38	-0.37
25	C	-3.430249	-1.28089	2.890168	0.004364	0.26	1.34	0.21
26	C	-4.193764	1.02175	1.428524	0.005151	0.26	1.59	0.2
27	C	-3.714279	2.037912	0.535364	-0.004159	-0.41	-1.38	-0.53
28	C	-1.437839	2.986974	-0.855878	0.088178	2.96	23.93	3.18
29	C	-0.038259	2.738439	-1.094134	-0.020925	-5.41	-9.89	-4.76
30	C	0.9526	3.210905	-0.185609	0.006438	1.83	2.96	1.57
31	C	2.074321	2.326317	0.134572	-0.035769	-4.47	-10.77	-5.37
32	C	2.859513	-0.04133	1.638132	-0.039119	-11.94	-5.96	-5.03
33	C	2.376761	-1.086383	2.542455	0.00849	2.02	1.79	3.45
34	C	0.039854	-2.072643	3.960872	-0.037938	-5.56	-6.21	-12.15
35	C	-1.390314	-1.824027	4.211947	0.003258	0.98	1.33	0.94
36	C	-4.022442	1.136599	2.837161	-0.004433	-0.96	-0.32	-0.28
37	C	-3.625399	-0.060671	3.597034	-0.004288	-0.24	-0.91	-0.28
38	C	-1.871902	3.695883	0.299608	-0.03825	-5.62	-6.28	-12.23
39	C	-3.055564	3.199882	1.023123	0.003227	0.99	0.95	1.34
40	C	0.5245	4.149733	0.796094	-0.025524	-4.72	-10.5	-5.56
41	C	-0.876344	4.399765	1.03032	0.088933	3.17	2.93	23.95
42	C	3.101507	1.260308	2.154548	0.076945	-0.49	18.69	-0.77

43	C	2.706963	2.446313	1.401258	0.066863	-1.5	15.59	-1.26
44	C	2.148823	-0.752164	3.907702	-0.027503	-5.67	-11.13	-4.88
45	C	0.996449	-1.249481	4.616127	0.089322	3.01	24.17	3.25
46	C	-3.142109	3.427697	2.425142	-0.003971	-0.38	-0.25	-1.15
47	C	-3.61941	2.411268	3.318975	0.004819	1.44	0.12	0.19
48	C	-2.85616	0.110001	4.779683	0.004456	0.2	1.41	0.13
49	C	-1.754216	-0.75826	5.082025	-0.003287	-1	-0.18	-0.33
50	C	-0.753692	0.018494	5.792982	0.005375	-0.08	1.57	0.1
51	C	0.596231	-0.221365	5.553072	-0.011314	-4.77	-3.39	-2.61
52	C	2.479453	0.555379	4.428583	0.074645	0.44	18.54	0.07
53	C	2.938908	1.548547	3.547636	0.021539	2.3	-4.89	-5.22
54	C	2.307525	3.452439	2.338791	0.026018	-4.77	-4.43	3.54
55	C	1.234603	4.310517	2.044045	0.072702	-0.1	17.89	0.24
56	C	-1.028706	4.679788	2.441855	-0.010418	-2.56	-3.39	-4.77
57	C	-2.144442	4.21264	3.131039	0.006791	0.14	-0.04	1.9
58	C	-2.533751	1.429574	5.294778	-0.00191	-0.63	-0.22	-0.46
59	C	-2.908887	2.560213	4.577442	-0.00132	-0.42	-0.09	-0.53
60	C	0.279976	4.64377	3.070971	0.000498	-4.1	-4.75	-1.49
61	C	-2.005281	3.686352	4.469758	0.004318	-0.95	-1.13	0.49
62	C	-1.237854	1.372711	5.938108	0.005415	0.81	-0.79	-0.97
63	C	1.524216	0.891586	5.453369	0.001016	-1.35	-4.85	-4.22
64	C	2.45205	2.911565	3.679453	-0.002781	-0.34	-1.51	-0.89
65	C	1.054991	2.210232	5.589739	0.062152	15.82	0.6	0.99
66	C	1.529742	3.237636	4.674944	-0.025298	-4.22	-3.72	-9.34
67	C	0.418495	4.128075	4.371468	0.058573	0.86	0.48	14.82
68	C	-0.348914	2.454296	5.831662	0.011374	2.12	-2.67	-2.62
69	C	-0.741556	3.638331	5.080405	0.014182	-2.39	-2.36	2.95
72	C	0.763433	-1.706678	-2.711068	-0.00383	-1.63	-1	-1.06
73	C	1.335595	-3.105047	-2.787237	-0.000071	-0.01	-0.21	-0.3
74	C	0.503161	-4.175928	-3.144192	-0.000239	0.05	-0.01	0.2
75	C	1.019336	-5.463731	-3.299968	0.000012	-0.06	-0.04	0.07
76	C	2.382348	-5.701762	-3.099817	-0.000048	-0.02	-0.01	0.1
77	C	3.220751	-4.642544	-2.743861	0.000072	-0.01	-0.03	0.16
78	C	2.699948	-3.354749	-2.589326	-0.000024	-0.04	0.04	0.19
83	C	0.666484	-1.022784	-4.083335	-0.00044	-0.27	-0.37	-0.07
86	C	2.029923	-0.71561	-4.722366	0.000275	0.17	0	-0.04
89	C	1.911751	0.158134	-5.979746	-0.000077	0.12	-0.02	-0.02
92	C	1.248311	-0.535441	-7.15276	0.000029	0.09	0	-0.01
95	C	-0.030785	-0.19931	-9.118664	0.000002	0.03	-0.02	-0.02
70	H	4.281509	-4.816587	-2.578816	0.000008	-0.15	-0.14	0.33
71	H	0.085249	-1.67837	-4.744931	-0.000027	0.38	-0.29	-0.34
79	H	3.357056	-2.539729	-2.29906	-0.000029	-0.54	-0.39	0.67
80	H	-0.559684	-3.99863	-3.290663	-0.000021	-0.25	-0.21	0.65
81	H	0.355436	-6.28137	-3.570833	-0.000008	-0.16	-0.14	0.24
82	H	2.785751	-6.705127	-3.214602	0.000011	-0.08	-0.08	0.24
84	H	0.101809	-0.090674	-3.987202	-0.000141	0.8	-0.2	-0.47
85	H	2.915594	0.447883	-6.324114	0.000001	0.29	-0.14	-0.15
87	H	2.65236	-0.174893	-3.998389	-0.000058	0.81	-0.26	-0.37
88	H	2.552576	-1.64473	-4.972814	0.000033	-0.13	-0.11	0.44
91	H	1.377979	1.090996	-5.768413	0.000011	0.44	-0.19	-0.22

94	H	-0.782187	-0.941597	-8.835472	0	0.14	-0.07	-0.07
96	H	-0.49821	0.648427	-9.621357	0	0.12	-0.06	-0.06
97	H	0.713061	-0.671234	-9.767216	0	0.11	-0.06	-0.06
90	O	0.597306	0.350509	-7.948269	0			
93	O	1.300449	-1.723958	-7.399182	-0.000004			

Table S8. Atomic number, type, coordinates of β_1-$PC_{71}BM$ radical anion structure optimized using B3LYP|6-31G+(d); Mulliken spin populations, and hyperfine coupling constants B3LYP|EPRII). Mulliken spin populations are unitless and sum to 1. The principal axes of the hyperfine coupling tensors for ^{13}C and ^{1}H are aligned with the principal axes of the g-tensor.

Atom Number	Atom Type	Xcoord (Å)	Ycoord (Å)	Zcoord (Å)	Spin Population	A_1 (MHz)	A_2 (MHz)	A_3 (MHz)
0	C	-1.620488	-3.340284	-4.218477	-0.025424	-10.27	-5.69	-4.97
1	C	-0.786577	-2.236768	-4.675141	0.081173	20.5	0.68	0.29
2	C	-1.656564	-1.125328	-4.964094	0.027088	5.72	-3.09	-3.19
3	C	-3.032112	-1.545079	-4.705518	0.010351	1.27	-3.99	-4.15
4	C	-3.010428	-2.918138	-4.260671	0.089896	0.97	1.3	23.2
5	C	0.458518	-2.007947	-4.053034	0.013593	0.79	-4.56	-3.92
6	C	0.857444	-0.658187	-3.707165	-0.008246	-3.6	-2.61	-2.08
7	C	0.019191	0.411399	-3.99318	-0.008174	-2.89	-1.45	-1.06
8	C	-1.258341	0.182095	-4.63633	0.010799	1.73	-1.77	-1.35
9	C	-2.198866	1.102378	-4.043612	0.016309	3.8	-0.66	-0.71
10	C	-3.512381	0.702549	-3.797447	0.016277	3.84	-0.46	-0.52
11	C	-3.937479	-0.637563	-4.136306	0.005565	-0.85	-1.25	0.86
12	C	-4.840771	-1.072712	-3.089624	-0.011161	-1.03	-1.37	-3.54
13	C	-4.819044	-2.393174	-2.653163	-0.003905	-2.09	-1.17	-2.51
14	C	-3.892197	-3.336881	-3.241388	0.016102	-4.7	-4.09	1.26
15	C	-3.427676	-4.188848	-2.177513	0.04708	-0.55	-0.2	11.55
16	C	-2.089758	-4.595638	-2.140874	0.011022	-3.58	-3.44	0.72
17	C	-1.174589	-4.167422	-3.183762	-0.001887	-0.58	-0.94	-0.18
18	C	0.109878	-3.927423	-2.55467	0.020595	3.18	-2.98	-3.27
19	C	0.911236	-2.864505	-2.990235	0.051509	12.68	-0.25	-0.57
20	C	-1.374176	-4.642198	-0.892474	0.05515	-0.02	13.46	0.16
21	C	-0.010092	-4.221385	-1.151024	0.043948	-0.95	9.87	-1.15
22	C	1.584619	-0.683357	-2.451114	0.06607	2.73	2.52	18.22
23	C	1.599396	-2.050755	-2.005217	-0.023532	-4.19	-8.85	-3.73
24	C	-4.072361	-3.781695	-0.941824	-0.019129	-3.45	-3.09	-7.15
25	C	-4.945621	-2.680505	-1.234358	0.0512	1.67	1.47	13.7
26	C	-4.995302	0.003317	-2.128671	0.028865	1.48	8.14	1.44
27	C	-4.166005	1.094479	-2.564714	-0.009317	-1.3	-3.33	-1.51
28	C	-1.497698	1.905755	-3.058463	-0.010383	-1.64	-3.63	-1.44
29	C	-0.123423	1.486362	-3.027721	0.030385	1.54	1.5	8.75
30	C	0.647817	1.541535	-1.832644	-0.009499	-1.55	-1.75	-3.69
31	C	1.546003	0.399379	-1.528935	-0.027125	-4.46	-4.87	-9.57
32	C	1.554947	-2.364602	-0.626616	0.008376	1.63	2.87	1.49
33	C	0.72503	-3.492009	-0.176321	-0.025298	-3.07	-7.59	-3.66
34	C	-2.01061	-4.334815	0.341796	-0.029132	-3.78	-4.43	-9.04
35	C	-3.414009	-3.890434	0.315926	0.007376	1.37	1.23	2.3
36	C	-5.201611	-0.254642	-0.745872	-0.010641	-1.51	-3.88	-1.74
37	C	-5.174516	-1.650911	-0.280223	-0.02055	-3.16	-3.5	-6.77
38	C	-2.142246	2.365594	-1.887115	0.001102	0.41	0.44	0.34
39	C	-3.529549	1.953686	-1.628335	0.000324	0.37	0.25	0.28
40	C	0.084181	2.280365	-0.761212	0.032773	1.07	1.05	8.91
41	C	-1.290901	2.642872	-0.777023	-0.012018	-1.83	-1.68	-4.12
42	C	1.775336	-1.273127	0.266509	-0.027721	-4.35	-3.97	-9.46

43	C	1.820577	0.078269	-0.175381	0.07132	2.12	1.97	18.91
44	C	0.184686	-3.454795	1.137975	0.044132	-1.06	9.63	-0.9
45	C	-1.173886	-3.878448	1.4001	0.057238	0.17	13.82	-0.03
46	C	-3.968294	1.854796	-0.279682	-0.009373	-1.34	-3.27	-1.16
47	C	-4.798681	0.769971	0.157601	0.030417	1.49	8.53	1.45
48	C	-4.743264	-1.913801	1.049794	0.050873	1.69	1.51	13.64
49	C	-3.870693	-3.014813	1.341738	-0.018166	-3.32	-2.98	-7.02
50	C	-3.018179	-2.63305	2.45405	0.042234	-0.16	10.65	-0.51
51	C	-1.687007	-3.056881	2.46666	0.007362	0.29	-3.47	-3.57
52	C	0.500962	-2.38598	2.050446	0.012507	-3.45	-3.32	1.46
53	C	1.290302	-1.30642	1.630727	0.058457	0.36	14.58	0.62
54	C	1.490147	0.954087	0.988141	-0.003996	-2.62	-1.81	-2.1
55	C	0.521115	2.175793	0.661204	-0.002204	-1.3	-0.77	-1.09
56	C	-1.794276	2.62601	0.581649	0.021326	-0.11	-0.07	5.5
57	C	-3.114441	2.242669	0.826496	0.009229	-0.86	2.07	-0.79
58	C	-4.409997	-0.84112	1.972068	-0.004622	-2.01	-2.47	-1.25
59	C	-4.441429	0.477414	1.533871	-0.011065	-1.29	-3.68	-1.61
60	C	-0.731617	2.25149	1.46877	0.000688	-1.82	-1.59	-0.6
61	C	-3.409762	1.405484	1.954942	0.009878	2.25	-0.45	-0.79
62	C	-3.352947	-1.290453	2.853209	0.017485	1.79	-4.04	-3.48
63	C	-0.642838	-2.137916	2.887267	-0.00107	0.34	-0.54	-0.26
64	C	1.012411	0.02812	2.059751	-0.004594	-4.53	-4.28	-2.01
65	C	-0.957684	-0.826385	3.266113	-0.024419	-9.66	-4.61	-5.14
66	C	-0.113446	0.28149	2.85002	0.070856	0.74	0.45	17.66
67	C	-1.000274	1.406187	2.543071	0.023263	-2.47	-2.44	4.75
68	C	-2.342215	-0.391436	3.257526	0.084914	21.71	0.98	1.29
69	C	-2.368686	0.973961	2.79552	0.00746	0.68	-3.96	-3.83
72	C	1.937153	2.395884	1.170395	-0.000667	-0.09	-0.3	0.03
73	C	2.940718	2.993529	0.209279	0.001683	-0.16	-0.12	0.12
74	C	4.13551	2.339888	-0.120066	-0.000403	0.23	0.28	0.48
75	C	5.083758	2.952197	-0.944805	0.000144	0.03	0	0.11
76	C	4.851375	4.232198	-1.453267	-0.000145	-0.04	-0.02	0.06
77	C	3.66267	4.894656	-1.131978	0.00012	-0.02	-0.03	0.06
78	C	2.718276	4.278676	-0.308694	-0.000881	-0.05	-0.02	0.12
83	C	2.070622	2.964529	2.59455	0.000391	0.06	-0.04	0.22
88	C	3.452031	2.830237	3.25912	0.000058	0.01	-0.01	0.13
91	C	3.865108	1.386972	3.566237	-0.000149	0.25	0.22	0.42
94	C	5.150252	1.29292	4.360656	0.000103	0.04	0.04	0.15
97	C	6.838946	-0.194122	5.105403	0.000006	-0.02	-0.02	0.04
70	H	6.664096	0.028134	6.162185	0	-0.07	-0.07	0.14
71	H	3.465337	5.887413	-1.529682	-0.000008	-0.1	-0.12	0.18
79	H	1.789528	4.793579	-0.074417	0.000029	-0.11	-0.16	0.45
80	H	4.320236	1.337762	0.256031	-0.000111	-0.19	-0.24	0.92
81	H	6.00051	2.423002	-1.193866	-0.000005	-0.11	-0.15	0.24
82	H	5.585388	4.706565	-2.100279	0.00001	-0.06	-0.06	0.18
84	H	3.431242	3.395004	4.19855	0.000034	-0.09	-0.11	0.34
85	H	7.075858	-1.250139	4.969151	0	-0.07	-0.08	0.15
86	H	1.314743	2.514916	3.244901	-0.000057	-0.42	-0.63	0.86
87	H	1.824635	4.033633	2.534435	0.000013	-0.17	-0.21	0.38
89	H	3.086192	0.87707	4.150879	-0.000024	-0.17	-0.33	0.54

90	H	4.220291	3.305482	2.638013	0.00001	-0.06	-0.09	0.35
92	H	3.978948	0.787074	2.657595	-0.000068	-0.1	-0.21	0.71
93	H	7.654315	0.435039	4.736917	0	-0.06	-0.07	0.13
95	O	5.689019	2.198294	4.967486	0			
96	O	5.644456	0.02993	4.339471	0.000004			

Table S9. Atomic number, type, coordinates of β_2-$PC_{71}BM$ radical anion structure optimized using B3LYP|6-31G+(d); Mulliken spin populations, and hyperfine coupling constants B3LYP|EPRII). Mulliken spin populations are unitless and sum to 1. The principal axes of the hyperfine coupling tensors for ^{13}C and ^{1}H are aligned with the principal axes of the g-tensor.

Atom Number	Atom Type	X coord (Å)	Y coord (Å)	Z coord (Å)	Spin Population	A_1 (MHz)	A_2 (MHz)	A_3 (MHz)
0	C	-1.782989	-3.361289	-4.087155	0.002091	-0.93	-4.75	-5.26
1	C	-0.930117	-2.25145	-4.508309	0.04119	9.24	-2.59	-2.82
2	C	-1.771581	-1.109138	-4.746757	0.077442	19.44	-0.01	-0.4
3	C	-3.145693	-1.505455	-4.460619	-0.021657	-9.51	-5.86	-4.89
4	C	-3.153233	-2.904805	-4.077897	0.099801	1.47	1.2	25.8
5	C	0.332235	-2.083116	-3.909155	0.014365	-1.93	2.16	-2.45
6	C	0.777008	-0.760854	-3.519194	-0.009677	-3.52	-1.95	-1.48
7	C	-0.035471	0.3429	-3.744569	-0.009007	-3.8	-2.66	-2.08
8	C	-1.331203	0.177611	-4.371357	0.015891	1.64	-4.28	-3.62
9	C	-2.229857	1.096081	-3.72533	0.046389	11.13	-0.62	-0.91
10	C	-3.550112	0.715658	-3.45497	0.025959	4.83	-2.19	-2.51
11	C	-4.011902	-0.605071	-3.832759	-0.002074	-0.91	-1.1	-0.24
12	C	-4.908775	-1.061005	-2.785595	0.004803	-3.19	-3.12	-0.23
13	C	-4.915446	-2.407003	-2.411543	0.034418	-0.68	-1.01	8.43
14	C	-4.027683	-3.339882	-3.056587	0.023481	-4.1	-4.7	2.74
15	C	-3.552502	-4.244708	-2.034316	0.003	-1.87	0.52	-1.95
16	C	-2.233203	-4.687191	-2.051132	-0.011098	-1.33	-1.86	-3.93
17	C	-1.330989	-4.244482	-3.097296	0.002861	-0.94	-1.35	0.42
18	C	-0.033473	-4.057185	-2.486303	0.023747	5.37	-0.65	-0.78
19	C	0.781151	-2.995799	-2.887458	0.025013	5.81	-0.84	-0.95
20	C	-1.490194	-4.800232	-0.812272	0.039054	1.66	10.59	1.59
21	C	-0.133707	-4.399489	-1.08357	-0.005889	-1.45	-3.04	-1.94
22	C	1.524546	-0.858989	-2.278032	0.042712	1.91	1.83	11.94
23	C	1.515349	-2.241984	-1.887286	-0.013633	-2.36	-4.98	-2.07
24	C	-4.167676	-3.886737	-0.765774	0.034042	0.73	0.56	8.96
25	C	-5.002083	-2.744469	-0.999889	-0.009911	-2.4	-2.03	-4.18
26	C	-5.016722	-0.02514	-1.788338	0.052645	0.56	13.34	0.71
27	C	-4.16672	1.071623	-2.202571	0.0282	-1.5	5.88	-1.64
28	C	-1.488304	1.835705	-2.720407	-0.021997	-3.92	-8.2	-3.47
29	C	-0.123424	1.384938	-2.737464	0.064129	2.61	2.43	17.67
30	C	0.668763	1.369086	-1.554905	-0.024737	-4.08	-4.48	-8.78
31	C	1.538314	0.188011	-1.312597	-0.014514	-2.36	-2.64	-5.3
32	C	1.494843	-2.609168	-0.521461	0.002066	0.61	0.8	0.53
33	C	0.645974	-3.733113	-0.09895	-0.001574	0.12	-0.32	-0.04
34	C	-2.094506	-4.536106	0.447399	-0.016154	-2.26	-5.47	-2.58
35	C	-3.487452	-4.060535	0.471416	-0.01225	-2.01	-2.26	-4.4
36	C	-5.19232	-0.332239	-0.410612	-0.025801	-3.56	-4.14	-8.43
37	C	-5.186995	-1.746508	-0.001686	0.004293	0.76	0.63	1.15
38	C	-2.096446	2.261648	-1.515787	0.006892	1.38	2.34	1.25
39	C	-3.488319	1.872971	-1.242649	-0.017332	-2.06	-5.3	-2.54
40	C	0.155094	2.087451	-0.445039	0.066732	2.03	1.9	17.52
41	C	-1.214136	2.477568	-0.415259	-0.024887	-3.98	-3.65	-8.68
42	C	1.76247	-1.562212	0.409418	-0.01518	-2.41	-2.23	-5.35

43	C	1.834498	-0.193416	0.022331	0.04344	1.4	1.34	11.49
44	C	0.128643	-3.733209	1.225016	-0.005611	-1.56	-2.74	-1.23
45	C	-1.222722	-4.136008	1.499599	0.04014	1.73	10.94	1.65
46	C	-3.902027	1.736987	0.110147	0.027554	-1.29	5.62	-1.18
47	C	-4.753273	0.646422	0.525794	0.055393	0.9	13.92	0.72
48	C	-4.731831	-2.074873	1.306635	-0.009336	-2.24	-1.89	-4.1
49	C	-3.897836	-3.216755	1.540968	0.032748	0.77	0.62	8.63
50	C	-3.006041	-2.887341	2.641952	0.002839	-1.4	-1.53	0.73
51	C	-1.692285	-3.341302	2.618125	-0.013019	-1.64	-4.44	-2.09
52	C	0.496696	-2.709661	2.181431	0.010735	-1.14	2.04	-1.05
53	C	1.297127	-1.637193	1.778845	0.028951	0.13	7.47	0.17
54	C	1.547705	0.643008	1.226731	-0.002723	-1.55	-0.97	-1.38
55	C	0.612548	1.909014	0.964553	-0.003842	-2.24	-1.51	-1.89
56	C	-1.688267	2.421615	0.951693	0.0518	0.56	13.34	0.73
57	C	-3.015986	2.053935	1.20396	0.009808	-2.81	-2.72	1.06
58	C	-4.36713	-1.047801	2.269154	0.030441	-0.22	7.94	-0.52
59	C	-4.372829	0.288903	1.871478	-0.002593	-1.43	-3.73	-3.25
60	C	-0.622607	1.981426	1.798115	-0.007801	-2.66	-4.59	-4.01
61	C	-3.305179	1.174678	2.304144	0.001736	1.05	-0.07	-0.14
62	C	-3.305277	-1.550336	3.103931	0.018373	1.73	-3.92	-3.41
63	C	-0.617318	-2.469691	3.054829	0.00728	2.01	-0.2	-0.52
64	C	1.063555	-0.310508	2.266683	-0.003058	-1.45	-2.55	-2.28
65	C	-0.895952	-1.162764	3.488367	-0.001381	-1.45	-4.74	-4.26
66	C	-0.035317	-0.059021	3.084488	0.032629	7.32	-1.68	-1.77
67	C	-0.889496	1.106026	2.851815	0.062456	15.57	0.42	0.18
68	C	-2.26121	-0.696145	3.521657	0.090701	23.15	1.36	1.58
69	C	-2.260571	0.691872	3.104758	-0.021888	-8.73	-4.42	-5.08
72	C	2.040731	2.058716	1.462681	-0.000578	0.6	0.4	0.7
74	C	2.741706	3.550366	5.464517	0.000007	0.03	0	0.11
76	C	3.258312	2.309803	5.082725	0.000072	-0.06	-0.04	0.07
77	C	3.010158	1.808674	3.801722	-0.000061	-0.04	-0.02	0.11
78	C	2.243425	2.537799	2.883085	0.000556	-0.01	-0.02	0.16
79	C	1.975478	4.286793	4.555836	0.000053	-0.04	-0.06	0.06
80	C	1.730216	3.78274	3.276971	0.000012	0.03	-0.01	0.15
81	C	7.374799	3.071259	-2.408916	0.000004	-0.01	-0.02	0.03
83	C	3.07561	2.638513	0.488556	0.000215	0.07	0.14	0.24
88	C	4.513449	2.155205	0.734276	-0.000156	-0.13	-0.11	0
91	C	5.449409	2.638369	-0.378562	0.000007	-0.01	-0.01	0.09
94	C	6.908271	2.281072	-0.142611	-0.000002	-0.03	-0.03	0.03
70	H	1.560086	5.249503	4.844714	0.00001	-0.09	-0.11	0.26
71	H	2.928957	3.9378	6.463236	0.000003	-0.08	-0.09	0.2
73	H	3.852652	1.726168	5.781626	0.000011	-0.09	-0.11	0.26
75	H	3.408754	0.839016	3.51712	0.000013	-0.21	-0.31	0.33
82	H	1.120963	4.352555	2.579283	-0.00006	-0.25	-0.35	0.4
84	H	4.537232	1.059428	0.769239	-0.00004	-0.19	-0.23	0.48
85	H	2.787717	2.398993	-0.538614	0.000318	-0.33	-0.13	0.6
86	H	6.854468	4.028485	-2.295779	0	-0.06	-0.07	0.13
87	H	3.0343	3.734351	0.571036	0.000011	-0.32	-0.36	0.29
89	H	5.124392	2.229744	-1.344402	0.000003	-0.12	-0.15	0.27
90	H	4.883526	2.50587	1.702435	-0.000009	-0.14	-0.16	0.24

92	H	5.383571	3.732572	-0.483855	-0.000007	-0.1	-0.12	0.21
93	H	8.292954	3.226298	-2.978419	0	-0.05	-0.05	0.1
96	H	6.734186	2.362357	-2.944723	0	-0.07	-0.09	0.16
95	O	7.33625	1.800165	0.881168	0.000003			
97	O	7.798301	2.543623	-1.146933	-0.000001			

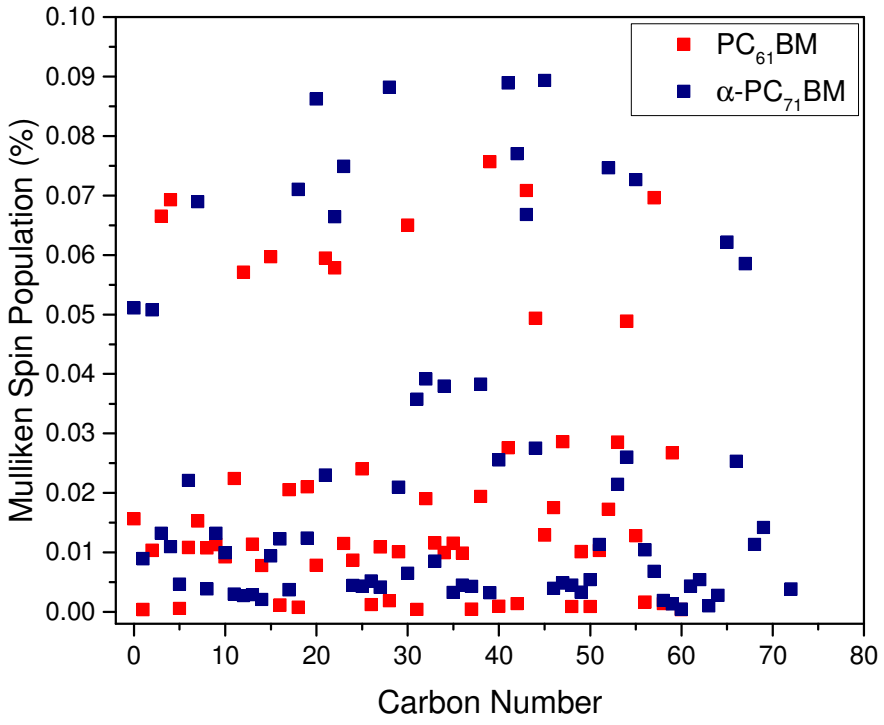


Figure S2. Mulliken spin populations for fullerene cage carbons for $PC_{61}BM$ (red) and α-$PC_{71}BM$ (blue) radical anions. Carbon numbers match those in Tables S6 and S7. Both molecules have approximately equal number of carbons with low spin density percentages (< 0.02 %), 60% for α-$PC_{71}BM$ and 66% for $PC_{61}BM$. The high spin density percentages (> .06 %) are also similar: 20% for $PC_{61}BM$ and 23% for α-$PC_{71}BM$.

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