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Supporting information for article:

When combined X-ray and polarized neutron diffraction data challenge high-level calculations: spin-resolved electron density of an organic radical

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Figure S1 Spin up charge density in planes defined by C1, N2, and O2 and C1, N1, and O1 of nitronyl nitroxide part; (C) spin down charge density in the same planes.

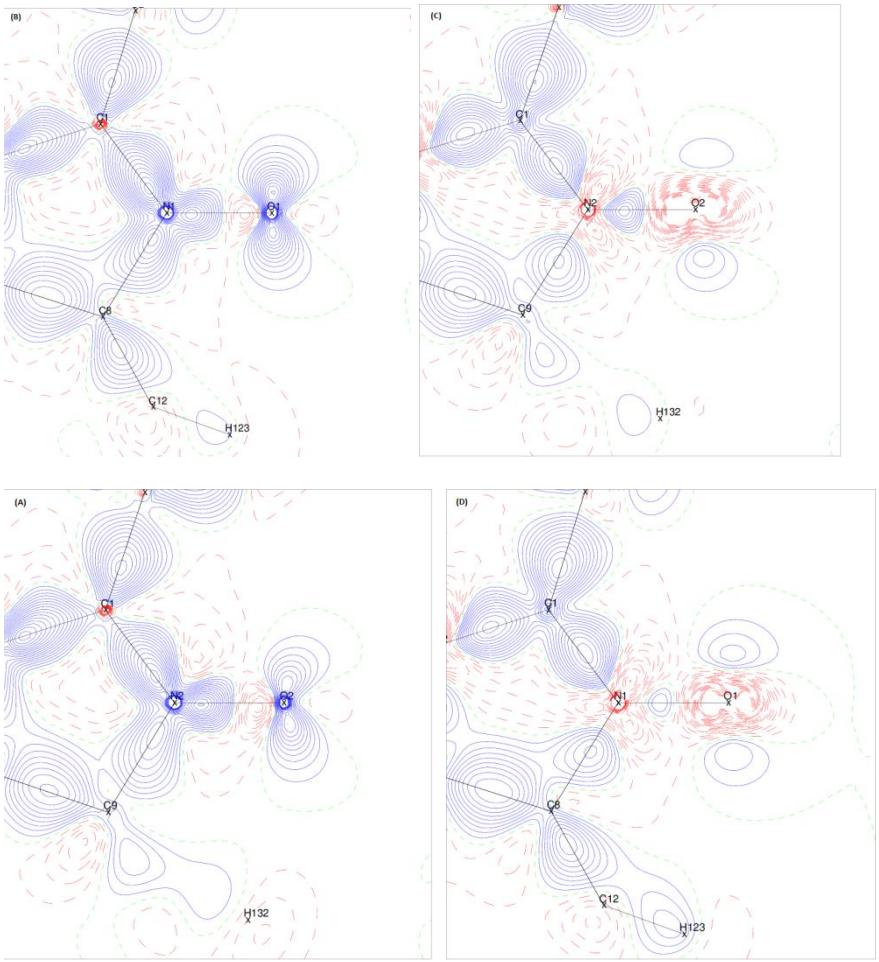

Table S1 Mulliken spin populations for the monomer using the ANO-L-VDZP basis set and considering different active spaces.

Atom	CASSCF(3,4)	CASSCF(5,6)	CASSCF(7,8)
O1	0.283	0.286	0.285
N1	0.329	0.326	0.323
C1	-0.181	-0.177	-0.174
N2	0.317	0.315	0.312
O2	0.235	0.237	0.243

Table S2 Mulliken spin populations for the dimer using the ANO-L-VDZP basis set and considering different active spaces.

Atom	CASSCF(6,6)	CASSCF(8,8)	CASSCF(10,10)
O1	0.313	0.313	0.311
N1	0.300	0.300	0.304
C1	-0.170	-0.170	-0.165
N2	0.329	0.329	0.319
O2	0.202	0.201	0.212
O'1	0.299	0.296	0.292
N'1	0.315	0.310	0.316
C'1	-0.180	-0.166	-0.166
N'2	0.331	0.336	0.330
O'2	0.207	0.202	0.206

Table S3 Mulliken spin populations for the monomer when unrestricted (U) and restricted open-shell (RO) DFT calculations are performed with the B3LYP, CAM-B3LYP and M06-2X functionals (cc-pVDZ basis-set). The results of the computations at unrestricted level with the M06-2X functional are reported in the text.

METHOD	O1	N1	C1	N2	O2	N1+O1	N2+O2
U-B3LYP	0.35	0.27	-0.19	0.26	0.34	0.62	0.60
U-CAM-B3LYP	0.36	0.30	-0.27	0.29	0.35	0.66	0.64
RO-B3LYP	0.28	0.21	0.01	0.19	0.25	0.49	0.44
RO-CAM-B3LYP	0.28	0.24	0.02	0.19	0.23	0.52	0.42
RO-M062X	0.29	0.24	0.02	0.19	0.22	0.53	0.41

Table S4 Bader spin populations for the monomer when unrestricted (U) and restricted open-shell (RO) DFT calculations are performed with the B3LYP, CAM-B3LYP and M06-2X functionals (cc-pVDZ basis-set). The results of the computations at unrestricted level with the M06-2X functional are reported in the text.

METHOD	**O1**	**N1**	**C1**	**N2**	**O2**	**N1+O1**	**N2+O2**
U-B3LYP	0.34	0.25	-0.14	0.24	0.33	0.59	0.57
U-CAM-B3LYP	0.36	0.27	-0.21	0.26	0.35	0.63	0.61
RO-B3LYP	0.28	0.21	0.01	0.19	0.26	0.49	0.45
RO-CAM-B3LYP	0.29	0.24	0.01	0.19	0.23	0.53	0.42
RO-M062X	0.30	0.24	0.01	0.18	0.21	0.54	0.40