Supplementary Data:

Clustering of Molecular Spins in the Crystals of Nitronylnitroxide and Iminonitroxide Triradicals Based on Benzene-1,3,5-Triyl Frameworks

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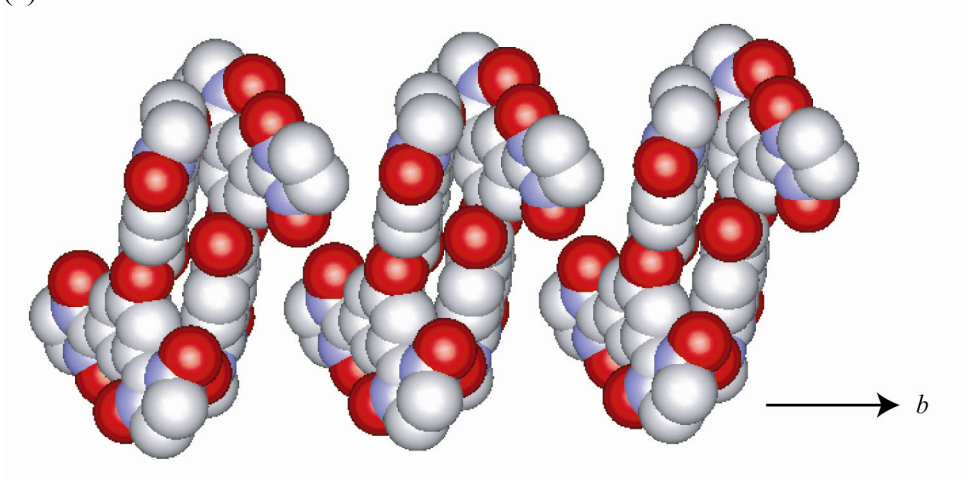
(a)



(b)

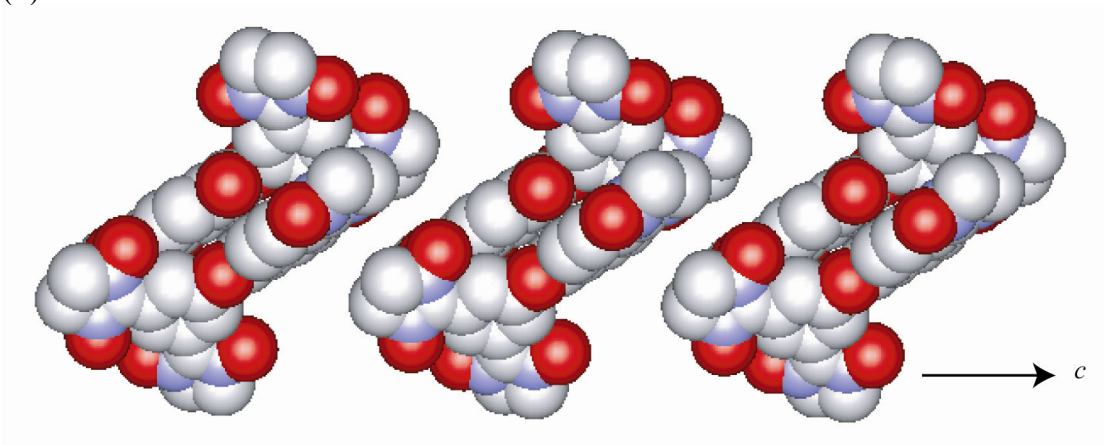
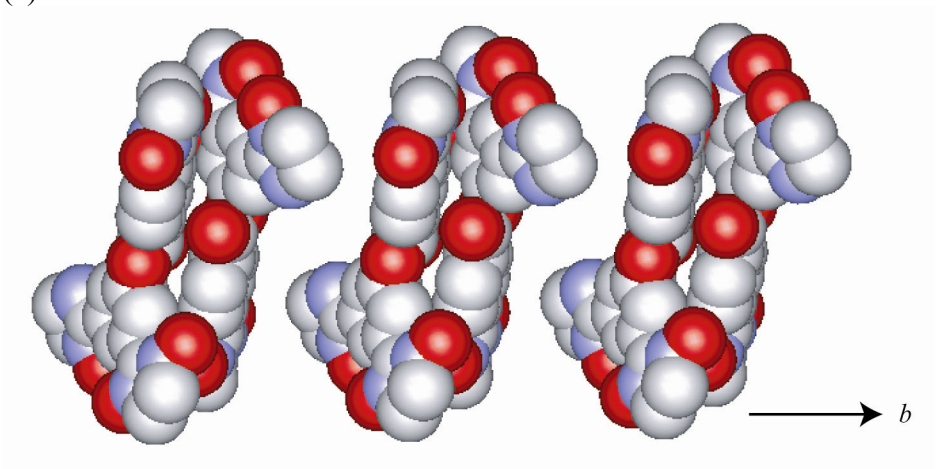


Fig. 1S Molecular packing of **trisNN** (**3**) in the head-to-tail motif along the *b* (a) and *c* axes (b). The methyl groups are omitted for clarity.

(a)



(b)

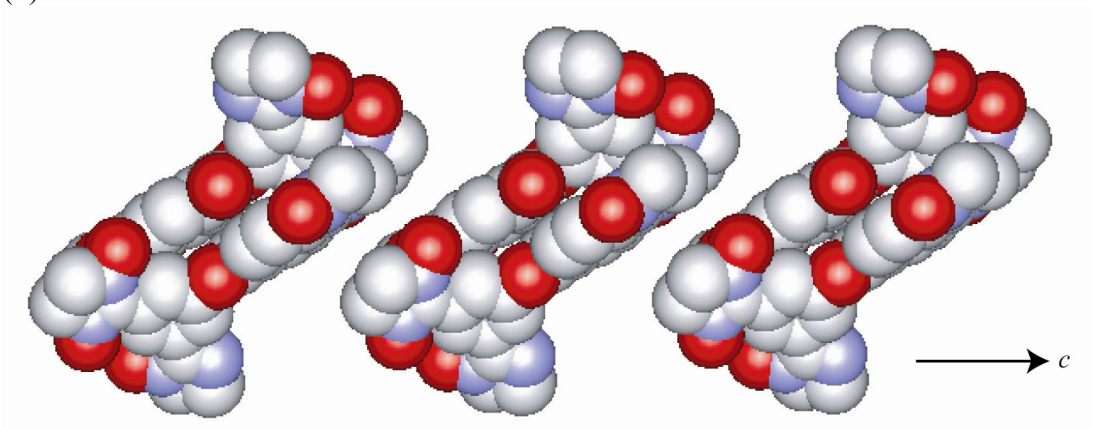


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Fig. 2S Molecular packing of **trisIN** (**4**) in the head-to-tail motif along the *b* (a) and *c* axes (b). The methyl groups are omitted for clarity.

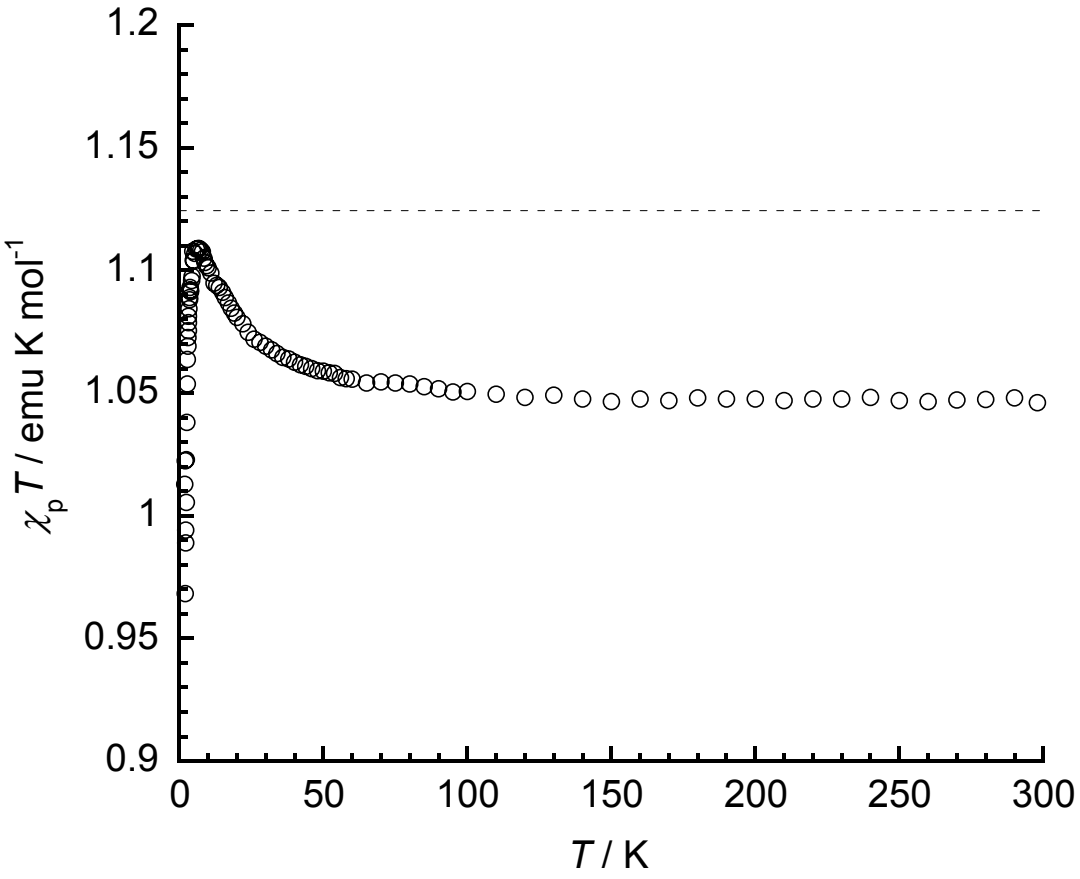

Fig. 3S Temperature dependence of magnetic susceptibility χ_p for **trisIN** (**4**) diluted in the PVC film in the $\chi_p T$ *vs.* T plots. The horizontal dotted lines show the Curie law for 3 moles of $S = 1/2$. Tthe $\chi_p T$ value of 1.05 emu K mol^{-1} at 298 K corresponds to the purity of 93% assuming $g = 2.0$ for the g-factor. The reduced purity is attributed to decomposition during the preparation of the PVC film by slow evaporation of a CH_2Cl_2 solution of **4**.

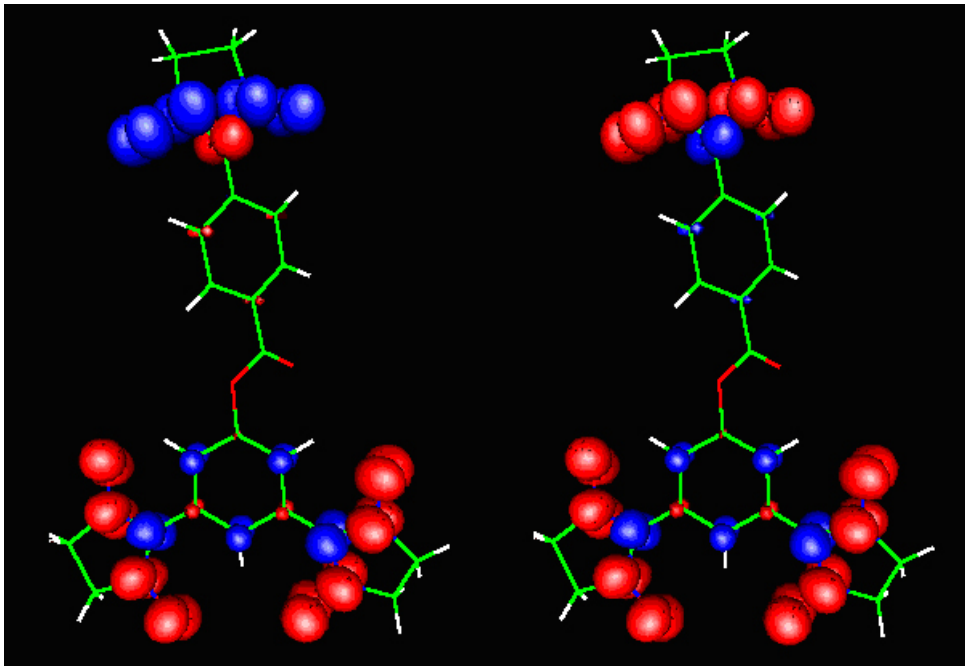

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Fig. 4S Three-dimensional representations of the spin density distribution in **trisNN** (**3**) for the lowest doublet (left) and the lowest quartet (right) states obtained from the DFT molecular orbital calculations. The red and blue surfaces denote the isocontour values of +0.005 au and –0.005 au, respectively. The methyl groups are replaced by hydrogen atoms in the calculations.

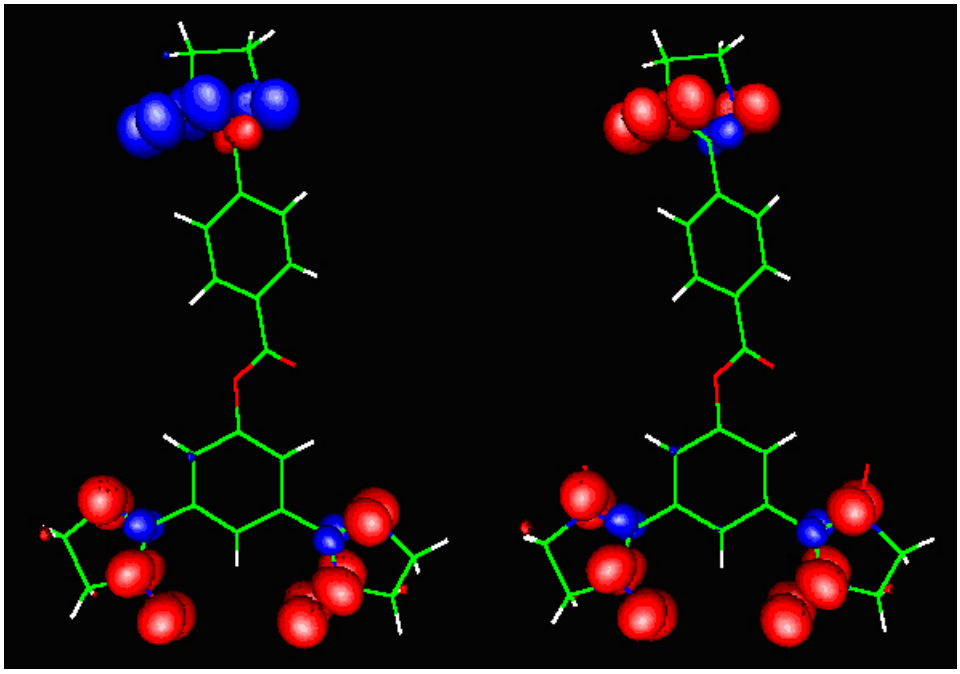


Fig. 5S Three-dimensional representations of the spin density distribution in **trisIN** (**4**) for the lowest doublet (left) and the lowest quartet (right) states obtained from the DFT molecular orbital calculations. The red and blue surfaces denote the isocontour values of +0.005 au and –0.005 au, respectively. The methyl groups are replaced by hydrogen atoms in the calculations.

Table 1S Atomic spin densities ρ of **trisNN** (**3**) calculated at UB3LYP/6-31+G(d,p) level.[a,b]

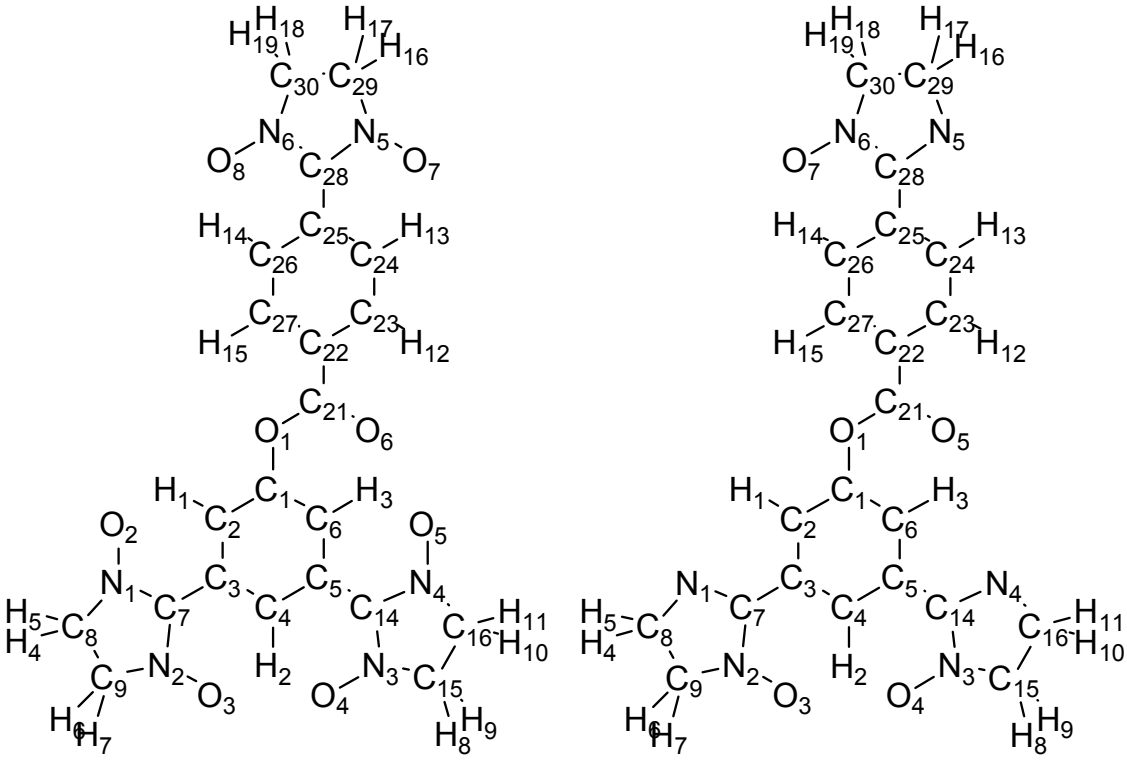
No.	atom	doublet	quartet
1	C1	0.060264	0.060066
2	C2	-0.118126	-0.118229
3	C3	0.082484	0.082471
4	C4	-0.096327	-0.096804
5	C5	0.068765	0.068938
6	C6	-0.091718	-0.092373
7	H1	0.003782	0.003800
8	H2	0.002898	0.002917
9	H3	0.003102	0.003135
10	O2	0.360757	0.360768
11	N1	0.301442	0.301447
12	C7	-0.221148	-0.221199
13	N2	0.259612	0.259624
14	O3	0.335420	0.335433
15	C8	-0.013741	-0.013738
16	C9	-0.016467	-0.016468
17	H4(C10)	0.003610	0.003611
18	H5(C11)	0.014756	0.014757
19	H6(C12)	0.002819	0.002819
20	H7(C13)	0.011074	0.011075
21	O4	0.354555	0.354565
22	N3	0.275956	0.275968
23	C14	-0.214357	-0.214395
24	N4	0.281916	0.281916
25	O5	0.343123	0.343166
26	C15	-0.017242	-0.017242
27	C16	-0.014886	-0.014896
28	H8(C17)	0.004310	0.004310
29	H9(C18)	0.013032	0.013032
30	H10(C19)	0.004205	0.004206
31	H11(C20)	0.012807	0.012808
32	O1	0.001262	-0.001372
33	O6	0.007242	-0.007165
34	C21	-0.004135	0.007662
35	C22	0.047345	-0.043253
36	C23	-0.032392	0.032388
37	C24	0.036919	-0.037066
38	C25	-0.032828	0.033172
39	C26	0.052032	-0.052022
40	C27	-0.025905	0.025911
41	H12	0.000933	-0.000956
42	H13	-0.001026	0.001033
43	H14	-0.001272	0.001276
44	H15	0.000963	-0.000976
45	O7	-0.352614	0.352639
46	N5	-0.291017	0.291040
47	C28	0.219463	-0.219502
48	N6	-0.279614	0.279638
49	O8	-0.341111	0.341143
50	C29	0.015899	-0.015898
51	C30	0.015410	-0.015414
52	H16(C31)	-0.013089	0.013090
53	H17(C32)	-0.003884	0.003885
54	H18(C33)	-0.003121	0.003122
55	H19(C34)	-0.012135	0.012136

[a] The expectation values $<S^2>$ before and after the spin-projection, *i.e.*, the annihilation of higher spin multiplicity components, are 1.9440 and 1.4941 for the doublet and 3.9443 and 3.7663 for the quartet state, respectively. [b] The atom numbering scheme is given in Scheme 1S.

Table 2S Atomic spin densities ρ of **trisIN** (**4**) calculated at UB3LYP/6-31+G(d,p) level.[a,b]

No.	atom	doublet	quartet
1	C1	0.026908	0.026666
2	C2	-0.052374	-0.052103
3	C3	0.031486	0.031420
4	C4	-0.029521	-0.029720
5	C5	0.025182	0.025293
6	C6	-0.039359	-0.039808
7	H1	0.001239	0.001249
8	H2	-0.000126	-0.000117
9	H3	0.000977	0.001008
10	N1	0.410064	0.410079
11	C7	-0.150523	-0.150549
12	N2	0.297873	0.297874
13	O3	0.448834	0.448834
14	C8	-0.012523	-0.012521
15	C9	-0.023480	-0.023482
16	H4(C10)	0.003666	0.003667
17	H5(C11)	0.018774	0.018775
18	H6(C12)	0.005569	0.005569
19	H7(C13)	0.018616	0.018616
20	O4	0.494624	0.494621
21	N3	0.332556	0.332560
22	C14	-0.119475	-0.119496
23	N4	0.300585	0.300612
24	C15	-0.028139	-0.028141
25	C16	-0.006501	-0.006503
26	H8(C17)	0.008081	0.008081
27	H9(C18)	0.023064	0.023064
28	H10(C19)	0.011018	0.011019
29	H11(C20)	0.002448	0.002449
30	O1	0.000718	-0.000986
31	O5	0.003488	-0.003585
32	C21	-0.003284	0.003178
33	C22	0.028575	-0.027002
34	C23	-0.023973	0.023966
35	C24	0.003534	-0.003622
36	C25	-0.000232	0.000428
37	C26	0.029751	-0.029803
38	C27	-0.014953	0.015236
39	H12	0.000933	-0.000956
40	H13	-0.001026	0.001033
41	H14	-0.001272	0.001276
42	H15	0.000963	-0.000976
43	N5	-0.373626	0.373663
44	C28	0.139914	-0.139937
45	N6	-0.323303	0.323305
46	O7	-0.461097	0.461098
47	C29	0.011503	-0.011505
48	C30	0.025101	-0.025100
49	H16(C31)	-0.013122	0.013124
50	H17(C32)	-0.002841	0.002842
51	H18(C33)	-0.006392	0.006392
52	H19(C34)	-0.019767	0.019767

[a] The expectation values $<S^2>$ before and after the spin-projection, *i.e.*, the annihilation of higher spin multiplicity components, are 1.8212 and 1.0017 for the doublet and 3.8212 and 3.7522 for the quartet state, respectively. [b] The atom numbering scheme is given in Scheme 1S.



Scheme 1S Atom numbering schemes of **trisNN** (**3**) (left) and **trisIN** (**4**) (right), corresponding to the DFT-calculated spin density distribution in Table 1S and Table 2S.