Supplementary Data:

Watson-Crick Pairing of Nucleobases Functionalized with Open-Shell Molecular Entities in Crystalline Solids

Tomoaki Ise,[a,b] Daisuke Shiomi,[*a,b] Kazunobu Sato,[a] and Takeji Takui[*a]

[a] Departments of Materials Science and Chemistry, Graduate School of Science, Osaka City University, Sumiyoshi-ku, Osaka 558-8585, Japan, Fax: +81 6 6605 3137; Tel: +81 6 6605 3149; E-mail: shiomi@sci.osaka-cu.ac.jp, and [b] PRESTO, Japan. Science and Technology Agency (JST), Honcho Kawaguchi, Saitama 332-0012, Japan.

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

No.	atom	ρ	No.	atom	ρ
1	C1	-0.206720	26	C10	0.023091
2	C2	-0.003473	27	C11	-0.039156
3	C3	-0.006094	28	C12	0.022517
4	C4	-0.005362	29	C13	-0.040339
5	C5	0.016805	30	C14	0.002260
6	C6	0.016188	31	H13	0.001737
7	C7	-0.006132	32	H14	-0.000839
8	N1	0.298271	33	H15	-0.000935
9	N2	0.297224	34	H16	0.001581
10	O1	0.323855	35	H17	-0.000092
11	O2	0.324361	36	H18	-0.000637
12	H1	-0.000619	37	C15	0.000125
13	H2	-0.000193	38	C16	0.000053
14	H3	-0.000339	39	C17	0.000037
15	H4	-0.001081	40	C18	-0.000327
16	H5	0.000838	41	C19	-0.000395
17	H6	-0.000871	42	N3	-0.001813
18	H7	-0.001071	43	N4	-0.000076
19	H8	0.000907	44	N5	-0.000084
20	H9	-0.000867	45	N6	0.000264
21	H10	-0.000352	46	N7	0.000003
22	H11	-0.000192	47	H19	-0.000003
23	H12	-0.000608	48	H20	0.000012
24	C8	0.037722	49	H21	-0.000002
25	C9	-0.049179	50	H22	-0.000001

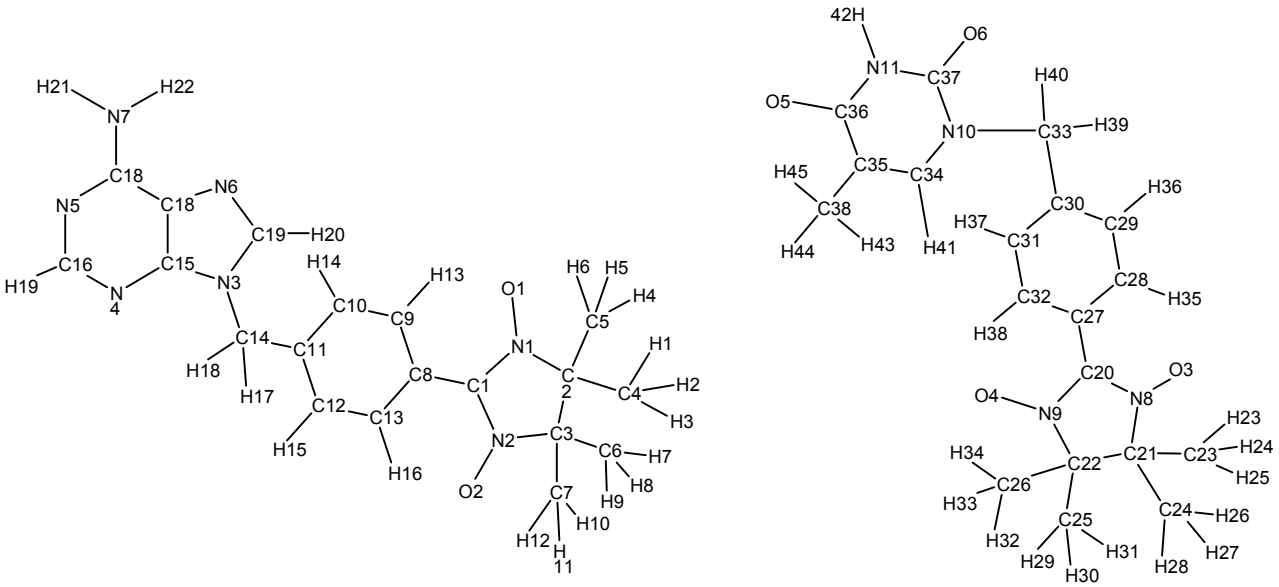
[a] The atom numbering scheme is given in Scheme 1S.

[b] The expectation value $<S^2>$ before and after the spin-projection, i.e., the annihilation of higher spin multiplicity components, are 0.8057 and 0.7505 for the doublet state.

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

No.	atom	ρ	No.	atom	ρ
1	C20	-0.217178	26	C29	0.016645
2	C21	-0.012096	27	C30	-0.031994
3	C22	-0.029930	28	C31	0.017147
4	C23	0.007637	29	C32	-0.034814
5	C24	0.007933	30	C33	0.001853
6	C25	0.014980	31	H35	0.001468
7	C26	0.014535	32	H36	-0.000698
8	O3	0.320348	33	H37	-0.000714
9	O4	0.317203	34	H38	0.001517
10	N8	0.296825	35	H39	-0.000833
11	N9	0.303931	36	H40	-0.000159
12	H23	0.000158	37	C34	-0.000785
13	H24	-0.000480	38	C35	0.000519
14	H25	-0.000768	39	C36	-0.000022
15	H26	-0.000592	40	C37	0.000030
16	H27	0.000043	41	C38	-0.000151
17	H28	-0.000462	42	O5	0.000037
18	H29	-0.000677	43	O6	-0.000098
19	H30	-0.000016	44	N10	-0.000525
20	H31	-0.000481	45	N11	-0.000015
21	H32	-0.000847	46	H41	0.000011
22	H33	0.000245	47	H42	-0.000005
23	H34	-0.000559	48	H43	0.000002
24	C27	0.041286	49	H44	0.000000
25	C28	-0.029456	50	H45	0.000003

[a] The atom numbering scheme is given in Scheme 1S.
[b] The expectation value $<S^2>$ before and after the spin-projection, i.e., the annihilation of higher spin multiplicity components, are 0.7999 and 0.7504 for the doublet state.



Scheme 1S. Atom numbering schemes of A-NN (left) and T-NN (right), corresponding to the DFT-calculated spin density distribution in Table 1S and Table 2S.