Supporting Information

for

Organic Magnetic Diradicals (Radical-Coupler-Radical): Standardization of Couplers for Strong Ferromagnetism

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Table of Contents

1. Zero-point corrected total energies, average spin square $< S^2 >$ and intramolecular magnetic exchange coupling constants (*J*).

Table S1. Calculated zero-point corrected total energies (sum of electronic and zero-point vibrational energies) in atonic unit (a.u.), the average spin square $< S^2 >$ and intramolecular magnetic exchange coupling constants (J in cm^{-1}) by using UB3LYP/6-311++G(d,p) level for DTDA group

Diradicals	E_T in au ($\langle S^2 \rangle_T$)	E_{BS} in au ($\langle S^2 \rangle_{BS}$)	J^Y (cm^{-1})
DTDA group-(i)	-2383.602223 (2.046667)	-2383.602001 (1.036274)	48.2
DTDA group-(ii)	-2119.072254 (2.038226)	-2119.0721 (1.033249)	33.6
DTDA group-(iii)	-2439.868364 (2.039695)	-2439.868227 (1.034581)	29.9
DTDA group-(iv)	-2135.122403 (2.037766)	-2135.122258 (1.033007)	31.7
DTDA group-(v)	-2135.117739 (2.037918)	-2135.117627 (1.034085)	24.5
DTDA group-(vi)	-2116.879249 (2.039272)	-2116.879137 (1.035019)	24.5
DTDA group-(vii)	-2272.655535 (2.042711)	-2272.655434 (1.035752)	22.0
DTDA group-(viii)	-2097.017377 (2.037178)	-2097.017306 (1.034722)	15.5
DTDA group-(ix)	-2129.113433 (2.036523)	-2129.113367 (1.034121)	14.4
DTDA group-(x)	-2135.120347 (2.03234)	-2135.1206 (1.040959)	-56.0
DTDA group-(xi)	-2119.072491 (2.032031)	-2119.072747 (1.041347)	-56.7
DTDA group-(xii)	-2097.018921 (2.031975)	-2097.019268 (1.044982)	-77.1
DTDA group-(xiii)	-2116.878012 (2.033071)	-2116.878413 (1.048321)	-89.3
DTDA group-(xiv)	-2439.867468 (2.032924)	-2439.867852 (1.048337)	-85.5
DTDA group-(xv)	-2383.603431 (2.034617)	-2383.603978 (1.06088)	-123.2

Table S2. Calculated zero-point corrected total energies (sum of electronic and zero-point vibrational energies) in atonic unit (a.u.), the average spin square $< S^2 >$ and intramolecular magnetic exchange coupling constants (J in cm^{-1}) by using UB3LYP/6-311++G(d,p) level for OVER group

Diradicals	E_T in au ($\langle S^2 \rangle_T$)	E_{BS} in au ($\langle S^2 \rangle_{BS}$)	J^Y (cm^{-1})
OVER group-(i)	-1396.018702 (2.059415)	-1396.018418 (1.04631)	61.5
OVER group-(ii)	-1131.487575 (2.04916)	-1131.487328 (1.042954)	53.8
OVER group-(iii)	-1452.285042 (2.051355)	-1452.284854 (1.04471)	41.0
OVER group-(iv)	-1147.538406 (2.04873)	-1147.538224 (1.042812)	39.7
OVER group-(v)	-1147.533778 (2.048363)	-1147.533666 (1.043555)	24.4
OVER group-(vi)	-1129.296007 (2.050463)	-1129.295857 (1.045064)	32.7
OVER group-(vii)	-1285.067489 (2.055074)	-1285.067346 (1.045637)	31.1
OVER group-(viii)	-1109.43266 (2.048306)	-1109.432553 (1.044965)	23.4
OVER group-(ix)	-1141.532188 (2.046996)	-1141.53215 (1.043938)	8.3
OVER group-(x)	-1147.536479 (2.041881)	-1147.536765 (1.051949)	-63.4
OVER group-(xi)	-1131.488135 (2.041676)	-1131.488438 (1.052715)	-67.2
OVER group-(xii)	-1109.435244 (2.042343)	-1109.435571 (1.05657)	-72.8
OVER group-(xiii)	-1129.295084 (2.043026)	-1129.295496 (1.060117)	-91.9
OVER group-(xiv)	-1452.284677 (2.043107)	-1452.285091 (1.060884)	-92.4
OVER group-(xv)	-1396.020191 (2.044707)	-1396.020776 (1.075455)	-132.4

Table S3. Calculated zero-point corrected total energies (sum of electronic and zero-point vibrational energies) in atonic unit (a.u.), the average spin square $< S^2 >$ and intramolecular magnetic exchange coupling constants (J in cm^{-1}) by using UB3LYP/6-311++G(d,p) level for NN group

Diradicals	E_T in au ($\langle S^2 \rangle_T$)	E_{BS} in au ($\langle S^2 \rangle_{BS}$)	J^Y (cm-1)
NN group-(i)	-1563.10501 (2.152948)	-1563.104403 (1.118735)	128.7
NN group-(ii)	-1298.563827 (2.120128)	-1298.563604 (1.10683)	48.3
NN group-(iii)	-1619.369583 (2.127576)	-1619.36926 (1.110529)	69.7
NN group-(iv)	-1314.614327 (2.12263)	-1314.614081 (1.108688)	53.2
NN group-(v)	-1314.607655 (2.119327)	-1314.60753 (1.110658)	27.2
NN group-(vi)	-1296.377764 (2.128626)	-1296.377504 (1.113988)	56.2
NN group-(vii)	-1452.146862 (2.114568)	-1452.146703 (1.103445)	34.5
NN group-(viii)	-1276.523033 (2.109524)	-1276.522952 (1.103676)	17.7
NN group-(ix)	-1308.608764 (2.111233)	-1308.608764 (1.108945)	0.0
NN group-(x)	-1314.611483 (2.107046)	-1314.612183 (1.138329)	-158.5
NN group-(xi)	-1298.564886 (2.103742)	-1298.565776 (1.141719)	-202.9
NN group-(xii)	-1276.524321 (2.095279)	-1276.525714 (1.146422)	-322.0
NN group-(xiii)	-1296.371229 (2.107816)	-1296.372822 (1.1735)	-373.9
NN group-(xiv)	-1619.370189 (2.101507)	-1619.371868 (1.16954)	-395.1
NN group-(xv)	-1563.112822 (2.105006)	-1563.115044 (1.207676)	-543.1

2. Calculated dihedral angles

Table S4. Calculated dihedral angles (˚) of DTDA group diradicals at UB3LYP/6-311++G(d,p) level

Diradical	Triplet state		BS state	
	Dihedral angle θ_1	Dihedral angle θ_2	Dihedral angle θ_1	Dihedral angle θ_2
DTDA group-(i)	-5.42	0.15	-5.38	0.10
DTDA group-(ii)	0.00	0.00	0.00	0.00
DTDA group-(iii)	-0.02	0.02	-0.02	0.02
DTDA group-(iv)	0.00	0.00	0.00	0.00
DTDA group-(v)	0.00	0.00	0.01	0.00
DTDA group-(vi)	0.01	0.00	0.00	0.00
DTDA group-(vii)	-0.01	0.00	0.00	0.00
DTDA group-(viii)	0.00	0.00	0.00	0.00
DTDA group-(ix)	0.00	0.00	0.00	0.00
DTDA group-(x)	-0.01	0.00	0.00	0.00
DTDA group-(xi)	0.00	-0.01	0.00	0.00
DTDA group-(xii)	0.00	0.00	-0.01	0.00
DTDA group-(xiii)	0.00	0.00	0.00	0.00
DTDA group-(xiv)	0.00	0.00	0.00	0.00
DTDA group-(xv)	-5.14	5.132	5.18	-5.17

Table S5. Calculated dihedral angles (˚) of OVER group diradicals at UB3LYP/6-311++G(d,p) level

Diradical	Triplet state		BS state	
	Dihedral angle θ_1	Dihedral angle θ_2	Dihedral angle θ_1	Dihedral angle θ_2
OVER group-(i)	-7.62	-0.33	-7.66	-0.54
OVER group-(ii)	0.00	0.00	-0.02	-0.05
OVER group-(iii)	-0.01	-0.12	-0.01	-0.13
OVER group-(iv)	0.12	-0.32	0.12	-0.32
OVER group-(v)	-2.81	-2.83	-3.60	-3.59
OVER group-(vi)	0.05	-0.03	0.05	-0.04
OVER group-(vii)	3.91	3.99	4.49	4.57
OVER group-(viii)	0.05	-0.05	-0.04	0.03
OVER group-(ix)	-1.34	0.71	-3.85	1.11
OVER group-(x)	-2.98	-0.75	-2.12	-0.57
OVER group-(xi)	0.01	0.01	0.00	0.00
OVER group-(xii)	-0.08	-0.08	-0.08	-0.08
OVER group-(xiii)	-0.48	-0.48	-0.47	-0.47
OVER group-(xiv)	-0.10	-0.10	-0.11	-0.11
OVER group-(xv)	7.53	-7.50	7.45	-7.44

Table S6. Calculated dihedral angles (˚) of NN group diradicals at UB3LYP/6-311++G(d,p) level

Diradical	Triplet state		BS state	
	Dihedral angle θ_1	Dihedral angle θ_2	Dihedral angle θ_1	Dihedral angle θ_2
NN group-(i)	-10.422	4.81	-10.26	6.45
NN group-(ii)	23.20	23.20	23.46	23.46
NN group-(iii)	-0.20	-0.79	-0.20	-0.81
NN group-(iv)	22.67	22.67	22.89	22.89
NN group-(v)	33.97	33.96	34.24	34.29
NN group-(vi)	2.71	-0.07	2.75	-0.05
NN group-(vii)	33.35	33.33	33.43	33.43
NN group-(viii)	0.99	0.48	1.00	0.52
NN group-(ix)	65.40	0.92	66.15	0.82
NN group-(x)	36.36	15.14	32.97	12.55
NN group-(xi)	16.09	16.09	14.67	14.67
NN group-(xii)	0.89	0.89	0.88	0.89
NN group-(xiii)	12.23	12.22	11.15	11.15
NN group-(xiv)	0.42	0.42	0.53	0.53
NN group-(xv)	10.35	-10.45	10.27	-10.04

3. Isotropic nucleus independent chemical shift at the center of the aromatic rings ($NICS(0)_{iso}$)

Table S7. Calculated isotropic nucleus independent chemical shift of parent coupler ($NICS_P$), triplet state ($NICS_T$), BS state ($NICS_{BS}$), difference in triplet and BS states ($\Delta NICS_{T-BS}$), difference in parent coupler and triplet state ($\Delta NICS_{P-T}$), difference in parent coupler and BS state ($\Delta NICS_{P-BS}$), and magnetic coupling constant *J* (in cm^{-1}) for DTDA group diradicals

diradicals	$NICS_P$	$NICS_T$	$NICS_{BS}$	$\Delta NICS_{T-BS}$	$\Delta NICS_{P-T}$	$\Delta NICS_{P-BS}$	*J* (cm^{-1})
DTDA group-(i)	-5.3576	-3.7225	-3.7618	0.0393	-1.6351	-1.5958	48.2
DTDA group-(ii)	-8.0460	-6.7091	-6.7896	0.0805	-1.3369	-1.2564	33.6
DTDA group-(iii)	-12.9180	-9.9378	-10.0020	0.0642	-2.9802	-2.9160	29.9
DTDA group-(iv)	-6.8422	-5.7693	-5.8503	0.0810	-1.0729	-0.9919	31.7
DTDA group-(v)	-6.8423	-6.1403	-6.1857	0.0454	-0.7020	-0.6566	24.5
DTDA group-(vi)	-11.9192	-9.5492	-9.6028	0.0536	-2.3700	-2.3164	24.5
DTDA group-(vii)	-17.0427	-13.2475	-13.2897	0.0422	-3.7952	-3.7530	22.0
DTDA group-(viii)	-13.6425	-10.5897	-10.6624	0.0727	-3.0528	-2.9801	15.5
DTDA group-(ix)	-13.0861	-10.4699	-10.4798	0.0099	-2.6162	-2.6063	14.4
DTDA group-(x)	-6.8429	-5.9372	-5.8594	-0.0778	-0.9057	-0.9835	-56.0
DTDA group-(xi)	-8.0459	-6.7842	-6.7067	-0.0775	-1.2617	-1.3392	-56.7
DTDA group-(xii)	-13.6429	-10.2764	-10.2451	-0.0313	-3.3665	-3.3978	-77.1
DTDA group-(xiii)	-11.9222	-9.354	-9.2843	-0.0697	-2.5682	-2.6379	-89.3
DTDA group-(xiv)	-12.9196	-9.3817	-9.3100	-0.0717	-3.5379	-3.6096	-85.5
DTDA group-(xv)	-5.3576	-3.1846	-2.9809	-0.2037	-2.173	-2.3767	-123.2

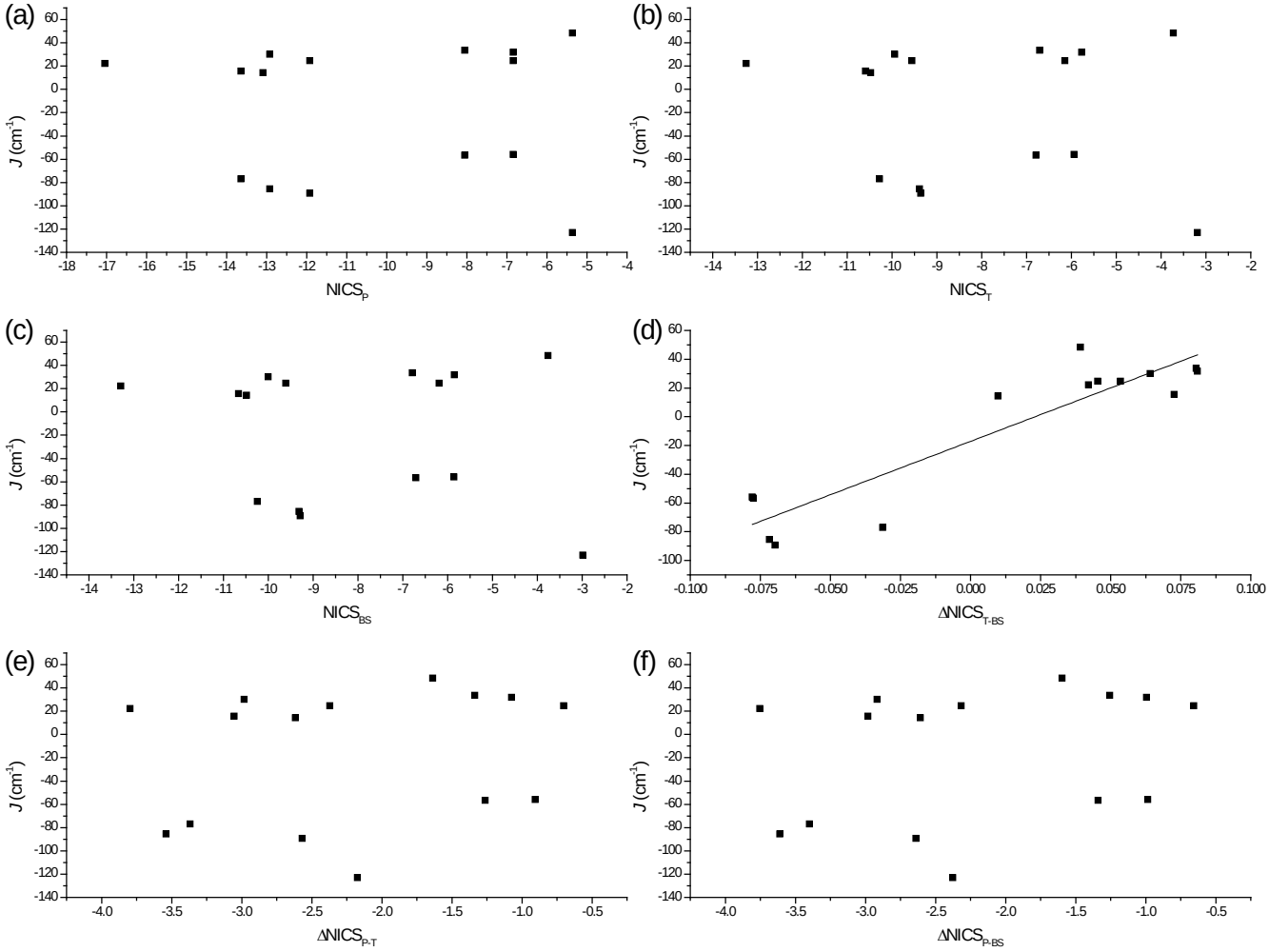

Figure S1. Correlation between (a) $NICS_P$, (b) $NICS_T$, (c) $NICS_{BS}$, (d) $\Delta NICS_{T\text{-}BS}$, (e) $\Delta NICS_{P\text{-}T}$, (f) $\Delta NICS_{P\text{-}BS}$ and magnetic coupling constant (J in cm^{-1}) for DTDA group diradicals.

Table S8. Calculated isotropic nucleus independent chemical shift of parent coupler ($NICS_P$), triplet state ($NICS_T$), BS state ($NICS_{BS}$), difference in triplet and BS states ($\Delta NICS_{T-BS}$), difference in parent coupler and triplet state ($\Delta NICS_{P-T}$), difference in parent coupler and BS state ($\Delta NICS_{P-BS}$), and magnetic coupling constant *J* (in cm^{-1}) for OVER group diradicals

diradicals	$NICS_P$	$NICS_T$	$NICS_{BS}$	$\Delta NICS_{T-BS}$	$\Delta NICS_{P-T}$	$\Delta NICS_{P-BS}$	*J* (cm^{-1})
OVER group-(i)	-5.3576	-3.4320	-3.4818	0.0498	-1.9256	-1.8758	61.5
OVER group-(ii)	-8.0460	-6.6352	-6.7487	0.1135	-1.4108	-1.2973	53.8
OVER group-(iii)	-12.918	-9.8983	-9.9802	0.0819	-3.0197	-2.9378	41.0
OVER group-(iv)	-6.8422	-5.7359	-5.843	0.1071	-1.1063	-0.9992	39.7
OVER group-(v)	-6.8423	-6.0008	-6.0648	0.0640	-0.8415	-0.7775	24.4
OVER group-(vi)	-11.9192	-9.615	-9.6843	0.0693	-2.3042	-2.2349	32.7
OVER group-(vii)	-17.0427	-13.951	-14.022	0.0710	-3.0917	-3.0207	31.1
OVER group-(viii)	-13.6425	-10.5579	-10.6480	0.0901	-3.0846	-2.9945	23.4
OVER group-(ix)	-13.0861	-10.4992	-10.5054	0.0062	-2.5869	-2.5807	8.3
OVER group-(x)	-6.8429	-5.8255	-5.7305	-0.0950	-1.0174	-1.1124	-63.4
OVER group-(xi)	-8.0459	-6.5765	-6.4696	-0.1069	-1.4694	-1.5763	-67.2
OVER group-(xii)	-13.6429	-10.2172	-10.2011	-0.0161	-3.4257	-3.4418	-72.8
OVER group-(xiii)	-11.9222	-9.4814	-9.4157	-0.0657	-2.4408	-2.5065	-91.9
OVER group-(xiv)	-12.9196	-9.454	-9.3739	-0.0801	-3.4656	-3.5457	-92.4
OVER group-(xv)	-5.3576	-3.082	-2.5045	-0.5775	-2.2756	-2.8531	-132.4

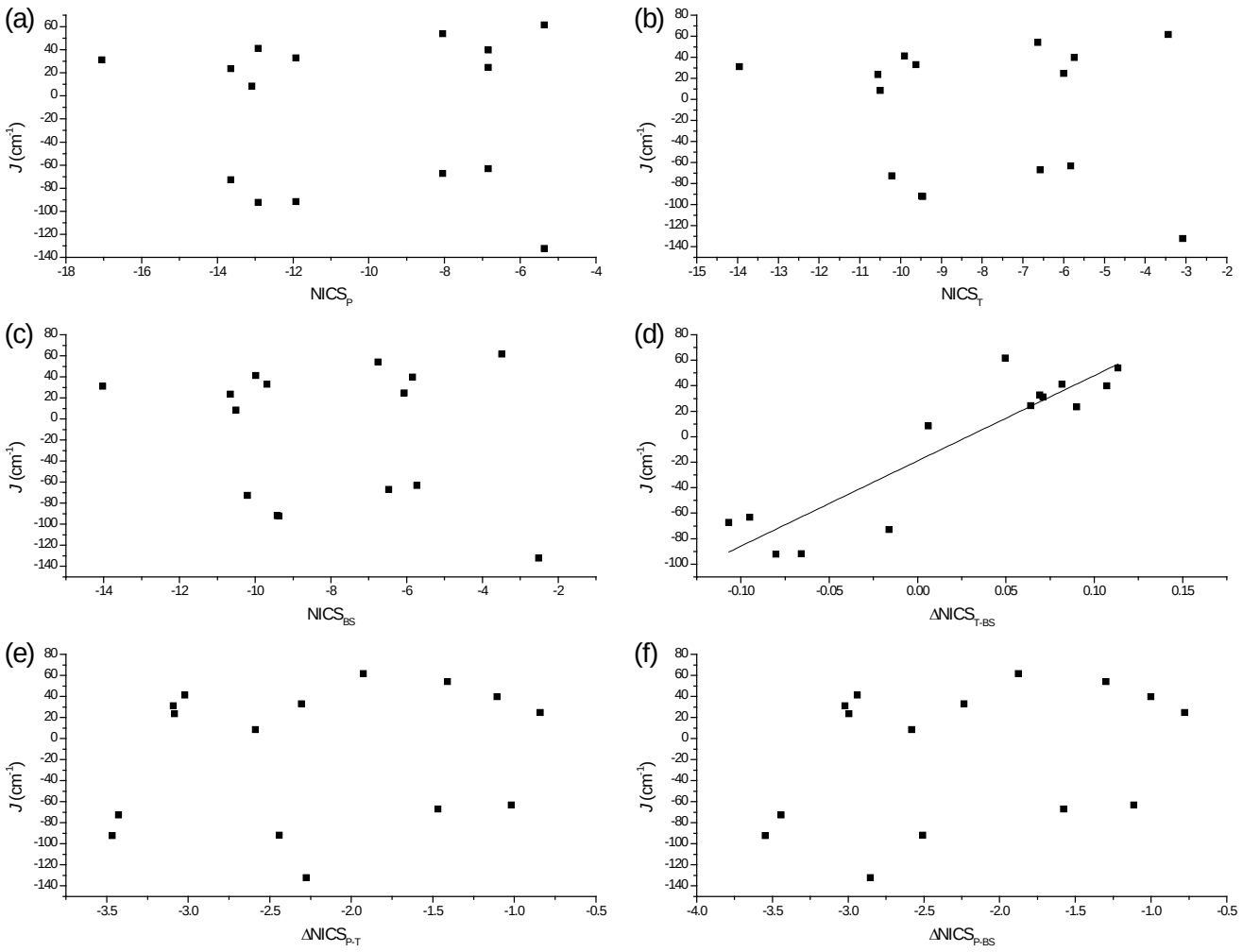

Figure S2. Correlation between (a) $NICS_P$, (b) $NICS_T$, (c) $NICS_{BS}$, (d) $\Delta NICS_{T\text{-}BS}$, (e) $\Delta NICS_{P\text{-}T}$, (f) $\Delta NICS_{P\text{-}BS}$ and magnetic coupling constant (J in cm^{-1}) for OVER group diradicals.

Table S9. Calculated isotropic nucleus independent chemical shift of parent coupler ($NICS_P$), triplet state ($NICS_T$), BS state ($NICS_{BS}$), difference in triplet and BS states ($\Delta NICS_{T-BS}$), difference in parent coupler and triplet state ($\Delta NICS_{P-T}$), difference in parent coupler and BS state ($\Delta NICS_{P-BS}$), and magnetic coupling constant *J* (in cm^{-1}) for NN group diradicals.

diradicals	$NICS_P$	$NICS_T$	$NICS_{BS}$	$\Delta NICS_{T-BS}$	$\Delta NICS_{P-T}$	$\Delta NICS_{P-BS}$	*J* (cm^{-1})
NN group-(i)	-5.3576	-5.7189	-5.8428	0.1239	0.3613	0.4852	128.7
NN group-(ii)	-8.0460	-7.3685	-7.4959	0.1274	-0.6775	-0.5501	48.3
NN group-(iii)	-12.918	-11.8744	-12.0062	0.1318	-1.0436	-0.9118	69.7
NN group-(iv)	-6.8422	-6.1977	-6.3229	0.1252	-0.6445	-0.5193	53.2
NN group-(v)	-6.8423	-6.6727	-6.7368	0.0641	-0.1696	-0.1055	27.2
NN group-(vi)	-11.9192	-11.0732	-11.1808	0.1076	-0.8460	-0.7384	56.2
NN group-(vii)	-17.0427	-14.0132	-14.0122	-0.0010	-3.0295	-3.0305	34.5
NN group-(viii)	-13.6425	-12.6252	-12.7025	0.0773	-1.0173	-0.9400	17.7
NN group-(ix)	-13.0861	-12.5333	-12.5420	0.0087	-0.5528	-0.5441	0.0
NN group-(x)	-6.8429	-6.5989	-6.3840	-0.2149	-0.2440	-0.4589	-158.5
NN group-(xi)	-8.0459	-7.6433	-7.3998	-0.2435	-0.4026	-0.6461	-202.9
NN group-(xii)	-13.6429	-12.7843	-12.4796	-0.3047	-0.8586	-1.1633	-322.0
NN group-(xiii)	-11.9222	-11.0455	-10.7641	-0.2814	-0.8767	-1.1581	-373.9
NN group-(xiv)	-12.9196	-11.9787	-11.6463	-0.3324	-0.9409	-1.2733	-395.1
NN group-(xv)	-5.3576	-6.9179	-6.7059	-0.2120	1.5603	1.3483	-543.1

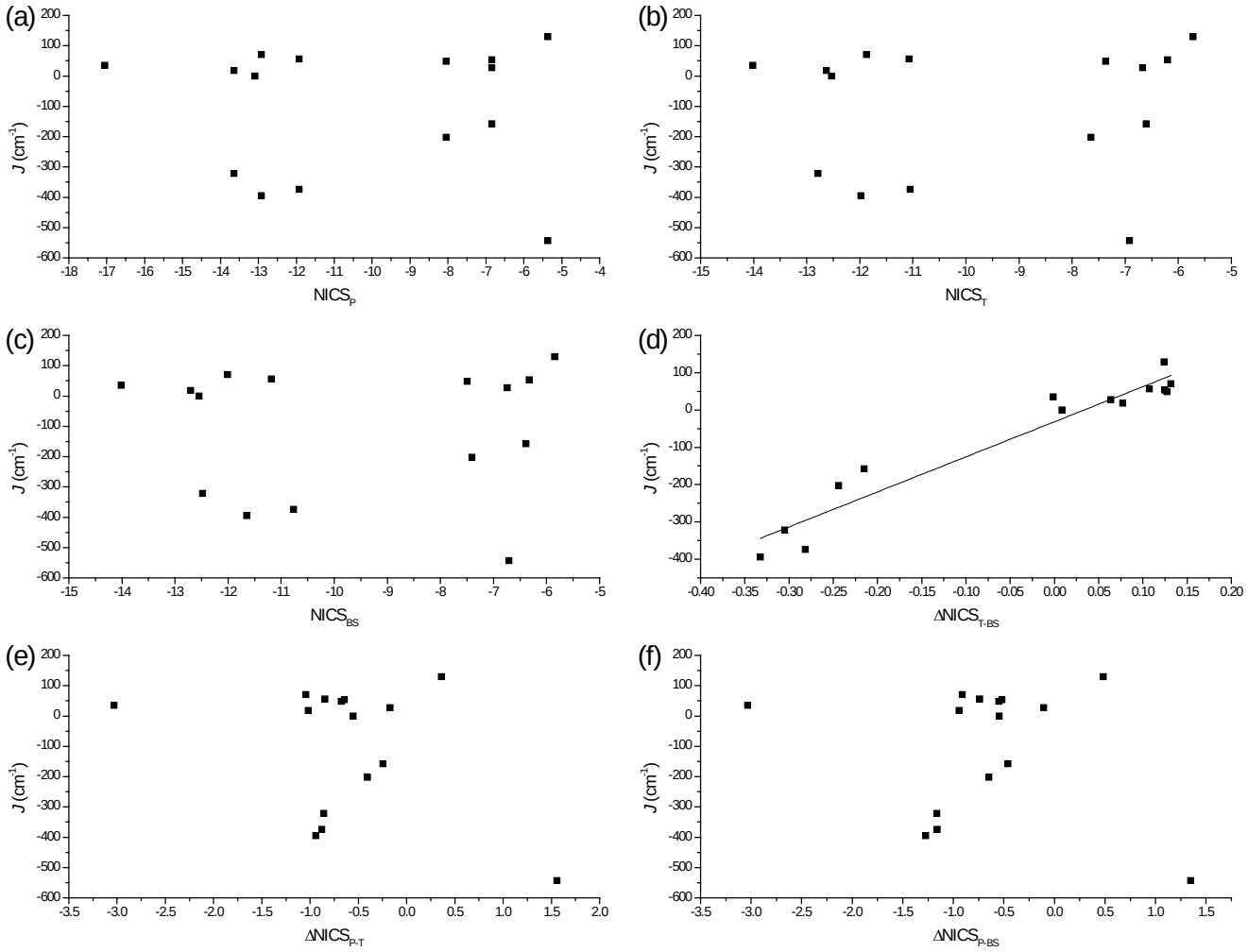

Figure S3. Correlation between (a) $NICS_P$, (b) $NICS_T$, (c) $NICS_{BS}$, (d) $\Delta NICS_{T\text{-}BS}$, (e) $\Delta NICS_{P\text{-}T}$, (f) $\Delta NICS_{P\text{-}BS}$ and magnetic coupling constant (J in cm^{-1}) for NN group diradicals.

4. Bond orders of diradicals

Table S10. Averaged bond orders of connecting bonds in triplet state (BO_T), BS state (BO_{BS}), difference between BO_T and BO_{BS} ($\Delta BO_{T\text{-}BS}$), and *J* values (in cm^{-1}) for DTDA group diradicals.

diradicals	BO_T	BO_{BS}	$\Delta BO_{T\text{-}BS}$	*J* (cm^{-1})
DTDA group-(i)	1.0466	1.0450	0.0015	48.2
DTDA group-(ii)	1.0270	1.0264	0.0006	33.6
DTDA group-(iii)	1.0459	1.0450	0.0009	29.9
DTDA group-(iv)	1.0264	1.0258	0.0006	31.7
DTDA group-(v)	1.0028	1.0023	0.0005	24.5
DTDA group-(vi)	1.0453	1.0445	0.0007	24.5
DTDA group-(vii)	1.0704	1.0698	0.0005	22.0
DTDA group-(viii)	1.0568	1.0564	0.0004	15.5
DTDA group-(ix)	1.0146	1.0143	0.0003	14.4
DTDA group-(x)	1.0177	1.0196	-0.0019	-56.0
DTDA group-(xi)	1.0290	1.0306	-0.0016	-56.7
DTDA group-(xii)	1.0608	1.0636	-0.0028	-77.1
DTDA group-(xiii)	1.0503	1.0535	-0.0032	-89.3
DTDA group-(xiv)	1.0610	1.0642	-0.0032	-85.5
DTDA group-(xv)	1.0690	1.0742	-0.0052	-123.2

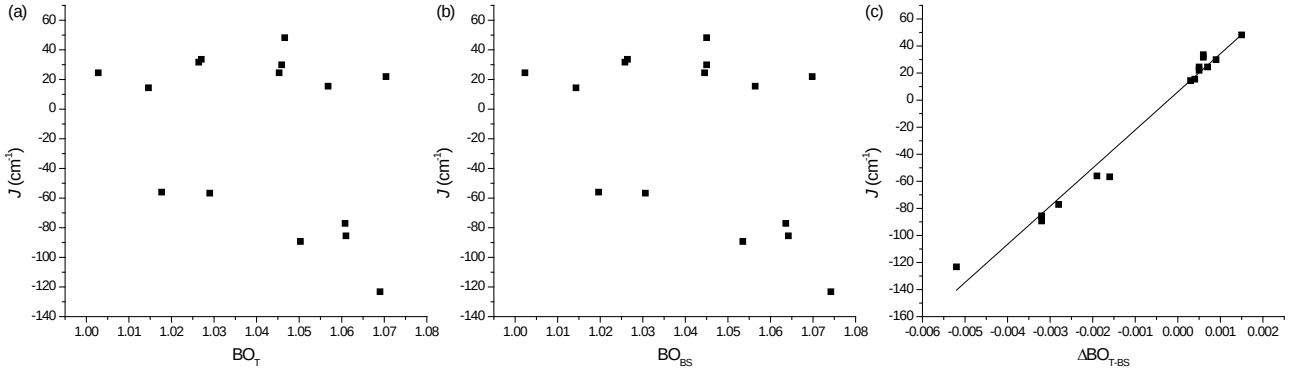

Figure S4. Correlation between BO_T, BO_{BS}, $\Delta BO_{T\text{-}BS}$ and *J* values (in cm^{-1}) for DTDA group diradicals.

Table S11. Averaged bond orders of connecting bonds in triplet state (BO_T), BS state (BO_{BS}), difference between BO_T and BO_{BS} (ΔBO_{T-BS}), and *J* values (in cm^{-1}) for OVER group diradicals.

diradicals	BO_T	BO_{BS}	ΔBO_{T-BS}	*J* (cm^{-1})
OVER group-(i)	1.0588	1.0567	0.0020	61.5
OVER group-(ii)	1.0389	1.0377	0.0012	53.8
OVER group-(iii)	1.0563	1.0551	0.0012	41.0
OVER group-(iv)	1.0388	1.0379	0.0009	39.7
OVER group-(v)	1.0168	1.0158	0.0010	24.4
OVER group-(vi)	1.0566	1.0557	0.0009	32.7
OVER group-(vii)	1.0735	1.0724	0.0010	31.1
OVER group-(viii)	1.0664	1.06585	0.0006	23.4
OVER group-(ix)	1.0292	1.0285	0.0007	8.3
OVER group-(x)	1.0310	1.0334	-0.0024	-63.4
OVER group-(xi)	1.0412	1.0439	-0.0027	-67.2
OVER group-(xii)	1.0716	1.0750	-0.0034	-72.8
OVER group-(xiii)	1.0623	1.0663	-0.004	-91.9
OVER group-(xiv)	1.0726	1.0762	-0.0036	-92.4
OVER group-(xv)	1.0817	1.0876	-0.0059	-132.4

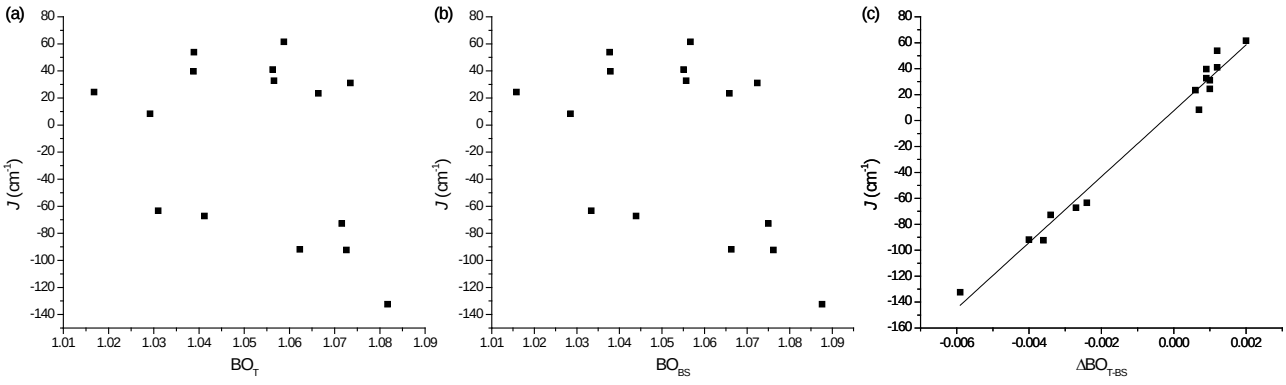

Figure S5. Correlation between BO_T, BO_{BS}, ΔBO_{T-BS} and *J* values (in cm^{-1}) for OVER group diradicals.

Table S12. Averaged bond orders of connecting bonds in triplet state (BO_T), BS state (BO_{BS}), difference between BO_T and BO_{BS} ($\Delta BO_{T\text{-}BS}$), and *J* values (in cm^{-1}) for NN group diradicals.

diradicals	BO_T	BO_{BS}	$\Delta BO_{T\text{-}BS}$	*J* (cm^{-1})
NN group-(i)	1.1108	1.1054	0.0054	128.7
NN group-(ii)	1.0793	1.0765	0.0028	48.3
NN group-(iii)	1.1127	1.1099	0.0027	69.7
NN group-(iv)	0.99	1.23645	-0.24645[b]	53.2
NN group-(v)	1.2454	1.2435	0.0019	27.2
NN group-(vi)	[a]	1.1051	[a]	56.2
NN group-(vii)	1.2703	1.2666	0.0037	34.5
NN group-(viii)	[a]	[a]	[a]	17.7
NN group-(ix)	1.0615	1.0609	0.0006	0.0
NN group-(x)	1.1598	1.1159	0.0439[b]	-158.5
NN group-(xi)	1.0925	1.0983	-0.0057	-202.9
NN group-(xii)	1.1315	1.1442	-0.0126	-322.0
NN group-(xiii)	1.0455	1.0626	-0.0171	-373.9
NN group-(xiv)	1.2075	1.2271	-0.0196	-395.1
NN group-(xv)	1.1727	1.1989	-0.0262	-543.1

[a] unable to obtain Wiberg bond indices by NBO analysis.

[b] not displayed in the linear regression.

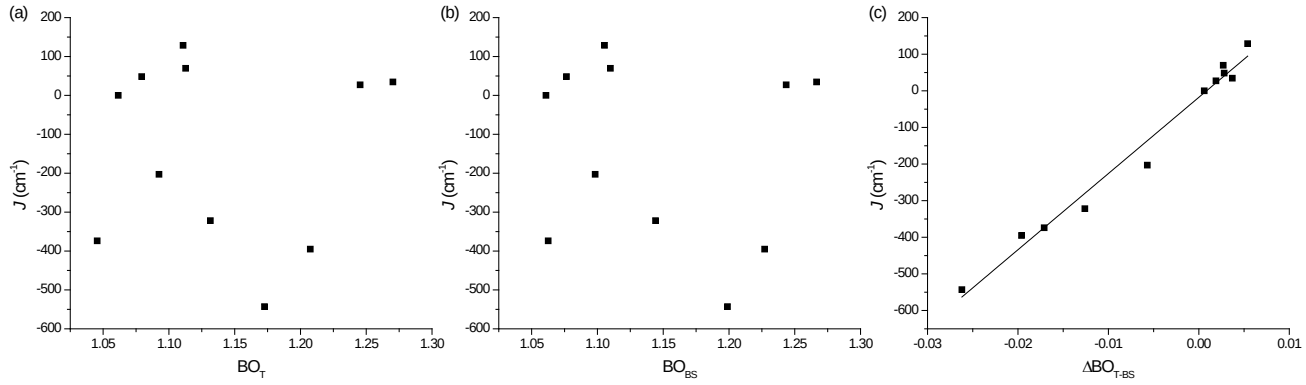

Figure S6. Correlation between BO_T, BO_{BS}, $\Delta BO_{T\text{-}BS}$ and *J* values (in cm^{-1}) for NN group diradicals.

5. Mulliken atomic spin density distributions.

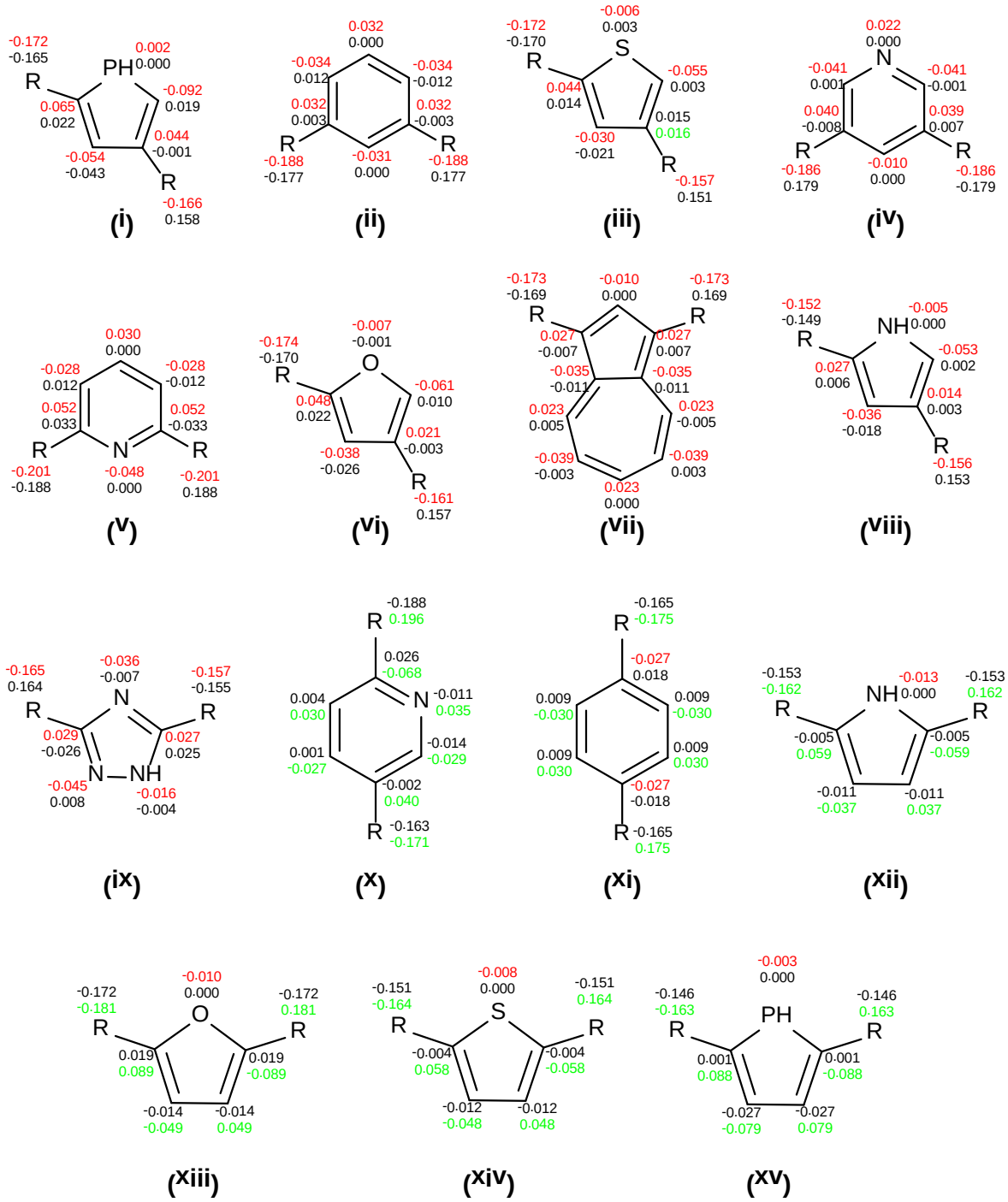

Figure S7. Mulliken atomic spin density distributions of DTDA group diradicals. Dominant spin polarizations are colored (red: dominant in triplet state, green: dominant in BS state).

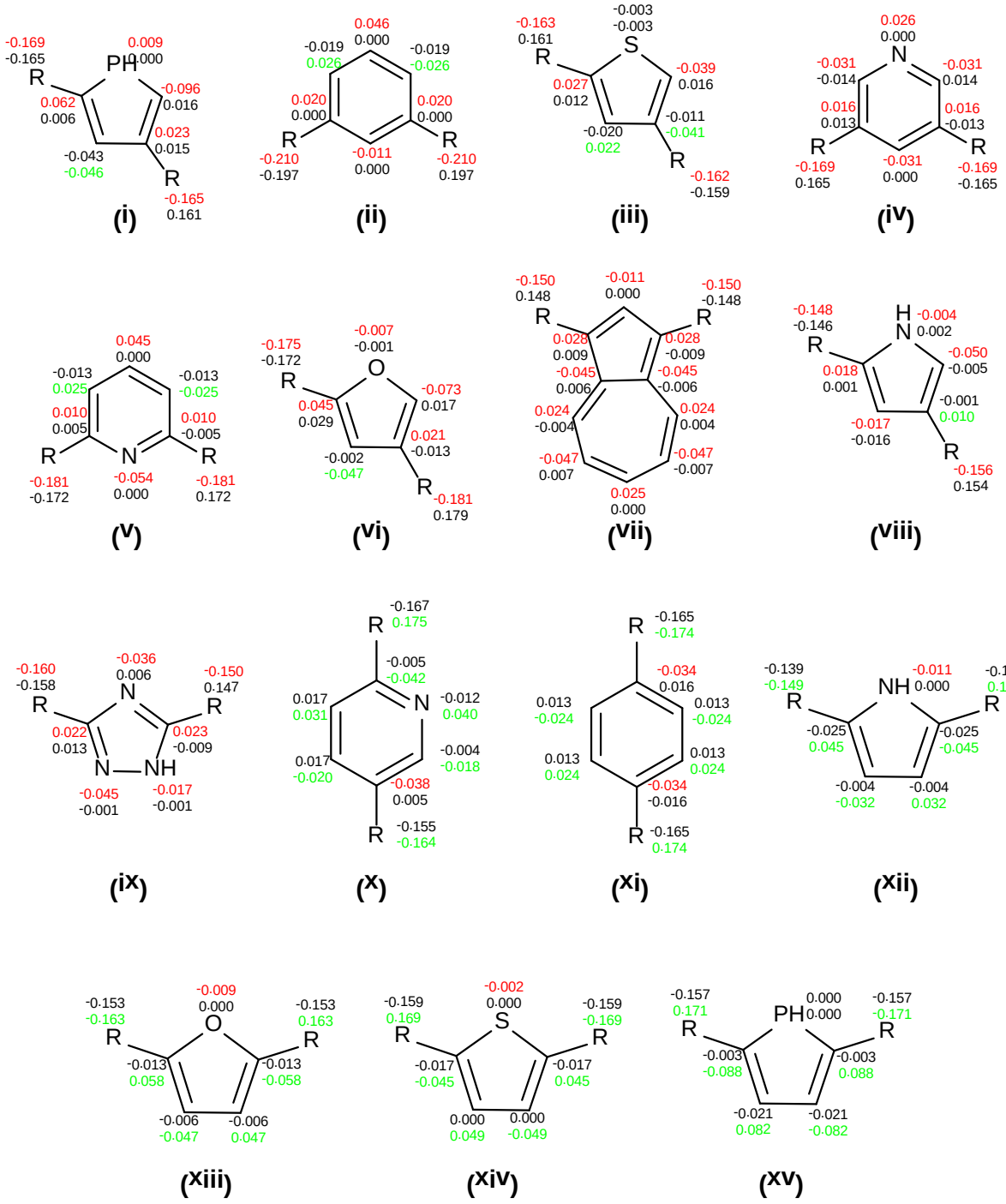

Figure S8. Mulliken atomic spin density distributions of OVER group diradicals. Dominant spin polarizations are colored (red: dominant in triplet state, green: dominant in BS state).

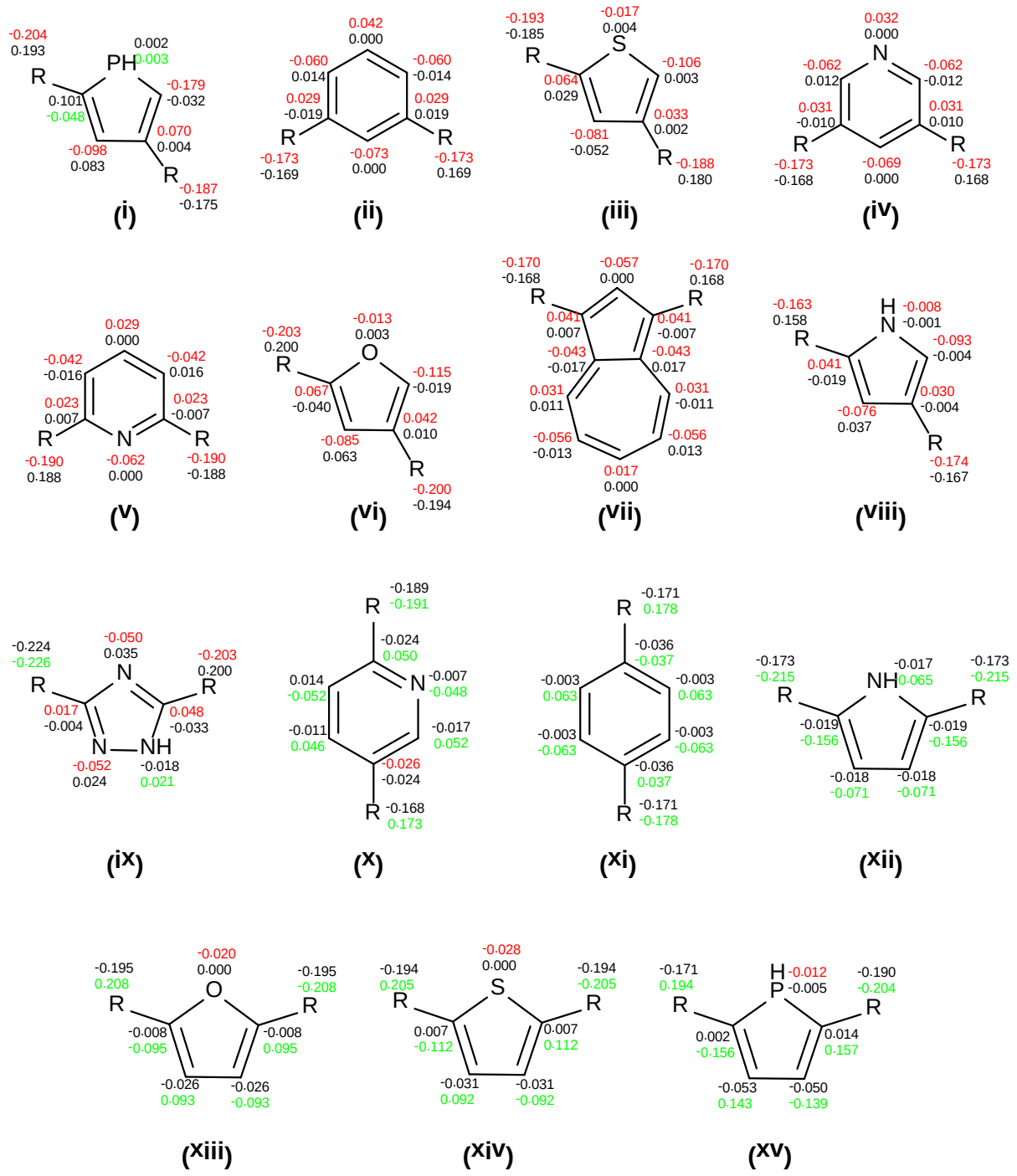

Figure S9. Mulliken atomic spin density distributions of NN group diradicals. Dominant spin polarizations are colored (red: dominant in triplet state, green: dominant in BS state).

6. Spin density distributions

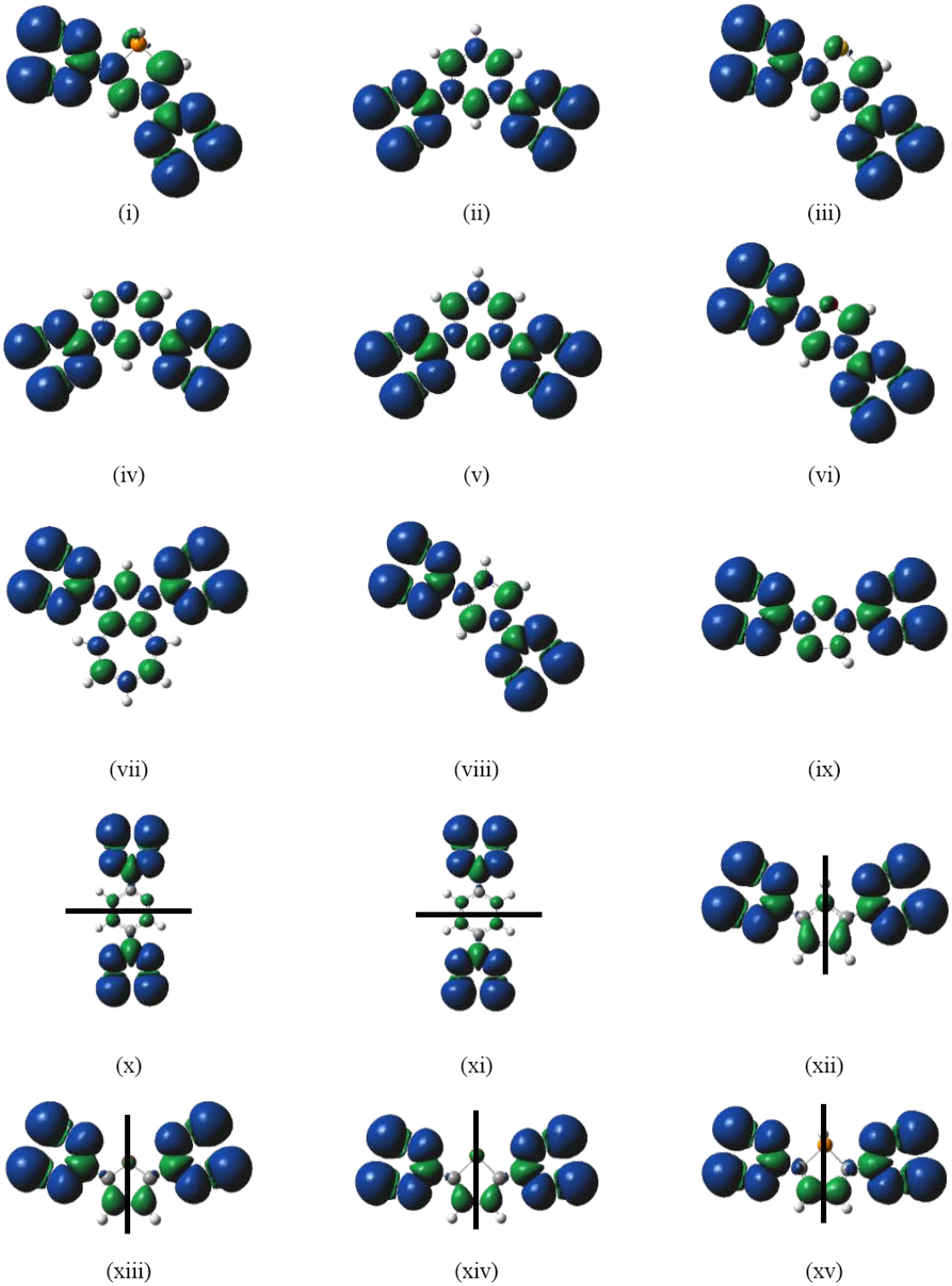

Figure S10. Plots of spin density distributions in the triplet states of DTDA group diradicals. Blue and green colors represent α- and β-spin, respectively. Black solid line indicates the mismatching of the spin polarization.

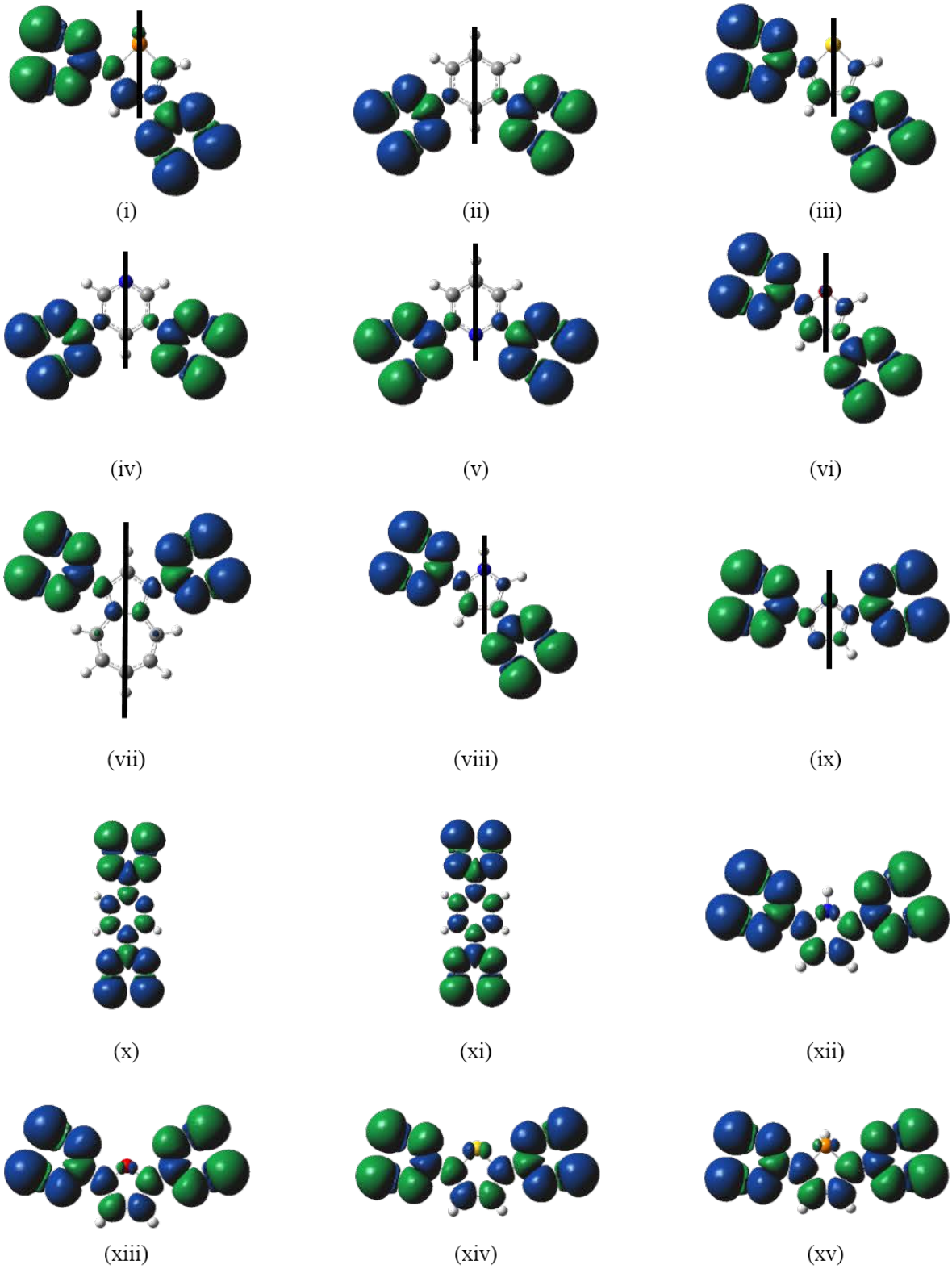

Figure S11. Plots of spin density distributions in the BS states of DTDA group diradicals. Blue and green colors represent α- and β-spin, respectively. Black solid line indicates the mismatching of the spin polarization.

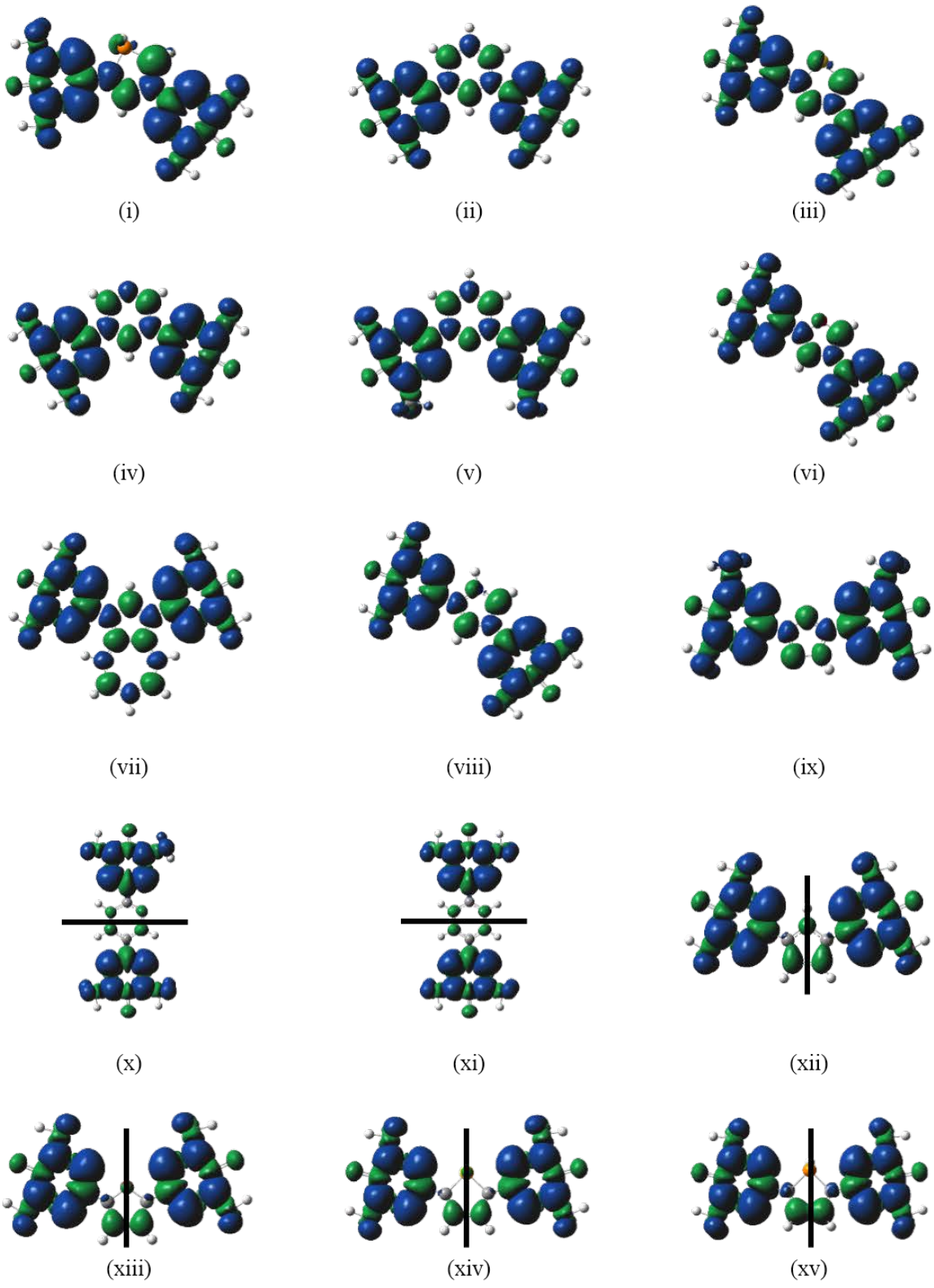

Figure S12. Plots of spin density distributions in the triplet states of VER group diradicals. Blue and green colors represent α- and β-spin, respectively. Black solid line indicates the mismatching of the spin polarization.

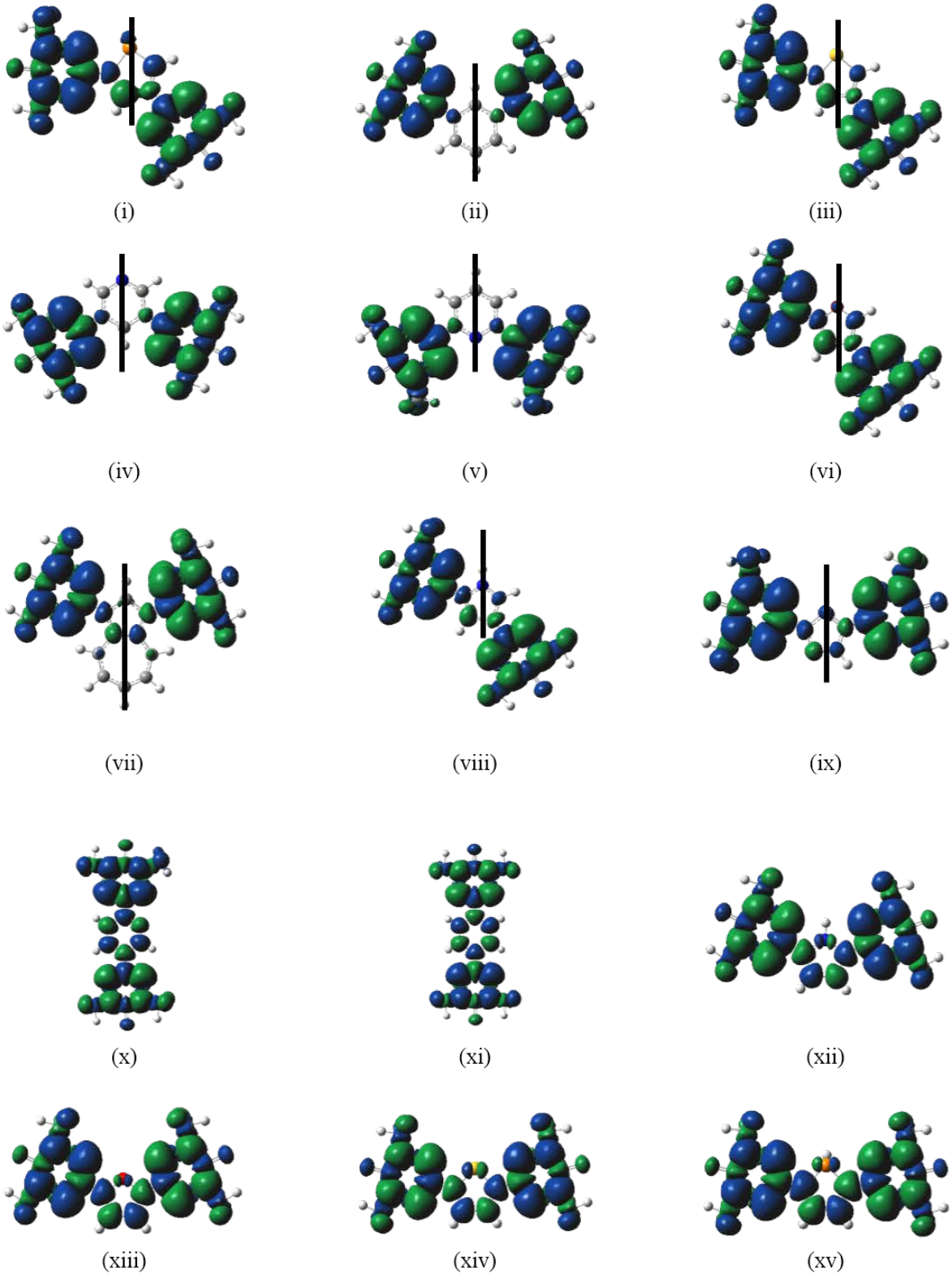

Figure S13. Plots of spin density distributions in the BS states of VER group diradicals. Blue and green colors represent α- and β-spin, respectively. Black solid line indicates the mismatching of the spin polarization.

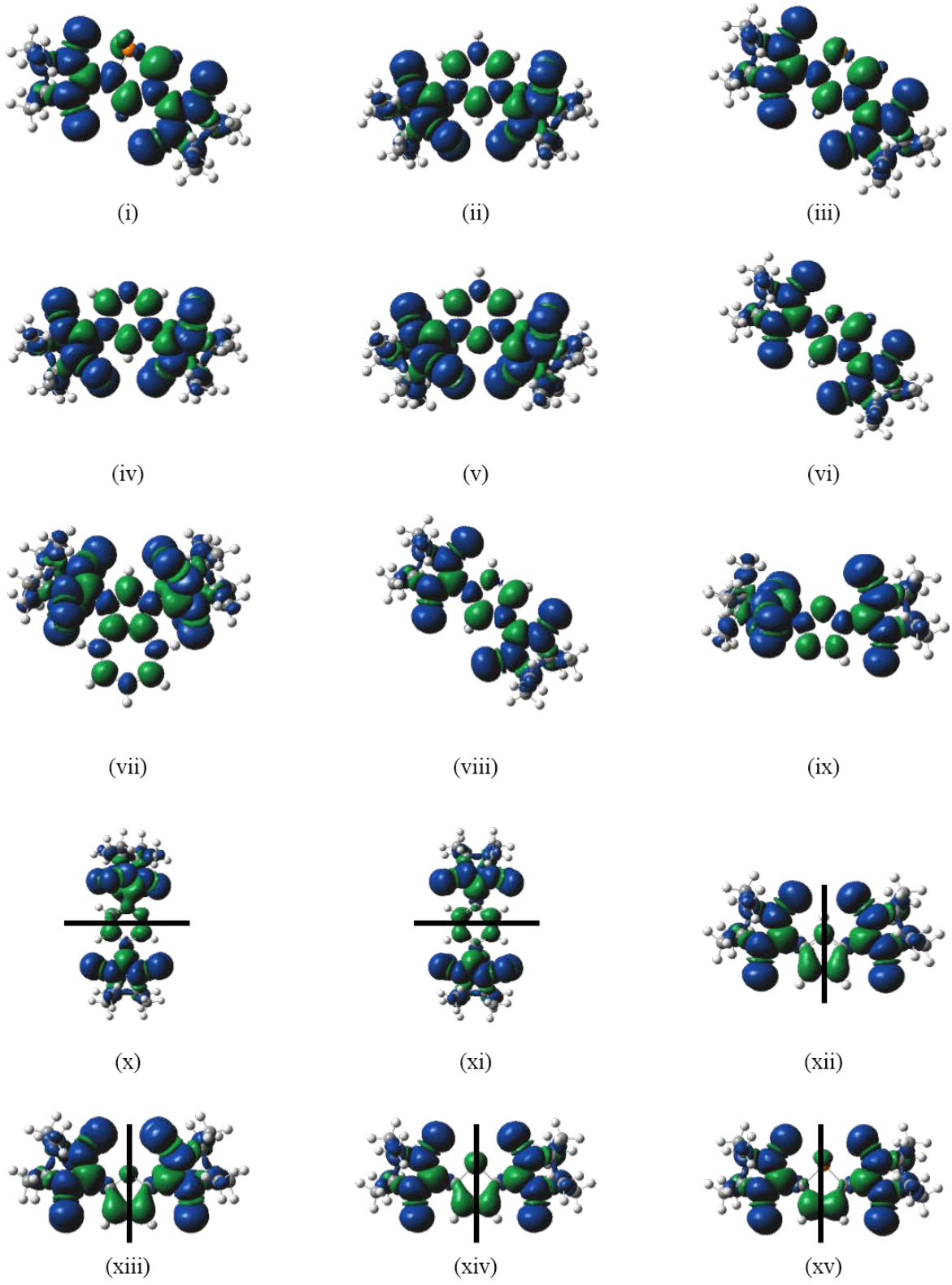

Figure S14. Plots of spin density distributions in the triplet states of NN group diradicals. Blue and green colors represent α- and β-spin, respectively. Black solid line indicates the mismatching of the spin polarization.

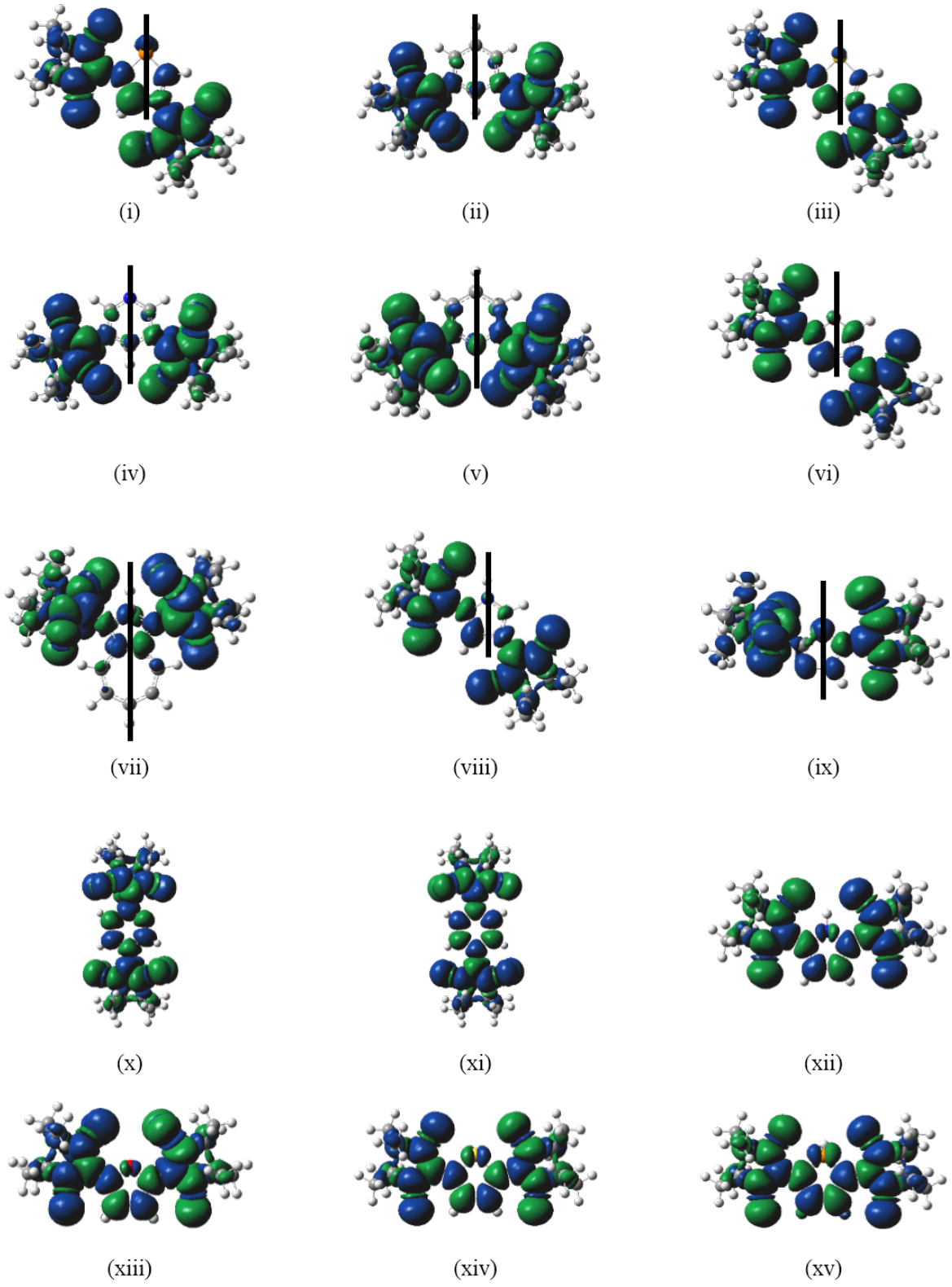

Figure S15. Plots of spin density distributions in the BS states of NN group diradicals. Blue and green colors represent α- and β-spin, respectively. Black solid line indicates the mismatching of the spin polarization.