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Supplementary Information:

Strain manipulation of the local spin flip on Ni@B_{80} endohedral fullerene

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SI. Cartesian coordinates of the equilibrium Ni@B_{80} structure and the distorted structures

In Tables S1-S3, the Cartesian coordinates of the distorted Ni@B_{80} structures under D1-, D2-, and D3-types strains are listed respectively, together with that of the equilibrium structure. Thereinto, the first columns of each table list the labels of B atoms (1-80) and the Ni (81) atom. And the coordinates of equilibrium structure, distorted structure under 0.5%, and 1.0% strains are listed successively. The coordinates of Ni atoms in each structure are highlighted with **bold blue**. Meanwhile, the coordinates of the atoms on which the strains applied are highlighted with ***bold italic red***. In structures under D1-type strains, the strains are applied on atoms with labels 65 and 75, respectively. The distance between atoms 65 and 75 in equilibrium is about 8.0408 Å, and this value is enhanced to 8.0810 Å and 8.1212 Å respectively under 0.5% and 1.0% D1-type strains. This indicates the different magnitudes of D1-type strains are simulated through adjusting the coordinates of corresponding atoms, and then freezing the distances between them. Similarly, the corresponding atoms for structures under D2-type strains are atoms 68 and 78. And the distances between them are 8.2410 Å, 8.2551 Å, and 8.2961 Å in equilibrium structure, distorted structures under 0.5% strain, and under 1.0% strains, respectively. It indicates that the simulation of the D2-types strains is achieved. In addition, it is notable that in scenarios under D3-type strains there is 10 corresponding atoms in total on which strains are applied. Here the corresponding relationship is that atoms 48-14, 58-24, 54-28, 44-18 and 39-9 respectively, and the distances between each pair of atoms are adjusted to simulate the D3-type strains. As a result, the distance between pentagons 48-58-54-44-39 and 14-24-28-18-9 are adjusted in proportion to the 0.5% and 1.0% tensile strains, and then the simulation of D3-type strains is achieved.

Table S1 The Cartesian coordinates of the optimized equilibrium Ni@B_{80} structure and the distorted structures with D1-type tensile strains are applied, the first column denotes the labels of the B (1-80) and Ni (81) atoms.

Label	Equilibrium Structure			0.5% D1-type Strain			1.0% D1-type Strain		
	X	Y	Z	X	Y	Z	X	Y	Z
1	4.0722	-0.2171	0.0000	0.1884	4.0257	0.6346	0.1449	3.9790	0.9075
2	3.7293	-1.2431	1.4005	1.2207	3.4750	1.9639	1.1871	3.3480	2.1955
3	3.0956	-2.6601	0.9494	2.6408	2.9292	1.4173	2.6115	2.8563	1.6092
4	3.0956	-2.6601	-0.9494	2.6355	3.2267	-0.4577	2.5983	3.2823	-0.2413
5	3.7293	-1.2431	-1.4005	1.2126	3.9126	-0.8012	1.1659	3.9730	-0.5326
6	3.9092	1.3395	0.0000	-1.3668	3.8533	0.6115	-1.4088	3.7908	0.8756
7	3.1373	-0.4519	2.6885	0.4371	2.6821	3.1439	0.4153	2.4660	3.3182
8	1.8551	-3.3205	1.7933	3.3114	1.5762	2.0555	3.2979	1.4695	2.1514
9	1.8551	-3.3205	-1.7933	3.3013	2.1376	-1.4859	3.2719	2.2727	-1.3422
10	3.1373	-0.4519	-2.6885	0.4215	3.5234	-2.1652	0.3745	3.6695	-1.9193
11	3.3575	2.2636	1.4081	-2.2831	3.0809	1.9187	-2.3123	2.9186	2.1300
12	1.9000	-1.0742	3.5238	1.0699	1.3335	3.7747	1.0639	1.0837	3.8529
13	0.4835	-3.9384	0.8109	3.9356	0.3791	0.8702	3.9308	0.3627	0.8870
14	1.2218	-2.4679	-3.0489	2.4499	1.7024	-2.8232	2.4221	1.9207	-2.7037
15	2.9499	1.3671	-2.7092	-1.3934	3.3288	-2.2122	-1.4342	3.4588	-1.9764
16	2.9499	1.3671	2.7092	-1.3934	2.4807	3.1380	-1.3986	2.2453	3.3022
17	1.2218	-2.4679	3.0489	2.4666	0.7478	3.1975	2.4649	0.5547	3.2346
18	0.4835	-3.9384	-0.8109	3.9311	0.6329	-0.7310	3.9190	0.7256	-0.6927
19	1.9000	-1.0742	-3.5238	1.0509	2.4372	-3.1852	1.0155	2.6644	-3.0135
20	3.3575	2.2636	-1.4081	-2.2899	3.5210	-0.8614	-2.3304	3.5473	-0.6131
21	2.3660	3.4491	0.9142	-3.4628	2.1697	1.2779	-3.4829	2.0395	1.4294
22	0.7891	0.2135	4.0090	-0.2081	0.1507	4.0824	-0.1987	-0.1325	4.0819
23	-0.9428	-3.5246	1.7418	3.5349	-1.1791	1.5675	3.5512	-1.2452	1.4768
24	-0.5878	-2.6373	-3.0629	2.6322	-0.0822	-3.1202	2.6232	0.1614	-3.1215
25	1.6157	1.7157	-3.5717	-1.7361	2.1437	-3.2725	-1.7690	2.3461	-3.1156
26	1.6157	1.7157	3.5717	-1.7167	1.0251	3.7816	-1.7169	0.7446	3.8441
27	-0.5878	-2.6373	3.0629	2.6487	-1.0414	2.9286	2.6671	-1.2112	2.8447
28	-0.9428	-3.5246	-1.7418	3.5256	-0.6334	-1.8728	3.5261	-0.4642	-1.9170
29	0.7891	0.2135	-4.0090	-0.2297	1.4058	-3.8362	-0.2554	1.6646	-3.7320
30	2.3660	3.4491	-0.9142	-3.4675	2.4562	-0.5276	-3.4960	2.4493	-0.3523
31	-4.0665	0.4579	0.0000	-0.4293	-4.0220	-0.6335	-0.3858	-3.9782	-0.9061
32	-3.7136	1.4806	-1.4050	-1.4581	-3.4605	-1.9657	-1.4241	-3.3362	-2.1963
33	-3.0110	2.8587	-0.9162	-2.8395	-2.8520	-1.3710	-2.8102	-2.7843	-1.5578
34	-3.0110	2.8587	0.9162	-2.8342	-3.1390	0.4385	-2.7974	-3.1949	0.2280
35	-3.7136	1.4806	1.4050	-1.4499	-3.8994	0.8085	-1.4030	-3.9625	0.5411
36	-3.9163	-1.0994	0.0000	1.1268	-3.8623	-0.6125	1.1690	-3.8024	-0.8770
37	-3.1336	0.6950	-2.7106	-0.6800	-2.6768	-3.1652	-0.6577	-2.4621	-3.3392
38	-1.7743	3.4976	-1.7616	-3.4889	-1.5024	-2.0114	-3.4755	-1.4005	-2.1023
39	-1.7743	3.4976	1.7616	-3.4790	-2.0538	1.4676	-3.4504	-2.1890	1.3297
40	-3.1336	0.6950	2.7106	-0.6645	-3.5249	2.1881	-0.6174	-3.6751	1.9425
41	-3.3497	-2.0145	-1.4286	2.0340	-3.0718	-1.9375	2.0634	-2.9106	-2.1479
42	-1.9013	1.3288	-3.5683	-1.3243	-1.3294	-3.8184	-1.3180	-1.0794	-3.8960

43	-0.4238	4.0940	-0.7819	-4.0915	-0.3258	-0.8321	-4.0869	-0.3138	-0.8449
44	-1.2036	2.7156	3.0612	-2.6977	-1.6879	2.8389	-2.6698	-1.9095	2.7208
45	-2.9507	-1.1329	2.7292	1.1593	-3.3340	2.2322	1.2003	-3.4679	1.9962
46	-2.9507	-1.1329	-2.7292	1.1458	-2.4798	-3.1577	1.1650	-2.2453	-3.3218
47	-1.2036	2.7156	-3.0612	-2.7143	-0.7295	-3.2067	-2.7125	-0.5385	-3.2427
48	-0.4238	4.0940	0.7819	-4.0874	-0.5705	0.7122	-4.0760	-0.6634	0.6787
49	-1.9013	1.3288	3.5683	-1.3051	-2.4469	3.2292	-1.2693	-2.6793	3.0568
50	-3.3497	-2.0145	1.4286	2.0408	-3.5182	0.8832	2.0810	-3.5484	0.6352
51	-2.3246	-3.1689	-0.8834	3.1824	-2.1354	-1.2407	3.2024	-2.0106	-1.3895
52	-0.7710	0.0461	-4.0184	-0.0513	-0.1330	-4.0885	-0.0604	0.1479	-4.0864
53	1.0197	3.8028	-1.7614	-3.8130	1.2557	-1.5740	-3.8292	1.3186	-1.4772
54	0.6320	2.9165	3.0613	-2.9115	0.1244	3.1259	-2.9028	-0.1223	3.1309
55	-1.5968	-1.4579	3.5512	1.4783	-2.1235	3.2557	1.5112	-2.3278	3.1005
56	-1.5968	-1.4579	-3.5512	1.4590	-1.0112	-3.7582	1.4595	-0.7349	-3.8196
57	0.6320	2.9165	-3.0613	-2.9278	1.0828	-2.9196	-2.9460	1.2488	-2.8325
58	1.0197	3.8028	1.7614	-3.8040	0.7042	1.9043	-3.8050	0.5301	1.9539
59	-0.7710	0.0461	4.0184	-0.0297	-1.3910	3.8486	-0.0037	-1.6532	3.7453
60	-2.3246	-3.1689	0.8834	3.1870	-2.4124	0.5042	3.2151	-2.4070	0.3325
61	3.7491	0.5389	1.4142	-0.5613	3.4792	1.9824	-0.5948	3.3298	2.2159
62	2.0451	-1.5344	1.8266	1.5244	1.7460	2.1203	1.5094	1.6149	2.2301
63	1.9162	-3.4150	0.0000	3.4004	1.9175	0.2941	3.3777	1.9315	0.4174
64	2.0451	-1.5344	-1.8266	1.5130	2.3184	-1.4860	1.4806	2.4344	-1.3261
65	***3.7491***	***0.5389***	***-1.4142***	***-0.5711***	***3.9401***	***-0.8127***	***-0.6197***	***3.9999***	***-0.5403***
66	1.6699	2.1801	1.8775	-2.1861	1.3409	2.1178	-2.1945	1.1698	2.2075
67	0.1571	-1.2624	3.8176	1.2716	-0.4332	3.7928	1.2864	-0.6788	3.7504
68	-0.9686	-3.9594	0.0000	3.9651	-0.9281	-0.1572	3.9733	-0.8704	-0.2264
69	0.1571	-1.2624	-3.8176	1.2510	0.7621	-3.7475	1.2326	1.0325	-3.6876
70	1.6699	2.1801	-1.8775	-2.1949	1.9295	-1.5893	-2.2181	2.0123	-1.4489
71	-3.7306	-0.2873	-1.4186	0.3097	-3.4618	-1.9836	0.3433	-3.3146	-2.2156
72	-2.0520	1.7657	-1.8762	-1.7556	-1.7470	-2.1703	-1.7404	-1.6153	-2.2805
73	-1.8075	3.6845	0.0000	-3.6705	-1.8123	-0.2767	-3.6488	-1.8304	-0.3928
74	-2.0520	1.7657	1.8762	-1.7441	-2.3347	1.5344	-1.7110	-2.4557	1.3733
75	***-3.7306***	***-0.2873***	***1.4186***	***0.3194***	***-3.9240***	***0.8204***	***0.3679***	***-3.9870***	***0.5493***
76	-1.6149	-1.8551	-1.8380	1.8609	-1.2948	-2.0699	1.8689	-1.1303	-2.1559
77	-0.1438	1.5506	-3.7376	-1.5589	0.4319	-3.7110	-1.5726	0.6689	-3.6688
78	1.0053	4.0139	0.0000	-4.0198	0.9642	0.1631	-4.0281	0.9061	0.2353
79	-0.1438	1.5506	3.7376	-1.5388	-0.7384	3.6707	-1.5200	-1.0064	3.6131
80	-1.6149	-1.8551	1.8380	1.8698	-1.8710	1.5595	1.8925	-1.9557	1.4236
81	**-0.2213**	**-1.7326**	**0.0000**	**1.7326**	**-0.2070**	**-0.0370**	**1.7324**	**-0.1866**	**-0.0528**

Table S2 The Cartesian coordinates of the optimized equilibrium Ni@B_{80} structure and the distorted structures with D2-type tensile strains are applied, the first column denotes the labels of the B (1-80) and Ni (81) atoms.

Label	Equilibrium Structure			0.5% D2-type Strain			1.0% D2-type Strain		
	X	Y	Z	X	Y	Z	X	Y	Z
1	4.0722	-0.2171	0.0000	4.0743	-0.1567	0.0000	-0.1039	4.0752	0.0000
2	3.7293	-1.2431	1.4005	3.7451	-1.1888	1.3992	-1.1411	3.7582	-1.3982
3	3.0956	-2.6601	0.9494	3.1333	-2.6161	0.9489	-2.5769	3.1653	-0.9483
4	3.0956	-2.6601	-0.9494	3.1333	-2.6161	-0.9489	-2.5769	3.1653	0.9483
5	3.7293	-1.2431	-1.4005	3.7451	-1.1888	-1.3992	-1.1411	3.7582	1.3982
6	3.9092	1.3395	0.0000	3.8910	1.3981	0.0000	1.4493	3.8748	0.0000
7	3.1373	-0.4519	2.6885	3.1420	-0.4061	2.6865	-0.3659	3.1456	-2.6847
8	1.8551	-3.3205	1.7933	1.9029	-3.2946	1.7929	-3.2716	1.9439	-1.7923
9	1.8551	-3.3205	-1.7933	1.9029	-3.2946	-1.7929	-3.2716	1.9439	1.7923
10	3.1373	-0.4519	-2.6885	3.1420	-0.4061	-2.6865	-0.3659	3.1456	2.6847
11	3.3575	2.2636	1.4081	3.3254	2.3149	1.4079	2.3598	3.2970	-1.4079
12	1.9000	-1.0742	3.5238	1.9142	-1.0460	3.5220	-1.0213	1.9262	-3.5201
13	0.4835	-3.9384	0.8109	0.5390	-3.9350	0.8113	-3.9319	0.5874	-0.8117
14	1.2218	-2.4679	-3.0489	1.2565	-2.4499	-3.0473	-2.4341	1.2867	3.0462
15	2.9499	1.3671	-2.7092	2.9293	1.4112	-2.7077	1.4500	2.9108	2.7062
16	2.9499	1.3671	2.7092	2.9293	1.4112	2.7077	1.4500	2.9108	-2.7062
17	1.2218	-2.4679	3.0489	1.2565	-2.4499	3.0473	-2.4341	1.2867	-3.0462
18	0.4835	-3.9384	-0.8109	0.5390	-3.9350	-0.8113	-3.9319	0.5874	0.8117
19	1.9000	-1.0742	-3.5238	1.9142	-1.0460	-3.5220	-1.0213	1.9262	3.5201
20	3.3575	2.2636	-1.4081	3.3254	2.3149	-1.4079	2.3598	3.2970	1.4079
21	2.3660	3.4491	0.9142	2.3158	3.4845	0.9117	3.5152	2.2714	-0.9091
22	0.7891	0.2135	4.0090	0.7859	0.2255	4.0066	0.2359	0.7829	-4.0041
23	-0.9428	-3.5246	1.7418	-0.8925	-3.5416	1.7415	-3.5556	-0.8481	-1.7411
24	-0.5878	-2.6373	-3.0629	-0.5497	-2.6467	-3.0616	-2.6547	-0.5164	3.0608
25	1.6157	1.7157	-3.5717	1.5909	1.7406	-3.5698	1.7621	1.5690	3.5681
26	1.6157	1.7157	3.5717	1.5909	1.7406	3.5698	1.7621	1.5690	-3.5681
27	-0.5878	-2.6373	3.0629	-0.5497	-2.6467	3.0616	-2.6547	-0.5164	-3.0608
28	-0.9428	-3.5246	-1.7418	-0.8925	-3.5416	-1.7415	-3.5556	-0.8481	1.7411
29	0.7891	0.2135	-4.0090	0.7859	0.2255	-4.0066	0.2359	0.7829	4.0041
30	2.3660	3.4491	-0.9142	2.3158	3.4845	-0.9117	3.5152	2.2714	0.9091
31	-4.0665	0.4579	0.0000	-4.0716	0.3991	0.0000	0.3475	-4.0744	0.0000
32	-3.7136	1.4806	-1.4050	-3.7332	1.4274	-1.4045	1.3807	-3.7490	1.4037
33	-3.0110	2.8587	-0.9162	-3.0507	2.8162	-0.9162	2.7793	-3.0853	0.9163
34	-3.0110	2.8587	0.9162	-3.0507	2.8162	0.9162	2.7793	-3.0853	-0.9163
35	-3.7136	1.4806	1.4050	-3.7332	1.4274	1.4045	1.3807	-3.7490	-1.4037
36	-3.9163	-1.0994	0.0000	-3.9005	-1.1564	0.0000	-1.2062	-3.8851	0.0000
37	-3.1336	0.6950	-2.7106	-3.1424	0.6506	-2.7098	0.6115	-3.1491	2.7086
38	-1.7743	3.4976	-1.7616	-1.8246	3.4750	-1.7618	3.4551	-1.8686	1.7620
39	-1.7743	3.4976	1.7616	-1.8246	3.4750	1.7618	3.4551	-1.8686	-1.7620
40	-3.1336	0.6950	2.7106	-3.1424	0.6506	2.7098	0.6115	-3.1491	-2.7086
41	-3.3497	-2.0145	-1.4286	-3.3204	-2.0642	-1.4285	-2.1075	-3.2941	1.4283
42	-1.9013	1.3288	-3.5683	-1.9191	1.3021	-3.5671	1.2788	-1.9342	3.5658

43	-0.4238	4.0940	-0.7819	-0.4812	4.0918	-0.7820	4.0893	-0.5310	0.7821
44	-1.2036	2.7156	3.0612	-1.2419	2.6999	3.0610	2.6858	-1.2753	-3.0606
45	-2.9507	-1.1329	2.7292	-2.9334	-1.1759	2.7281	-1.2134	-2.9176	-2.7269
46	-2.9507	-1.1329	-2.7292	-2.9334	-1.1759	-2.7281	-1.2134	-2.9176	2.7269
47	-1.2036	2.7156	-3.0612	-1.2419	2.6999	-3.0610	2.6858	-1.2753	3.0606
48	-0.4238	4.0940	0.7819	-0.4812	4.0918	0.7820	4.0893	-0.5310	-0.7821
49	-1.9013	1.3288	3.5683	-1.9191	1.3021	3.5671	1.2788	-1.9342	-3.5658
50	-3.3497	-2.0145	1.4286	-3.3204	-2.0642	1.4285	-2.1075	-3.2941	-1.4283
51	-2.3246	-3.1689	-0.8834	-2.2796	-3.2055	-0.8829	-3.2374	-2.2399	0.8823
52	-0.7710	0.0461	-4.0184	-0.7713	0.0353	-4.0155	0.0262	-0.7714	4.0131
53	1.0197	3.8028	-1.7614	0.9635	3.8181	-1.7596	3.8310	0.9141	1.7579
54	0.6320	2.9165	3.0613	0.5904	2.9277	3.0609	2.9371	0.5540	-3.0605
55	-1.5968	-1.4579	3.5512	-1.5747	-1.4810	3.5492	-1.5010	-1.5553	-3.5474
56	-1.5968	-1.4579	-3.5512	-1.5747	-1.4810	-3.5492	-1.5010	-1.5553	3.5474
57	0.6320	2.9165	-3.0613	0.5904	2.9277	-3.0609	2.9371	0.5540	3.0605
58	1.0197	3.8028	1.7614	0.9635	3.8181	1.7596	3.8310	0.9141	-1.7579
59	-0.7710	0.0461	4.0184	-0.7713	0.0353	4.0155	0.0262	-0.7714	-4.0131
60	-2.3246	-3.1689	0.8834	-2.2796	-3.2055	0.8829	-3.2374	-2.2399	-0.8823
61	3.7491	0.5389	1.4142	3.7405	0.5950	1.4137	0.6439	3.7318	-1.4131
62	2.0451	-1.5344	1.8266	2.0648	-1.5048	1.8241	-1.4791	2.0821	-1.8223
63	1.9162	-3.4150	0.0000	1.9637	-3.3880	0.0000	-3.3643	2.0054	0.0000
64	2.0451	-1.5344	-1.8266	2.0648	-1.5048	-1.8241	-1.4791	2.0821	1.8223
65	3.7491	0.5389	-1.4142	3.7405	0.5950	-1.4137	0.6439	3.7318	1.4131
66	1.6699	2.1801	1.8775	1.6381	2.2072	1.8746	2.2308	1.6098	-1.8716
67	0.1571	-1.2624	3.8176	0.1751	-1.2603	3.8164	-1.2579	0.1905	-3.8147
68	***-0.9686***	***-3.9594***	***0.0000***	***-0.9135***	***-3.9880***	***0.0000***	***-4.0146***	***-0.8653***	***0.0000***
69	0.1571	-1.2624	-3.8176	0.1751	-1.2603	-3.8164	-1.2579	0.1905	3.8147
70	1.6699	2.1801	-1.8775	1.6381	2.2072	-1.8746	2.2308	1.6098	1.8716
71	-3.7306	-0.2873	-1.4186	-3.7231	-0.3417	-1.4179	-0.3891	-3.7168	1.4174
72	-2.0520	1.7657	-1.8762	-2.0762	1.7382	-1.8759	1.7141	-2.0969	1.8754
73	-1.8075	3.6845	0.0000	-1.8588	3.6577	0.0000	3.6333	-1.9030	0.0000
74	-2.0520	1.7657	1.8762	-2.0762	1.7382	1.8759	1.7141	-2.0969	-1.8754
75	-3.7306	-0.2873	1.4186	-3.7231	-0.3417	1.4179	-0.3891	-3.7168	-1.4174
76	-1.6149	-1.8551	-1.8380	-1.5876	-1.8810	-1.8360	-1.9035	-1.5634	1.8339
77	-0.1438	1.5506	-3.7376	-0.1654	1.5489	-3.7356	1.5476	-0.1837	3.7338
78	***1.0053***	***4.0139***	***0.0000***	***0.9505***	***4.0539***	***0.0000***	***4.0911***	***0.9020***	***0.0000***
79	-0.1438	1.5506	3.7376	-0.1654	1.5489	3.7356	1.5476	-0.1837	-3.7338
80	-1.6149	-1.8551	1.8380	-1.5876	-1.8810	1.8360	-1.9035	-1.5634	-1.8339
81	**-0.2213**	**-1.7326**	**0.0000**	**-0.1975**	**-1.7432**	**0.0000**	**-1.7530**	**-0.1769**	**0.0000**

Table S3 The Cartesian coordinates of the optimized equilibrium $Ni@B_{80}$ structure and the distorted structures with D3-type tensile strains are applied, the first column denotes the labels of the B (1-80) and Ni (81) atoms.

Label	Equilibrium Structure			0.5% D3-type Strain			1.0% D3-type Strain		
	X	Y	Z	X	Y	Z	X	Y	Z
1	4.0722	-0.2171	0.0000	0.3035	1.9399	3.5666	0.4613	1.6090	3.6996
2	3.7293	-1.2431	1.4005	1.1860	0.4627	3.9690	1.2535	0.0539	3.9598
3	3.0956	-2.6601	0.9494	2.6261	0.4205	3.2348	2.6751	-0.0088	3.1922
4	3.0956	-2.6601	-0.9494	2.8103	2.0766	2.3193	2.9662	1.7067	2.4292
5	3.7293	-1.2431	-1.4005	1.4516	2.9004	2.6194	1.6592	2.5779	2.8232
6	3.9092	1.3395	0.0000	-1.2498	2.0037	3.3830	-1.0874	1.7809	3.5479
7	3.1373	-0.4519	2.6885	0.2636	-0.8716	4.0499	0.2505	-1.2245	3.9466
8	1.8551	-3.3205	1.7933	3.1767	-0.9726	2.5731	3.1308	-1.3738	2.4088
9	***1.8551***	***-3.3205***	***-1.7933***	***3.5290***	***2.1591***	***0.8450***	***3.6890***	***1.8835***	***0.9650***
10	3.1373	-0.4519	-2.6885	0.7726	3.8098	1.4600	1.0272	3.6233	1.7545
11	3.3575	2.2636	1.4081	-2.3164	0.5963	3.5542	-2.2400	0.4311	3.6168
12	1.9000	-1.0742	3.5238	0.7768	-2.2522	3.3858	0.6678	-2.5698	3.1609
13	0.4835	-3.9384	0.8109	3.8615	-0.8308	0.9149	3.8137	-1.1308	0.7674
14	***1.2218***	***-2.4679***	***-3.0489***	***2.7840***	***3.0263***	***-0.3371***	***2.9875***	***2.8873***	***-0.1313***
15	2.9499	1.3671	-2.7092	-1.0426	3.9025	1.2400	-0.7837	3.8372	1.5724
16	2.9499	1.3671	2.7092	-1.5552	-0.8154	3.8481	-1.5664	-1.0434	3.7801
17	1.2218	-2.4679	3.0489	2.1948	-2.2925	2.5994	2.0732	-2.6324	2.3520
18	***0.4835***	***-3.9384***	***-0.8109***	***4.0228***	***0.5850***	***0.1333***	***4.0671***	***0.3343***	***0.1115***
19	1.9000	-1.0742	-3.5238	1.4476	3.8877	-0.0077	1.7001	3.7914	0.2904
20	3.3575	2.2636	-1.4081	-2.0493	3.0483	2.2001	-1.8330	2.9680	2.4732
21	2.3660	3.4491	0.9142	-3.4762	0.6557	2.4206	-3.4098	0.6552	2.5141
22	0.7891	0.2135	4.0090	-0.5767	-3.0913	2.6147	-0.7449	-3.2577	2.3475
23	-0.9428	-3.5246	1.7418	3.3264	-2.2935	0.1057	3.2014	-2.5310	-0.1530
24	***-0.5878***	***-2.6373***	***-3.0629***	***2.9152***	***2.1506***	***-1.9245***	***3.0393***	***2.1346***	***-1.7900***
25	1.6157	1.7157	-3.5717	-1.3375	4.0436	-0.3517	-1.0848	4.1287	0.0050
26	1.6157	1.7157	3.5717	-2.0164	-2.1788	3.0892	-2.1241	-2.3085	2.9199
27	-0.5878	-2.6373	3.0629	2.3236	-3.1894	1.0297	2.1393	-3.4050	0.7210
28	***-0.9428***	***-3.5246***	***-1.7418***	***3.6674***	***0.7466***	***-1.5754***	***3.6948***	***0.6597***	***-1.5558***
29	0.7891	0.2135	-4.0090	0.1834	3.8898	-1.2453	0.4166	3.9634	-0.9194
30	2.3660	3.4491	-0.9142	-3.2986	2.2501	1.5398	-3.1373	2.3068	1.7690
31	-4.0665	0.4579	0.0000	-0.5447	-1.9163	-3.5691	-0.7014	-1.5702	-3.7034
32	-3.7136	1.4806	-1.4050	-1.4231	-0.4308	-3.9646	-1.4878	-0.0068	-3.9608
33	-3.0110	2.8587	-0.9162	-2.8263	-0.3918	-3.1504	-2.8795	0.0483	-3.1284
34	-3.0110	2.8587	0.9162	-3.0040	-1.9894	-2.2672	-3.1533	-1.6044	-2.3812
35	-3.7136	1.4806	1.4050	-1.6895	-2.8768	-2.6107	-1.8976	-2.5382	-2.8178
36	-3.9163	-1.0994	0.0000	1.0090	-1.9865	-3.3969	0.8471	-1.7462	-3.5558
37	-3.1336	0.6950	-2.7106	-0.5056	0.9140	-4.0645	-0.4874	1.2821	-3.9595
38	-1.7743	3.4976	-1.7616	-3.3556	0.9987	-2.4912	-3.3127	1.4078	-2.3465
39	***-1.7743***	***3.4976***	***1.7616***	***-3.7024***	***-2.0767***	***-0.7950***	***-3.8507***	***-1.7761***	***-0.9129***
40	-3.1336	0.6950	2.7106	-1.0180	-3.8067	-1.4535	-1.2755	-3.6000	-1.7509
41	-3.3497	-2.0145	-1.4286	2.0686	-0.5533	-3.5642	1.9907	-0.3750	-3.6052
42	-1.9013	1.3288	-3.5683	-1.0286	2.3127	-3.4157	-0.9161	2.6504	-3.1929

43	-0.4238	4.0940	-0.7819	-4.0171	0.8472	-0.8531	-3.9713	1.1562	-0.7184
44	***-1.2036***	***2.7156***	***3.0612***	***-3.0331***	***-3.0060***	***0.3517***	***-3.2301***	***-2.8405***	***0.1399***
45	-2.9507	-1.1329	2.7292	0.8059	-3.8989	-1.2376	0.5427	-3.8123	-1.5683
46	-2.9507	-1.1329	-2.7292	1.3222	0.8529	-3.8648	1.3325	1.0935	-3.7821
47	-1.2036	2.7156	-3.0612	-2.4420	2.3341	-2.5968	-2.3165	2.6885	-2.3530
48	***-0.4238***	***4.0940***	***0.7819***	***-4.1756***	***-0.5171***	***-0.0999***	***-4.2172***	***-0.2557***	***-0.0801***
49	-1.9013	1.3288	3.5683	-1.7064	-3.9050	0.0210	-1.9582	-3.7833	-0.2825
50	-3.3497	-2.0145	1.4286	1.7982	-3.0401	-2.1901	1.5857	-2.9444	-2.4646
51	-2.3246	-3.1689	-0.8834	3.1931	-0.6378	-2.3764	3.1256	-0.6247	-2.4558
52	-0.7710	0.0461	-4.0184	0.3175	3.1310	-2.6107	0.4896	3.3183	-2.3444
53	1.0197	3.8028	-1.7614	-3.5988	2.3707	-0.0559	-3.4453	2.5795	0.1943
54	***0.6320***	***2.9165***	***3.0613***	***-3.1938***	***-2.1026***	***1.9542***	***-3.3153***	***-2.0690***	***1.8173***
55	-1.5968	-1.4579	3.5512	1.0810	-3.9922	0.3513	0.8287	-4.0595	-0.0005
56	-1.5968	-1.4579	-3.5512	1.7565	2.1926	-3.0700	1.8660	2.3383	-2.9009
57	0.6320	2.9165	-3.0613	-2.6021	3.2350	-0.9982	-2.4034	3.4560	-0.6875
58	***1.0197***	***3.8028***	***1.7614***	***-3.9463***	***-0.7015***	***1.6440***	***-3.9804***	***-0.6020***	***1.6372***
59	-0.7710	0.0461	4.0184	-0.4441	-3.8657	1.2578	-0.6759	-3.9188	0.9290
60	-2.3246	-3.1689	0.8834	3.0230	-2.1793	-1.5261	2.8818	-2.2506	-1.7524
61	3.7491	0.5389	1.4142	-0.5900	0.6223	3.9453	-0.5092	0.3198	3.9797
62	2.0451	-1.5344	1.8266	1.3986	-0.7459	2.7067	1.3726	-1.0544	2.5963
63	1.9162	-3.4150	0.0000	3.4465	0.6122	1.7629	3.5212	0.2601	1.7391
64	2.0451	-1.5344	-1.8266	1.7525	2.4357	0.9457	1.9330	2.2590	1.1136
65	3.7491	0.5389	-1.4142	-0.3220	3.0855	2.5824	-0.1016	2.8699	2.8270
66	1.6699	2.1801	1.8775	-2.3232	-0.6374	2.3081	-2.3457	-0.6934	2.2747
67	0.1571	-1.2624	3.8176	0.8973	-3.3656	2.0089	0.7034	-3.5792	1.7021
68	-0.9686	-3.9594	0.0000	3.9263	-0.8277	-0.7459	3.8384	-1.0067	-0.8771
69	0.1571	-1.2624	-3.8176	1.6244	3.2878	-1.6695	1.8180	3.3109	-1.4119
70	1.6699	2.1801	-1.8775	-1.9586	2.6353	0.4994	-1.7860	2.6937	0.7437
71	-3.7306	-0.2873	-1.4186	0.3388	-0.5858	-3.9372	0.2611	-0.2710	-3.9718
72	-2.0520	1.7657	-1.8762	-1.6268	0.8056	-2.7433	-1.6032	1.1310	-2.6386
73	-1.8075	3.6845	0.0000	-3.7136	-0.5367	-1.6772	-3.7667	-0.1663	-1.6613
74	-2.0520	1.7657	1.8762	-1.9909	-2.4654	-0.9358	-2.1653	-2.2539	-1.1101
75	-3.7306	-0.2873	1.4186	0.0705	-3.0571	-2.5714	-0.1467	-2.8267	-2.8146
76	-1.6149	-1.8551	-1.8380	1.9943	0.6580	-2.2481	2.0199	0.7306	-2.1983
77	-0.1438	1.5506	-3.7376	-1.1932	3.3262	-1.9661	-0.9963	3.5500	-1.6656
78	1.0053	4.0139	0.0000	-3.9809	0.8483	0.7760	-3.9191	1.0188	0.9030
79	-0.1438	1.5506	3.7376	-1.9062	-3.1847	1.6332	-2.0935	-3.1870	1.3822
80	-1.6149	-1.8551	1.8380	1.6396	-2.5449	-0.4789	1.4877	-2.5997	-0.7155
81	**-0.2213**	**-1.7326**	**0.0000**	**1.7312**	**-0.2585**	**-0.1439**	**1.7074**	**-0.3560**	**-0.1404**

SII. The spin components of the equilibrium structure and the strained structures

During the investigation of the spin-dynamics processes in the $Ni@B_{80}$ structures, the initial and final states are selected based on the requirements of the Λ process. In principle, the initial and final states of a Λ process should be selected to be (or close to) the ground state. At the same time, the expectation values of the spin components for the selected states are better to be concentrated along one direction (*z*-axis in this manuscript) with the influence of the same magnetic field.

Table S4 lists the spin components of the equilibrium structure and the strained structures. Based on the spin components of the structures and the theoretical requirements of Λ process, we select states $|1\rangle$ and $|3\rangle$ as the initial and final states to achieve the spin-flip processes on Ni@B_{80} structures, except for the structures with D1-type strains are applied. When the D1-type strains are applied, states $|4\rangle$ and $|6\rangle$ are selected as the initial and final states, respectively. The different selection for scenarios under D1-type strains is out of considering that the spin component $\langle S_z \rangle$ of state $|3\rangle$ lost its dominate position, which may lead to the difficulty in identifying the spin-flip processes. The spin components $\langle S_z \rangle$ are 0.747 under 0.5% strain and 0.767 under 1.0% strain, while $\langle S_y \rangle$ are up to 0.587 and 0.543 for 0.5% and 1.0% strains, respectively. Therefore, states $|4\rangle$ and $|6\rangle$ are selected for the spin-flip processes of the structures under D1-type strains. Besides, when 0.5% D3-type tensile strain is applied on the Ni@B_{80} structures, the comparisons of $\langle S_y \rangle$ versus $\langle S_z \rangle$ are 0.358 vs. -0.914 and 0.266 vs. 0.877, for the initial and final states, respectively. The obvious differences between the spin components $\langle S_y \rangle$ and $\langle S_z \rangle$ do not influence the identification of the spin-flip processes. Therefore, states $|1\rangle$ and $|3\rangle$ are suitable for the structures under D3-type strains. As for scenarios in the equilibrium structure and the structures under D2-type strains, the values of $\langle S_x \rangle$ and $\langle S_y \rangle$ are zero, and the opposite $\langle S_z \rangle$ values of states $|1\rangle$ and $|3\rangle$ make them almost the perfect initial and final states for the spin-flip processes.

Table S4 The expectation values of spin components of low-lying substates on the equilibrium structure and the strained structures (the initial and final states are highlighted in bold).

Structure		State	$\langle S_x \rangle$	$\langle S_y \rangle$	$\langle S_z \rangle$	$\langle S \rangle$
Equilibrium structure		$\|1\rangle$	0.000	0.000	**-0.451**	0.451
		$\|2\rangle$	0.000	0.000	0.000	0.000
		$\|3\rangle$	0.000	0.000	**0.360**	0.360
D1	0.5%	$\|1\rangle$	-0.002	0.086	-0.996	0.999
		$\|2\rangle$	0.016	-0.672	0.246	0.716
		$\|3\rangle$	-0.014	0.587	0.747	0.950
		$\|4\rangle$	0.008	0.022	**-0.864**	0.864
		$\|5\rangle$	-0.021	-0.055	0.000	0.058
		$\|6\rangle$	0.012	0.031	**0.863**	0.864
	1.0%	$\|1\rangle$	-0.002	0.167	-0.985	0.999
		$\|2\rangle$	0.007	0.710	0.219	0.743
		$\|3\rangle$	-0.006	0.543	0.767	0.940
		$\|4\rangle$	0.016	0.026	**-0.870**	0.870
		$\|5\rangle$	-0.039	-0.061	0.000	0.073
		$\|6\rangle$	0.023	0.035	**0.869**	0.870
D2	0.5%	$\|1\rangle$	0.000	0.000	**-0.910**	0.910
		$\|2\rangle$	0.000	0.000	0.000	0.000
		$\|3\rangle$	0.000	0.000	**0.910**	0.910
	1.0%	$\|1\rangle$	0.000	0.000	**-0.995**	0.995
		$\|2\rangle$	0.000	0.000	0.000	0.000
		$\|3\rangle$	0.000	0.000	**0.995**	0.995
D3	0.5%	$\|1\rangle$	0.014	0.358	**-0.914**	0.982
		$\|2\rangle$	-0.024	-0.624	0.037	0.625

		|3⟩	0.010	0.266	**0.877**	0.916
	1.0%	|1⟩	-0.208	0.076	**-0.673**	0.708
		|2⟩	0.334	-0.122	0.025	0.357
		|3⟩	-0.126	0.047	**0.648**	0.661

SIII. Comparison between different basis sets for the Ni atom

In order to take the effect of using different basis sets for the Ni atom into account, we examine the spin-flip process on the equilibrium structure with triple-zeta Los Alamos basis set adopted for the Ni atom on Ni@B_{80}. As listed in Table S5, among all five low-lying spin states we examined, the calculated spin density distributions on Ni atom do not show significant difference when we change the basis set from LanL2DZ to LanL2TZ. Fig. S1 shows the spin-flip processes achieved on Ni@B_{80} with the double- and triple-zeta Los Alamos basis sets applied on Ni atom. Figs. S1 (a) and (b) in the left panel show the time-resolved occupations of each state with LanL2DZ and LanL2TZ basis set, respectively. The results show that both spin-flip processes can be achieved within 600 fs with less Rabi-like cycles, which demonstrates that the effect of using a triple- versus double-zeta basis set does not significantly influence the time consumption. Figs. S1 (c) and (d) in the right penal show the corresponding time-resolved expectation values of the spin components with two different basis sets. The spin components $\langle S_x \rangle$ and $\langle S_y \rangle$ almost do not contribute to the expectation value of the total spin during the spin-flip processes. In the case using the LanL2DZ basis set, the opposite spin components $\langle S_z \rangle$ of the initial state (-0.451) and the final state (0.360) show the reversal of the spin direction. Similarly, in the case using the LanL2TZ basis set, the same spin direction reversal can also be found with the values -0.585 and 0.701 for the initial state and the final state, respectively. The reversals of the spin direction indicate the spin-flip scenarios are both achieved, and the using of the triple-zeta versus double-zeta does not influence the achievement of the spin-flip process.

Additionally, Table S6 lists the optimized parameters of the adopted laser pulses with using the LanL2DZ and LanL2TZ basis sets, respectively. In the scenario of using LanL2TZ basis set for the Ni atom, the fidelity of the spin-flip process reaches to 99.0%, and the energy of the adopted laser pulse is 2.94 eV. Although the different laser pulses are adopted, both of the ultrafast spin-flip processes achieved with high fidelities. Without compromising the accuracy of main conclusions in our present work, these results partially explain why the effects of changing a larger basis set (LanL2TZ) for the Ni atom are negligible. There is no doubt that a triple-zeta basis set considers more orbitals to address the higher accuracy, but a double-zeta basis set is adopted in our manuscript as a tradeoff between the accuracy and the efficiency.

Table S5 Calculated Mulliken spin density (without SOC) of the lowest five triplet states on Ni atom of Ni@B_{80} equilibrium structure with adopting different basis sets for the Ni atom.

Basis Set	State 1	State 2	State 3	State 4	State 5
LanL2DZ	1.591	0.982	0.983	0.978	0.963
LanL2TZ	1.670	1.110	1.131	1.006	0.951

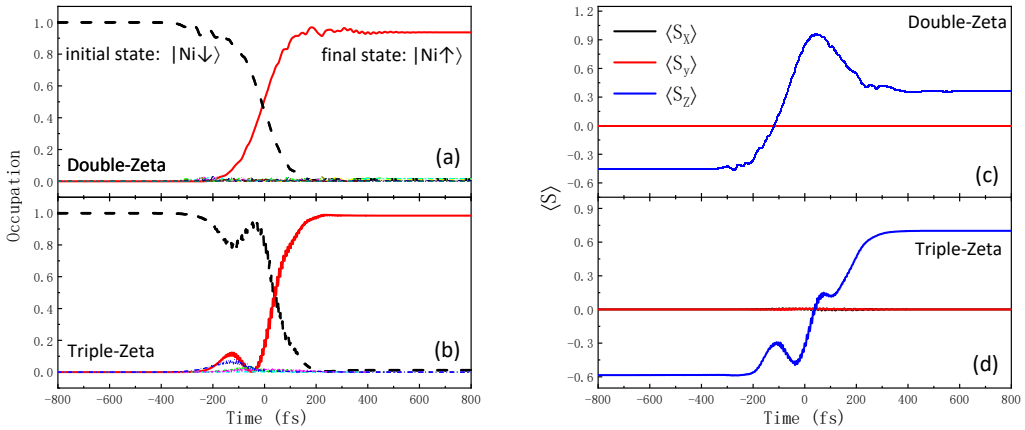

Fig. S1 Spin-flip processes achieved on $Ni@B_{80}$ with adopting different basis sets for the Ni atom. Left panel: Time-resolved occupation of the initial (dashed black line), the final (solid red line), and the intermediate (dash-dotted line) states with adopting (a) LanL2DZ and (b) LanL2TZ basis sets, respectively. Right panel: Time-resolved expectation values of the spin components with adopting (c) LanL2DZ and (d) LanL2TZ basis sets, respectively. The corresponding basis sets are denoted as Double-Zeta (for LanL2DZ) and Triple-Zeta (for LanL2TZ), respectively.

Table S6 Optimized laser parameters of each Λ process in the $Ni@B_{80}$ with adopting different basis sets for the Ni atom. θ and φ represent the angles of incidence in spherical coordinates, and γ is the angle between the linearly polarized light and the optical plane. FWHM is the full width at half maximum of the laser pulse.

Basis Set	Fidelity (%)	$\theta(°)$	$\varphi(°)$	$\gamma(°)$	FWHM (fs)	Energy (eV)
LanL2DZ	96.6	181.58	258.30	226.28	299.71	2.41
LanL2TZ	99.0	196.36	36.60	332.90	300.00	2.94