Pt_3Ni

Pt_3Ni bulk

Source	Spin moment Ni, Pt
This work	0.61, 0.20
[1] VASP PBE	0.67, 0.16
[2] DFTB	0.65, 0.16
[3] DFTB	0.58, 0.10

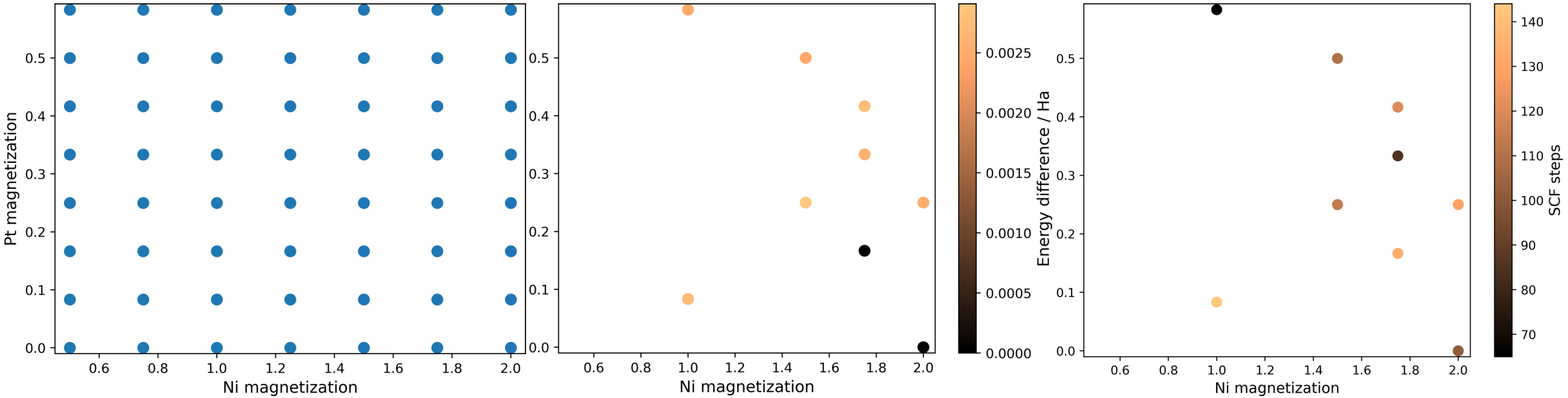
- Pt_3Ni 2x2x2 bulk: 24 Pt atoms, 8 Ni atoms. EPS_SCF=1.0E-6
- Multiple minima exist with different Pt spin moments
- 1D grid search performed for CUTOFF (600), KPOINTS (1x1x1), ADDED_MOS (30), ELECTRONIC_TEMPERATURE (300)
- K-points seem to improve interface SCF convergence but decrease bulk convergence

Multiplicity	Magnetization Ni, Pt	Energy / Ha	Spin moment Ni, Pt
0		-4239.1402	0.00, 0.00
17		-4239.1402	0.00, 0.00
17	2.00, 0.00	-4239.1707	0.63, 0.00
17	2.00, 0.00	-4239.1712	0.63, 0.10
17	2.00, 0.00	-4239.1743	0.61, 0.21

[1] Sharma, B. K., Kwon, O., Odkhuu, D. & Hong, S. C. Electronic structure and magnetism of various surfaces of the catalytic material Pt3Ni: Density-functional study. *Journal of Magnetism and Magnetic Materials* 339, 89–93 (2013).
[2] Paudyal, D., Saha-Dasgupta, T. & Mookerjee, A. Magnetic properties of X–Pt (X = Fe,Co,Ni) alloy systems. *J. Phys.: Condens. Matter* **16**, 2317–2334 (2004).
[3] Singh, P. P. Relativity and magnetism in Ni–Pd and Ni–Pt alloys. *Journal of Magnetism and Magnetic Materials* **261**, 347–352 (2003).

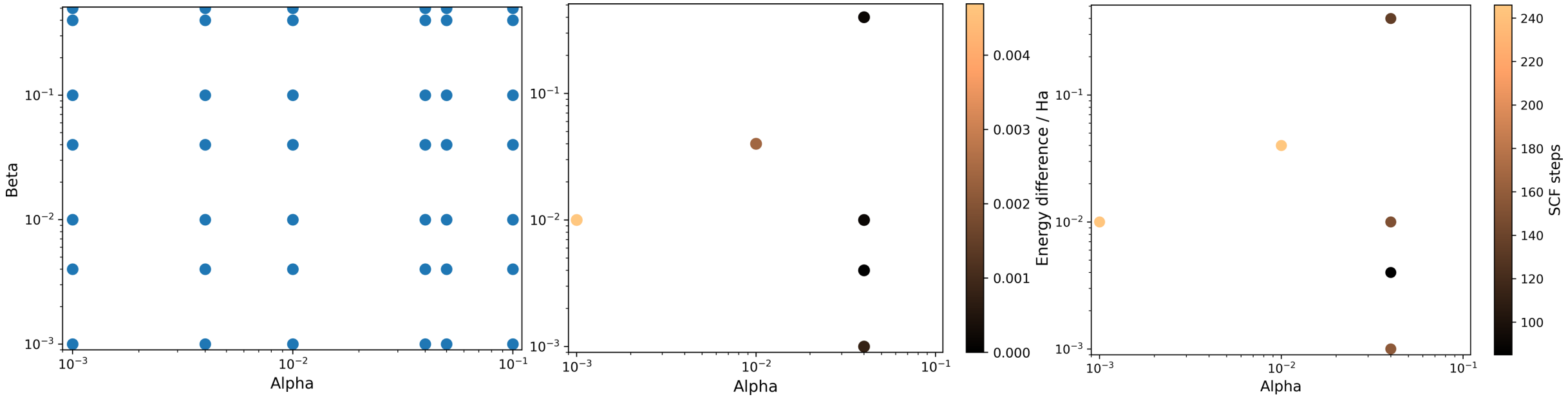
Pt_3Ni bulk optimization

- Pt_3Ni 2x2x2 bulk: 24 Pt atoms, 8 Ni atoms. EPS_SCF=5.0E-7
- 8x8 2D grid search for Pt, Ni magnetisations which sum to odd integer multiplicity
- Minimum at Ni=2.0, Pt=0.0. Finite Pt magnetisation does not improve convergence

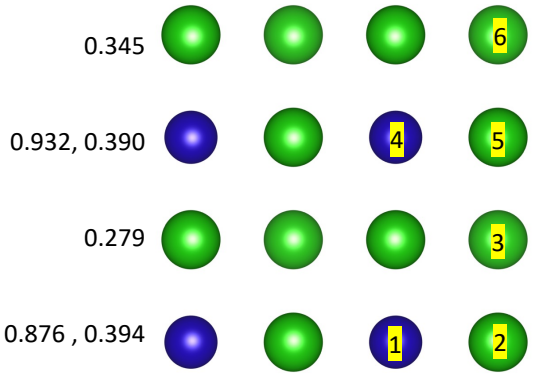


Pt_3Ni bulk optimization

- Pt_3Ni 2x2x2 bulk: 24 Pt atoms, 8 Ni atoms. EPS_SCF=1.0E-6
- 8x8 2D grid search for BROYDEN_MIXING ALPHA, BETA
- Minimum at ALPHA=4.0E-2, BETA=4.0E-3



$Pt_3Ni(001)$ slab in vacuum



- Convergence is challenging, EPS_SCF=1E-7 required 12 restarts to converge EPS_SCF=5.0E-7
- Slab (right) spin moments larger than bulk (left), consistent with literature [1]

Hirshfeld Charges

#Atom	Element	Kind	Ref Charge	Population		Spin moment	Net charge
1	Ni	1	18.000	9.422	8.809	0.612	-0.231
2	Ni	1	18.000	9.422	8.809	0.612	-0.231
3	Pt	2	18.000	9.066	8.857	0.209	0.077
4	Pt	2	18.000	9.066	8.857	0.209	0.077
5	Ni	1	18.000	9.422	8.809	0.612	-0.231
6	Ni	1	18.000	9.422	8.809	0.612	-0.231
7	Pt	2	18.000	9.066	8.857	0.209	0.077
8	Pt	2	18.000	9.066	8.857	0.209	0.077
9	Pt	3	18.000	9.066	8.857	0.209	0.077
10	Pt	3	18.000	9.066	8.857	0.209	0.077
11	Pt	3	18.000	9.066	8.857	0.209	0.077
12	Pt	3	18.000	9.066	8.857	0.209	0.077
13	Pt	3	18.000	9.066	8.857	0.209	0.077
14	Pt	3	18.000	9.066	8.857	0.209	0.077
15	Pt	3	18.000	9.066	8.857	0.209	0.077
16	Pt	3	18.000	9.066	8.857	0.209	0.077
17	Ni	4	18.000	9.422	8.809	0.612	-0.231
18	Ni	4	18.000	9.422	8.809	0.612	-0.231
19	Pt	5	18.000	9.066	8.857	0.209	0.077
20	Pt	5	18.000	9.066	8.857	0.209	0.077
21	Ni	4	18.000	9.422	8.809	0.612	-0.231
22	Ni	4	18.000	9.422	8.809	0.612	-0.231
23	Pt	5	18.000	9.066	8.857	0.209	0.077
24	Pt	5	18.000	9.066	8.857	0.209	0.077
25	Pt	6	18.000	9.066	8.857	0.209	0.077
26	Pt	6	18.000	9.066	8.857	0.209	0.077
27	Pt	6	18.000	9.066	8.857	0.209	0.077
28	Pt	6	18.000	9.066	8.857	0.209	0.077
29	Pt	6	18.000	9.066	8.857	0.209	0.077
30	Pt	6	18.000	9.066	8.857	0.209	0.077
31	Pt	6	18.000	9.066	8.857	0.209	0.077
32	Pt	6	18.000	9.066	8.857	0.209	0.077

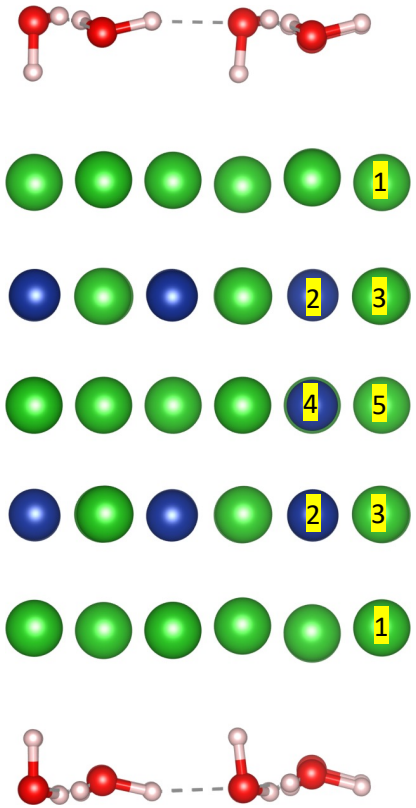
Hirshfeld Charges

#Atom	Element	Kind	Ref Charge	Population		Spin moment	Net charge
1	Ni	1	18.000	9.516	8.640	0.876	-0.156
2	Ni	1	18.000	9.516	8.640	0.876	-0.156
3	Pt	2	18.000	9.173	8.779	0.394	0.049
4	Pt	2	18.000	9.173	8.779	0.394	0.049
5	Ni	1	18.000	9.516	8.640	0.876	-0.156
6	Ni	1	18.000	9.516	8.640	0.876	-0.156
7	Pt	2	18.000	9.173	8.779	0.394	0.049
8	Pt	2	18.000	9.173	8.779	0.394	0.049
9	Pt	3	18.000	9.094	8.815	0.279	0.091
10	Pt	3	18.000	9.094	8.815	0.279	0.091
11	Pt	3	18.000	9.094	8.815	0.279	0.091
12	Pt	3	18.000	9.094	8.815	0.279	0.091
13	Pt	3	18.000	9.094	8.815	0.279	0.091
14	Pt	3	18.000	9.094	8.815	0.279	0.091
15	Pt	3	18.000	9.094	8.815	0.279	0.091
16	Pt	3	18.000	9.094	8.815	0.279	0.091
17	Ni	4	18.000	9.555	8.623	0.932	-0.178
18	Ni	4	18.000	9.555	8.623	0.932	-0.178
19	Pt	5	18.000	9.148	8.759	0.390	0.093
20	Pt	5	18.000	9.148	8.759	0.390	0.093
21	Ni	4	18.000	9.555	8.623	0.932	-0.178
22	Ni	4	18.000	9.555	8.623	0.932	-0.178
23	Pt	5	18.000	9.148	8.759	0.390	0.093
24	Pt	5	18.000	9.148	8.759	0.390	0.093
25	Pt	6	18.000	9.169	8.825	0.345	0.006
26	Pt	6	18.000	9.169	8.825	0.345	0.006
27	Pt	6	18.000	9.169	8.825	0.345	0.006
28	Pt	6	18.000	9.169	8.825	0.345	0.006
29	Pt	6	18.000	9.169	8.825	0.345	0.006
30	Pt	6	18.000	9.169	8.825	0.345	0.006
31	Pt	6	18.000	9.169	8.825	0.345	0.006
32	Pt	6	18.000	9.169	8.825	0.345	0.006

[1] Sharma, B. K., Kwon, O., Odkhuu, D. & Hong, S. C. Electronic structure and magnetism of various surfaces of the catalytic material Pt3Ni: Density-functional study. *Journal of Magnetism and Magnetic Materials* 339, 89–93 (2013).

$Pt_3Ni(111)$ Yike structure

- $Pt_3Ni(111)$ surface calculated by Yike using VASP. 45 Pt atoms, 15 Ni atoms
- CUTOFF (600), ADDED_MOS (30), ELECTRONIC_TEMPERATURE (300)



Kpoints	Multiplicity	Energy / Ha	IASD	SCF steps	Restarts
1x1x1	(0*15)+1=1	-8223.3280	0.0	85	0
1x1x1	(2*15)+1=31	-8223.4133	28.6	286	19
4x4x1	(0*15)+1=1	-8223.6260	0.0	74	0
4x4x1	(2*15)+1=31	-8223.6865	24.4	245	19

[1] Sharma, B. K., Kwon, O., Odkhuu, D. & Hong, S. C. Electronic structure and magnetism of various surfaces of the catalytic material Pt3Ni: Density-functional study. *Journal of Magnetism and Magnetic Materials* 339, 89–93 (2013).