

Supporting Information

SBH10: A Benchmark Database of Barrier Heights on Transition Metal Surfaces

Shaama Mallikarjun Sharada,¹ Thomas Bligaard,¹ Alan C. Luntz,¹ Geert-Jan Kroes,²
Jens K. Nørskov^{*1}

¹Department of Chemical Engineering, Stanford University, 443 Via Ortega, Stanford
CA 94305, USA

²Leiden Institute of Chemistry, Gorlaeus Laboratories, Leiden University, P.O. Box
9502, 2300 RA Leiden, The Netherlands

*Corresponding author: norskov@stanford.edu

Computational procedure

All calculations are carried out with the Vasp 5.3.5 simulation package^{1,2,3,4} using the Atomic Simulation Environment (ASE) interface⁵ and the default Vasp projected augmented wave (PAW) potentials.^{6,7} Since Ru-based adsorption energies are sensitive to the underlying pseudopotential,^{8,9} the hard `_pv` pseudopotential with 14 valence electrons is employed for all calculations except those with the HSE06 functional. A hard pseudopotential, `_h`, is used for N₂ dissociation reactions. Since the model surfaces must not only be large enough to accurately estimate barriers but also small enough for carrying out hybrid calculations, slabs are modeled using 2x2 unit cells with 6 layers. The exception is the Ni(211) slab used to model step sites in Ni(111) for CH₄ dissociation, which is 3x3x4 in size.

For CH₄ dissociation on Ni(111) and Ni(211), the slabs are completely frozen since combined quantum dynamics and reactive scattering studies employed a frozen lattice approximation. Since the choice of slab flexibility cannot be clearly determined for CH₄ dissociation on Ni(100), we calculate barriers using both a fully frozen slab model and one with the top 2 layers flexible. The BEEF-vdW barriers from the two setups differ by less than 0.2 eV, and therefore, we employ the flexible lattice model for CH₄ dissociation on Ni(100).

Similarly, the top two slab layers are allowed to relax for the remaining reactions. At least 10 Å vacuum separates the periodic images along the z axis. The plane-wave cutoff is set to 500 eV for GGA-vdW and meta-GGA calculations, and 400 eV for the screened hybrid functional. The convergence of the N₂/Ru(0001) barrier using the harder pseudopotential with respect to cutoff energy is verified via single point calculations using an energy cutoff of 900 eV. Gamma-centered k-point grids of (4x4x1) and (4x6x1) are employed for reactions on terrace and step sites, respectively. A gradient tolerance of 0.05 eV/ Å is used during geometry relaxation. Spin-polarized calculations are performed for dissociation reactions on the Ni surface. Transition state structures are determined using the “fixed-bond length” method¹⁰ or the dimer method.¹¹ The fixed-bond length method is initiated from the dissociated/chemisorbed state, for which the adsorbate locations are described in Table S1. Vibrational analysis is carried out using the BEEF-vdW functional to verify the transition state geometries and to determine the zero-point corrected barrier heights. Zero-point energies calculated with BEEF-vdW are employed for ground state corrections across all functionals.

Table S1. Adsorbate positions in the chemisorbed state of the dissociating system. These geometries are employed to initialize the fixed-bond length method as well as to calculate energies of the final state.

	Dissociation reaction, site type	Final state, adsorption sites
1	H ₂ /Cu(111), terrace	Hollow ¹²
2	H ₂ /Cu(100), terrace	Hollow ¹²
3	H ₂ /Pt(111), terrace	FCC, Bridge
4	H ₂ /Ru(0001), terrace	Hollow, Hollow ¹³
5	N ₂ /Ru(0001), terrace	Hollow, Hollow ¹⁴
6	N ₂ /Ru(0001), step	Hollow, Bridge ¹⁴
7	CH ₄ /Ru(0001), terrace	FCC, FCC ¹⁵
8	CH ₄ /Ni(100), terrace	Hollow (CH ₃)*, Hollow (H) ¹⁶
9	CH ₄ /Ni(111), terrace	FCC (CH ₃), Hollow (H) ¹⁶
10	CH ₄ /Ni(111), step	Bridge (CH ₃), Hollow (H) ¹⁷

*The minimum energy configuration with BEEF-vdW corresponds to CH₃ at the bridge site and not hollow site.

PBE Barrier Heights

Figure S1 demonstrates that, along similar lines to the MS2 functional, PBE overbinds chemisorbed H and as a result, underestimates barrier heights.

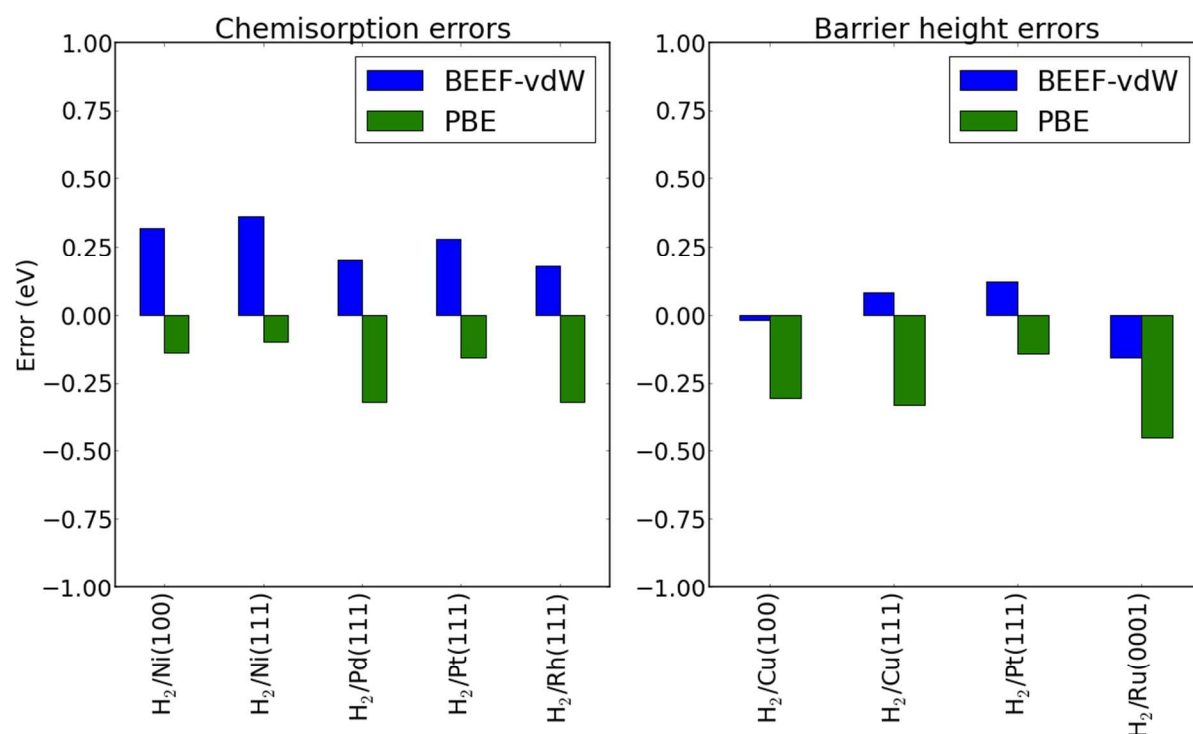


Figure S1. Comparison between BEEF-vdW and PBE errors (eV) in the prediction of H chemisorption energies in the ADS41 database and barrier heights for H₂ dissociation reactions in the SBH10 database. Chemisorption energy errors (per adsorbate) are scaled by a factor of 2 for appropriate comparison with barrier heights.

Transition state geometries

Vasp POSCAR files corresponding to the BEEF-vdW transition state geometries of the reactions constituting the SBH10 database are listed below:

1. H₂ dissociation on Cu(111) terrace

```

H Cu
1.0000000000000000
5.1832765538605026 0.0000000000000000 0.0000000000000000
2.5916382769302513 4.4888491704834541 0.0000000000000000
0.0000000000000000 0.0000000000000000 31.5003189605748659

```

2 24
 Selective dynamics
 Cartesian

1.4876980920747711	0.8614212612400526	11.5809712078238611	T	T	T
2.3830217316054481	1.3782534408336331	11.5807960711610143	T	T	T
1.2958191384651254	0.7481415284139090	0.0000000000000000	F	F	F
3.8874574153953767	0.7481415284139090	0.0000000000000000	F	F	F
2.5916382769302508	2.9925661136556361	0.0000000000000000	F	F	F
5.1832765538605017	2.9925661136556361	0.0000000000000000	F	F	F
0.0000000000000000	1.4962830568278180	2.1160637921149732	F	F	F
2.5916382769302517	1.4962830568278180	2.1160637921149732	F	F	F
1.2958191384651256	3.7407076420695446	2.1160637921149732	F	F	F
3.8874574153953771	3.7407076420695446	2.1160637921149732	F	F	F
0.0000000000000000	0.0000000000000000	4.2321275842299464	F	F	F
2.5916382769302513	0.0000000000000000	4.2321275842299464	F	F	F
1.2958191384651256	2.2444245852417271	4.2321275842299464	F	F	F
3.8874574153953771	2.2444245852417271	4.2321275842299464	F	F	F
1.2958191384651254	0.7481415284139090	6.3481913763449214	F	F	F
3.8874574153953767	0.7481415284139090	6.3481913763449214	F	F	F
2.5916382769302508	2.9925661136556361	6.3481913763449214	F	F	F
5.1832765538605017	2.9925661136556361	6.3481913763449214	F	F	F
-0.0159816921390811	1.4896224270397787	8.4367739611807355	T	T	T
2.5760009496869833	1.4880679145209954	8.3874920193659381	T	T	T
1.2867797492425008	3.7363663809113410	8.4596742949180790	T	T	T
3.8728808590549302	3.7315271923931315	8.4368408723348374	T	T	T
-0.0082399923984361	-0.0019705203954350	10.5537801913838418	T	T	T
2.6054743353305945	-0.0311317567107552	10.5273966216620423	T	T	T
1.2739898730747206	2.2750983332625108	10.5274867237150289	T	T	T
3.8804758127920662	2.2431163345760829	10.5353983677235838	T	T	T

2. H₂ dissociation on Cu(100) terrace

H Cu
 1.0000000000000000
 5.1832765538605026 0.0000000000000000 0.0000000000000000
 0.0000000000000000 5.1832765538605026 0.0000000000000000
 0.0000000000000000 0.0000000000000000 29.2628250000000030

2 24
 Selective dynamics
 Cartesian

1.9958085240946926	1.2958190909367027	10.1880488136399254	T	T	T
3.1874459602545917	1.2958190909422944	10.1880571726712308	T	T	T
1.2958191384651256	1.2958191384651256	0.0000000000000000	F	F	F
3.8874574153953771	1.2958191384651256	0.0000000000000000	F	F	F
1.2958191384651256	3.8874574153953771	0.0000000000000000	F	F	F

3.8874574153953771	3.8874574153953771	0.0000000000000000	F	F	F
0.0000000000000000	0.0000000000000000	1.8325650000000007	F	F	F
2.5916382769302513	0.0000000000000000	1.8325650000000007	F	F	F
0.0000000000000000	2.5916382769302513	1.8325650000000007	F	F	F
2.5916382769302513	2.5916382769302513	1.8325650000000007	F	F	F
1.2958191384651256	1.2958191384651256	3.6651299999999996	F	F	F
3.8874574153953771	1.2958191384651256	3.6651299999999996	F	F	F
1.2958191384651256	3.8874574153953771	3.6651299999999996	F	F	F
3.8874574153953771	3.8874574153953771	3.6651299999999996	F	F	F
0.0000000000000000	0.0000000000000000	5.4976950000000002	F	F	F
2.5916382769302513	0.0000000000000000	5.4976950000000002	F	F	F
0.0000000000000000	2.5916382769302513	5.4976950000000002	F	F	F
2.5916382769302513	2.5916382769302513	5.4976950000000002	F	F	F
1.2782335459131786	1.2958189804949678	7.2785152650404203	T	T	T
3.9050620295664005	1.2958193654089443	7.2785325771859650	T	T	T
1.3175970107066259	3.8874571724640550	7.3835190624141678	T	T	T
3.8656625319144879	3.8874576312695419	7.3834965842394400	T	T	T
-0.0000006789675716	0.0012490058761559	9.1519324835598148	T	T	T
2.5916389125973991	-0.0036903624247988	9.1969735146613818	T	T	T
-0.0000006232760780	2.5903891317760515	9.1519326421691680	T	T	T
2.5916387074632459	2.5953286043937416	9.1969735725650992	T	T	T

3. H₂ dissociation on Pt(111) terrace

H Pt

1.0000000000000000		
5.6469406124201447	0.0000000000000000	0.0000000000000000
2.8234703062100723	4.8903940240179002	0.0000000000000000
0.0000000000000000	0.0000000000000000	26.3267692568574212

2 24

Selective dynamics

Cartesian

2.3710563300046772	0.0470766388111095	13.2910551880569514	T	T	T
3.3893904715161276	-0.0252753049637876	13.2927347440277650	T	T	T
1.4117351531050362	0.8150656706696500	0.0000000000000000	F	F	F
4.2352054593151083	0.8150656706696500	0.0000000000000000	F	F	F
2.8234703062100723	3.2602626826785999	0.0000000000000000	F	F	F
5.6469406124201447	3.2602626826785999	0.0000000000000000	F	F	F
0.0000000000000000	1.6301313413392999	2.3053538513714837	F	F	F
2.8234703062100723	1.6301313413392999	2.3053538513714837	F	F	F
1.4117351531050359	4.0753283533482501	2.3053538513714837	F	F	F
4.2352054593151083	4.0753283533482501	2.3053538513714837	F	F	F
0.0000000000000000	0.0000000000000000	4.6107077027429675	F	F	F
2.8234703062100723	0.0000000000000000	4.6107077027429675	F	F	F
1.4117351531050362	2.4451970120089501	4.6107077027429675	F	F	F

4.2352054593151083	2.4451970120089501	4.6107077027429675	F	F	F
1.4117351531050362	0.8150656706696500	6.9160615541144530	F	F	F
4.2352054593151083	0.8150656706696500	6.9160615541144530	F	F	F
2.8234703062100723	3.2602626826785999	6.9160615541144530	F	F	F
5.6469406124201447	3.2602626826785999	6.9160615541144530	F	F	F
0.0018429251997256	1.6358728107649267	9.1993678760893403	T	T	T
2.8262687382676184	1.6309215212105495	9.2097103177363806	T	T	T
1.4013371431699917	4.0930110311476602	9.2212567329970714	T	T	T
4.2479299642876764	4.0912737430156607	9.2210971363969829	T	T	T
0.0058252085913934	-0.0024159484313133	11.5224004533226019	T	T	T
2.8297405103921389	-0.0191504595480565	11.6501878654324891	T	T	T
1.4151759066865237	2.4355874269503941	11.5061880296818568	T	T	T
4.2373175405294745	2.4350197899672277	11.5019249017842657	T	T	T

4. H₂ dissociation on Ru(0001) terrace

Ru H

1.0000000000000000

5.4457599999999999	0.0000000000000000	0.0000000000000000
2.7228800000000000	4.7161665029131443	0.0000000000000000
0.0000000000000000	0.0000000000000000	25.6300499999999971

24 2

Selective dynamics

Cartesian

0.0000000000000000	1.5720555009710480	0.0000000000000000	F	F	F
2.7228800000000000	1.5720555009710480	0.0000000000000000	F	F	F
1.3614399999999998	3.9301387524276201	0.0000000000000000	F	F	F
4.0843200000000000	3.9301387524276201	0.0000000000000000	F	F	F
0.0000000000000000	0.0000000000000000	2.1460099999999995	F	F	F
2.7228800000000000	0.0000000000000000	2.1460099999999995	F	F	F
1.3614400000000000	2.3580832514565722	2.1460099999999995	F	F	F
4.0843200000000000	2.3580832514565722	2.1460099999999995	F	F	F
0.0000000000000000	1.5720555009710480	4.2920199999999999	F	F	F
2.7228800000000000	1.5720555009710480	4.2920199999999999	F	F	F
1.3614399999999998	3.9301387524276201	4.2920199999999999	F	F	F
4.0843200000000000	3.9301387524276201	4.2920199999999999	F	F	F
0.0000000000000000	0.0000000000000000	6.4380299999999986	F	F	F
2.7228800000000000	0.0000000000000000	6.4380299999999986	F	F	F
1.3614400000000000	2.3580832514565722	6.4380299999999986	F	F	F
4.0843200000000000	2.3580832514565722	6.4380299999999986	F	F	F
0.0113605875579721	1.5682240856000120	8.5953923457114279	T	T	T
2.7370193985630196	1.5771783393301695	8.6117459293055560	T	T	T
1.3713036297688137	3.9270044146025382	8.6206333887193676	T	T	T
4.0864712593420354	3.9194524132358275	8.6074358861726488	T	T	T
0.0142083939301368	-0.0050938924523756	10.6690964240924462	T	T	T

2.7769817030050583	-0.0167732737701897	10.7668232934355821	T	T	T
1.3717437777738668	2.3535095012880167	10.6676848539829905	T	T	T
4.0833863589862354	2.3644399909895566	10.6815432307627791	T	T	T
2.4306356027290463	-0.4782810813612458	12.3369421812573528	T	T	T
3.0618767147143808	0.4611285415602422	12.3441676876769115	T	T	T

5. N₂ dissociation on Ru(0001) terrace

Ru N

1.0000000000000000		
5.4457599999999999	0.0000000000000000	0.0000000000000000
2.7228800000000000	4.7161665029131443	0.0000000000000000
0.0000000000000000	0.0000000000000000	25.6300499999999971

24 2

Selective dynamics

Cartesian

0.0000000000000000	1.5720555009710480	0.0000000000000000	F	F	F
2.7228800000000000	1.5720555009710480	0.0000000000000000	F	F	F
1.3614399999999998	3.9301387524276201	0.0000000000000000	F	F	F
4.0843200000000000	3.9301387524276201	0.0000000000000000	F	F	F
0.0000000000000000	0.0000000000000000	2.1460099999999995	F	F	F
2.7228800000000000	0.0000000000000000	2.1460099999999995	F	F	F
1.3614400000000000	2.3580832514565722	2.1460099999999995	F	F	F
4.0843200000000000	2.3580832514565722	2.1460099999999995	F	F	F
0.0000000000000000	1.5720555009710480	4.2920199999999999	F	F	F
2.7228800000000000	1.5720555009710480	4.2920199999999999	F	F	F
1.3614399999999998	3.9301387524276201	4.2920199999999999	F	F	F
4.0843200000000000	3.9301387524276201	4.2920199999999999	F	F	F
0.0000000000000000	0.0000000000000000	6.4380299999999986	F	F	F
2.7228800000000000	0.0000000000000000	6.4380299999999986	F	F	F
1.3614400000000000	2.3580832514565722	6.4380299999999986	F	F	F
4.0843200000000000	2.3580832514565722	6.4380299999999986	F	F	F
-0.0300683650043998	1.5892647012794574	8.6350210394138820	T	T	T
2.7377590376172782	1.5817522327891413	8.6193177849111020	T	T	T
1.3771099372209172	3.9209336394166718	8.5617817028433389	T	T	T
4.0833634237255287	3.9121949683431056	8.6196580098343585	T	T	T
0.0117676025111950	0.0509225812133375	10.7556535691723685	T	T	T
2.7926510557396051	-0.0407887303754571	10.7099909174245695	T	T	T
1.3224274454905605	2.3231861569734140	10.7550912102090805	T	T	T
4.0612201388978395	2.3708099677557071	10.8291473277753791	T	T	T
1.3921112247401506	-0.2381646776925015	12.1016320085225644	T	T	T
2.2666607300807327	1.2748116210746814	12.1004422968141743	T	T	T

6. N₂ dissociation on Ru(1010) step

Ru N

```
1.0000000000000000
  5.445759999999999  0.000000000000000  0.000000000000000
  0.000000000000000  4.292019999999999  0.000000000000000
  0.000000000000000  0.000000000000000  34.2624717582539020
```

24 2

Selective dynamics

Cartesian

```
1.361440000000000  0.000000000000000  0.000000000000000  F F F
4.084320000000000  0.000000000000000  0.000000000000000  F F F
1.361440000000000  2.146010000000000 -1.5720555009710484 F F F
4.084320000000000  2.146010000000000 -1.5720555009710484 F F F
0.000000000000000  0.000000000000000  2.3580832514565717 F F F
2.722880000000000  0.000000000000000  2.3580832514565717 F F F
0.000000000000000  2.146010000000000  0.7860277504855233 F F F
2.722880000000000  2.146010000000000  0.7860277504855233 F F F
1.361440000000000  0.000000000000000  4.7161665029131452 F F F
4.084320000000000  0.000000000000000  4.7161665029131452 F F F
1.361440000000000  2.146010000000000  3.1441110019420950 F F F
4.084320000000000  2.146010000000000  3.1441110019420950 F F F
0.000000000000000  0.000000000000000  7.0742497543697151 F F F
2.722880000000000  0.000000000000000  7.0742497543697151 F F F
0.000000000000000  2.146010000000000  5.5021942533986685 F F F
2.722880000000000  2.146010000000000  5.5021942533986685 F F F
1.3616823369693998 -0.0087407054952561  9.4332027358147759 T T T
4.0843175873659732  0.0118543676146137  9.4448865653677210 T T T
1.3614482048570928  2.1374863170347842  7.9141471946673700 T T T
4.0843489190137134  2.1423186990526388  7.8593723269530180 T T T
-0.0857234539859228  0.0271378338581212  11.7953311686531723 T T T
2.8092300816477960  0.0273721005103804  11.7952611432429144 T T T
-0.0352739567894253  2.1831178144054006  10.2762959211668736 T T T
2.7584314219639139  2.1835144731435556  10.2764318309604512 T T T
1.3617414978214115  1.0198149006194366  12.6545645882091105 T T T
1.3617336686275869  2.5732939263201180  11.7477346690833446 T T T
```

7. CH₄ dissociation on Ru(0001) terrace

Ru C H

```
1.0000000000000000
  5.445759999999999  0.000000000000000  0.000000000000000
  2.722880000000000  4.7161665029131443  0.000000000000000
  0.000000000000000  0.000000000000000  26.6628034350128331
```

24 1 4

Selective dynamics

Cartesian

0.0000000000000000	1.5720555009710480	0.0000000000000000	F	F	F
2.7228800000000000	1.5720555009710480	0.0000000000000000	F	F	F
1.3614399999999998	3.9301387524276201	0.0000000000000000	F	F	F
4.0843200000000000	3.9301387524276201	0.0000000000000000	F	F	F
0.0000000000000000	0.0000000000000000	2.1460099999999995	F	F	F
2.7228800000000000	0.0000000000000000	2.1460099999999995	F	F	F
1.3614400000000000	2.3580832514565722	2.1460099999999995	F	F	F
4.0843200000000000	2.3580832514565722	2.1460099999999995	F	F	F
0.0000000000000000	1.5720555009710480	4.2920199999999999	F	F	F
2.7228800000000000	1.5720555009710480	4.2920199999999999	F	F	F
1.3614399999999998	3.9301387524276201	4.2920199999999999	F	F	F
4.0843200000000000	3.9301387524276201	4.2920199999999999	F	F	F
0.0000000000000000	0.0000000000000000	6.4380299999999986	F	F	F
2.7228800000000000	0.0000000000000000	6.4380299999999986	F	F	F
1.3614400000000000	2.3580832514565722	6.4380299999999986	F	F	F
4.0843200000000000	2.3580832514565722	6.4380299999999986	F	F	F
-0.0015654427534141	1.5675537571010469	8.5776305426647053	T	T	T
2.7140683695450063	1.5608197358245053	8.6088558655555758	T	T	T
1.3528208627045399	3.9202073327161107	8.6112144482151098	T	T	T
4.0920772230034590	3.9261437886694681	8.6141012841325253	T	T	T
-0.0131541642526940	-0.0123309240422989	10.6526824346925455	T	T	T
2.7090528112151580	-0.0385499104158684	10.7740174497875412	T	T	T
1.3528544906255418	2.3389545822985425	10.6743873411406618	T	T	T
4.0802155673069240	2.3502935926879509	10.6541195142504144	T	T	T
2.6491769457401197	-0.1399462349800229	13.0844460250952768	T	T	T
3.4536265409487772	0.4744945879374750	13.4876582365150579	T	T	T
1.7309238353386065	0.0161205407752039	13.6540561133487586	T	T	T
2.9117358257928938	-1.1974031466302739	13.1368500350402044	T	T	T
2.0061621274328942	0.8357433093867128	12.0505642402553246	T	T	T

8. CH₄ dissociation on Ni(100) terrace

Ni C H

1.0000000000000000

4.9978448715621422	0.0000000000000000	0.0000000000000000
0.0000000000000000	4.9978448715621422	0.0000000000000000
0.0000000000000000	0.0000000000000000	24.4677784350128320

24 1 4

Selective dynamics

Cartesian

1.2494612178905355	1.2494612178905355	0.0000000000000000	F	F	F
3.7483836536716066	1.2494612178905355	0.0000000000000000	F	F	F
1.2494612178905355	3.7483836536716066	0.0000000000000000	F	F	F
3.7483836536716066	3.7483836536716066	0.0000000000000000	F	F	F
0.0000000000000000	0.0000000000000000	1.7670049999999993	F	F	F

2.4989224357810711	0.0000000000000000	1.7670049999999993	F	F	F
0.0000000000000000	2.4989224357810711	1.7670049999999993	F	F	F
2.4989224357810711	2.4989224357810711	1.7670049999999993	F	F	F
1.2494612178905355	1.2494612178905355	3.5340100000000003	F	F	F
3.7483836536716066	1.2494612178905355	3.5340100000000003	F	F	F
1.2494612178905355	3.7483836536716066	3.5340100000000003	F	F	F
3.7483836536716066	3.7483836536716066	3.5340100000000003	F	F	F
0.0000000000000000	0.0000000000000000	5.3010149999999996	F	F	F
2.4989224357810711	0.0000000000000000	5.3010149999999996	F	F	F
0.0000000000000000	2.4989224357810711	5.3010149999999996	F	F	F
2.4989224357810711	2.4989224357810711	5.3010149999999996	F	F	F
1.2609854530870526	1.2610009610899751	7.0755741221228652	T	T	T
3.7414152785900465	1.2702963401040537	7.0788281024354704	T	T	T
1.2550033039130013	3.7382940160222606	7.0590887959459030	T	T	T
3.7506992597378792	3.7317745065434260	7.0559553456341639	T	T	T
0.0045833817156291	0.0071943237014594	8.7228615070188749	T	T	T
2.5284896871117239	-0.0233851826939427	8.7654888110749809	T	T	T
-0.0046560755939989	2.5041007690178803	8.8162891159626415	T	T	T
2.4704980864169537	2.4974634849145643	8.9451634729006919	T	T	T
1.5994847744094671	2.5981745727315908	10.7240687349655222	T	T	T
1.6396055668651235	3.5326236458908968	11.2811122493584914	T	T	T
0.5407546902165918	2.2547858858951373	10.7633817292283851	T	T	T
2.1789697348423203	1.8262646073735596	11.2442101879404692	T	T	T
2.8776982957646102	3.6876637969041863	9.8158188746312778	T	T	T

9. CH₄ dissociation on Ni(111) terrace

Ni C H

1.0000000000000000

4.9978448715621422	0.0000000000000000	0.0000000000000000
2.4989224357810711	4.3282606229465888	0.0000000000000000
0.0000000000000000	0.0000000000000000	25.8345615591069802

24 1 4

Selective dynamics

Cartesian

1.2494612178905355	0.7213767704910981	0.0000000000000000	F	F	F
3.7483836536716062	0.7213767704910981	0.0000000000000000	F	F	F
2.4989224357810711	2.8855070819643922	0.0000000000000000	F	F	F
4.9978448715621422	2.8855070819643922	0.0000000000000000	F	F	F
0.0000000000000000	1.4427535409821961	2.0403616248188285	F	F	F
2.4989224357810711	1.4427535409821961	2.0403616248188285	F	F	F
1.2494612178905353	3.6068838524554905	2.0403616248188285	F	F	F
3.7483836536716062	3.6068838524554905	2.0403616248188285	F	F	F
0.0000000000000000	0.0000000000000000	4.0807232496376589	F	F	F
2.4989224357810711	0.0000000000000000	4.0807232496376589	F	F	F

1.2494612178905355	2.1641303114732944	4.0807232496376589	F	F	F
3.7483836536716066	2.1641303114732944	4.0807232496376589	F	F	F
1.2494612178905355	0.7213767704910981	6.1210848744564892	F	F	F
3.7483836536716062	0.7213767704910981	6.1210848744564892	F	F	F
2.4989224357810711	2.8855070819643922	6.1210848744564892	F	F	F
4.9978448715621422	2.8855070819643922	6.1210848744564892	F	F	F
0.0000000000000000	1.4427535409821961	8.1614464992753177	F	F	F
2.4989224357810711	1.4427535409821961	8.1614464992753177	F	F	F
1.2494612178905353	3.6068838524554905	8.1614464992753177	F	F	F
3.7483836536716062	3.6068838524554905	8.1614464992753177	F	F	F
0.0000000000000000	0.0000000000000000	10.2018081240941463	F	F	F
2.4989224357810711	0.0000000000000000	10.2018081240941463	F	F	F
1.2494612178905355	2.1641303114732944	10.2018081240941463	F	F	F
3.7483836536716066	2.1641303114732944	10.2018081240941463	F	F	F
1.3186911604041154	2.1286106300378771	12.2699371930332397	T	T	T
2.1794335151943893	2.6208574161222433	12.7278246651553602	T	T	T
0.3971802995682441	2.4713411776340535	12.7439370099920311	T	T	T
1.4141528953721489	1.0478267491334192	12.4160865116003567	T	T	T
1.2410654501823202	3.4043881699262610	11.1801033208591676	T	T	T

10. CH₄ dissociation on Ni(211) step

Ni C H

1.0000000000000000		
6.1210000000000004	0.0000000000000000	0.0000000000000000
0.0000000000000000	7.496999999999999	0.0000000000000000
0.0000000000000000	0.0000000000000000	22.5000000000000000

36 1 4

Selective dynamics

Cartesian

4.0807232500000001	1.2494612199999999	7.935144479999999	F	F	F
4.0807232500000001	3.7483836500000001	7.935144479999999	F	F	F
4.0807232500000001	6.2473060900000004	7.935144479999999	F	F	F
2.0403616200000001	0.0000000000000000	7.2137677000000000	F	F	F
2.0403616200000001	2.4989224399999999	7.2137677000000000	F	F	F
2.0403616200000001	4.9978448699999998	7.2137677000000000	F	F	F
0.0000000000000000	1.2494612199999999	6.4923909300000000	F	F	F
0.0000000000000000	3.7483836500000001	6.4923909300000000	F	F	F
0.0000000000000000	6.2473060900000004	6.4923909300000000	F	F	F
4.0807232500000001	0.0000000000000000	5.7710141600000000	F	F	F
4.0807232500000001	2.4989224399999999	5.7710141600000000	F	F	F
4.0807232500000001	4.9978448699999998	5.7710141600000000	F	F	F
2.0403616200000001	1.2494612199999999	5.0496373900000000	F	F	F
2.0403616200000001	3.7483836500000001	5.0496373900000000	F	F	F
2.0403616200000001	6.2473060900000004	5.0496373900000000	F	F	F

0.0000000000000000	0.0000000000000000	4.3282606200000000	F	F	F
0.0000000000000000	2.4989224399999999	4.3282606200000000	F	F	F
0.0000000000000000	4.9978448699999998	4.3282606200000000	F	F	F
4.0807232500000001	1.2494612199999999	3.6068838500000000	F	F	F
4.0807232500000001	3.7483836500000001	3.6068838500000000	F	F	F
4.0807232500000001	6.2473060900000004	3.6068838500000000	F	F	F
2.0403616200000001	0.0000000000000000	2.8855070800000000	F	F	F
2.0403616200000001	2.4989224399999999	2.8855070800000000	F	F	F
2.0403616200000001	4.9978448699999998	2.8855070800000000	F	F	F
0.0000000000000000	1.2494612199999999	2.1641303100000000	F	F	F
0.0000000000000000	3.7483836500000001	2.1641303100000000	F	F	F
0.0000000000000000	6.2473060900000004	2.1641303100000000	F	F	F
4.0807232500000001	0.0000000000000000	1.4427535400000000	F	F	F
4.0807232500000001	2.4989224399999999	1.4427535400000000	F	F	F
4.0807232500000001	4.9978448699999998	1.4427535400000000	F	F	F
2.0403616200000001	1.2494612199999999	0.7213767700000000	F	F	F
2.0403616200000001	3.7483836500000001	0.7213767700000000	F	F	F
2.0403616200000001	6.2473060900000004	0.7213767700000000	F	F	F
0.0000000000000000	0.0000000000000000	0.0000000000000000	F	F	F
0.0000000000000000	2.4989224399999999	0.0000000000000000	F	F	F
0.0000000000000000	4.9978448699999998	0.0000000000000000	F	F	F
3.8841046765216349	2.6452380079959057	9.5878974958559784	T	T	T
4.6473725216544670	3.2769127430602794	10.0645659058376982	T	T	T
2.9803905556672246	2.6499733079367269	10.1957427269418623	T	T	T
4.3015078128597919	1.5933220096648713	9.7036672086006348	T	T	T
2.7041753190355386	3.4284587448806128	8.5398306544303573	T	T	T

References

1. Kresse, G.; Hafner, J. Ab Initio Molecular Dynamics for Liquid Metals. *Phys. Rev. B.* **1993**, *47*, 558.
2. Kresse, G.; Hafner, J. Ab Initio Molecular-Dynamics Simulation of the Liquid-Metal–Amorphous-Semiconductor Transition in Germanium. *Phys. Rev. B.* **1994**, *49*, 14251.
3. Kresse, G.; Furthmüller, J. Efficiency of Ab-Initio Total Energy Calculations for Metals and Semiconductors Using a Plane-Wave Basis Set. *Comput. Mat. Sci.* **1996**, *6*, 15.
4. Kresse, G.; Furthmüller, J. Efficient Iterative Schemes for Ab Initio Total-Energy Calculations Using a Plane-Wave Basis Set. *Phys. Rev. B.* **1996**, *54*, 11169.
5. Bahn, S. R.; Jacobsen, K. W. An Object-Oriented Scripting Interface to a Legacy Electronic Structure Code. *Comput. Sci. Eng.* **2002**, *4* (3), 56–66.

-
6. Blöchl, P.E. Projected Augmented-Wave Method. *Phys. Rev. B.* **1994**, *50*, 17953.
 7. Kresse, G.; Joubert, J. From Ultrasoft Pseudopotentials to the Projector Augmented-Wave Method. *Phys. Rev. B.* **1999**, *59*, 1758.
 8. Briquet, L.; Sarwar, M.; Mugo, J.; Jones, G.; Calle-Vallejo, F. A New Type of Scaling Relations to Assess the Accuracy of Computational Predictions of Catalytic Activities Applied to the Oxygen Evolution Reaction. *Chem. Cat. Chem.* **2017**, *2*, 1261–1268.
 9. Kiejna, A.; Kresse, G.; Rogal, J.; De Sarkar, A.; Reuter, K.; Scheffler, M. Comparison of the Full-Potential and Frozen-Core Approximation Approaches to Density-Functional Calculations of Surfaces. *Phys. Rev. B.* **2006**, *73* (3), 6–9.
 10. Falsig, H.; Shen, J.; Khan, T. S.; Guo, W.; Jones, G.; Dahl, S.; Bligaard, T. On the Structure Sensitivity of Direct NO Decomposition over Low-Index Transition Metal Facets. *Top. Catal.* **2014**, *57* (1), 80–88.
 11. Henkelman, G.; Jonsson, H. A Dimer Method for Finding Saddle Points on High Dimensional Potential Surfaces Using only First Derivatives. *J. Chem. Phys.* **1999**, *111*, 7010.
 12. Kratzer, P.; Hammer, B.; Nørskov, J. Geometric and Electronic Factors Determining the Differences in Reactivity of H₂ on Cu(100) and Cu(111). *Surf. Sci.* **1996**, *359*, 45–53.
 13. Nieto, P.; Farías, D.; Miranda, R.; Luppi, M.; Baerends, E. J.; Somers, M. F.; van der Niet, M. J. T. C.; Olsen, R. A.; Kroes, G. J. Diffractive and Reactive Scattering of H₂ from Ru(0001): Experimental and Theoretical Study. *Phys. Chem. Chem. Phys.* **2011**, *13* (111), 8583–8597.
 14. Dahl, S.; Logadottir, A.; Egeberg, R. C.; Larsen, J. H.; Chorkendorff, I.; Törnqvist, E.; Nørskov, J. K. Role of Steps in Activation on Ru(0001). *Phys. Rev. Lett.* **1999**, *83* (9), 1814–1817.
 15. Ciobîcă, I. M.; Frechard, F.; van Santen, R. A.; Kleyn, A. W.; Hafner, J. A DFT Study of Transition States for C-H Activation on the Ru(0001) Surface, *J. Phys. Chem. B.* **2000**, *104*, 3364–3369.
 16. Nave, S.; Tiwari, A. K.; Jackson, B. Methane Dissociation and Adsorption on Ni(111), Pt(111), Ni(100), Pt(100), and Pt(110)-(1X2): Energetic Study. *J. Chem. Phys.* **2010**, *132*, 054705.

-
17. Abild-Pedersen, F.; Lytken, O.; Engbæk, J.; Nielsen, G.; Chorkendorff, I.; Nørskov, J. K. Methane Activation on Ni(111): Effects of Poisons and Step Defects. *Surf. Sci.* **2005**, *590* (2–3), 127–137.