Supporting information to:

Dynamics of H_2 dissociation on the closepacked (111) surface of the noblest metal: H_2 + Au(111)

Mark Wijzenbroek, Darcey Helstone, Jörg Meyer, and Geert-Jan Kroes*

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

*Corresponding author

Table S1. Lattice constants for fcc Au metal computed for the six density functionals employed in this work.

Functional	Lattice constant (Å)
PBE	4.1816
SRP48	4.2022
RPBE	4.2258
PBE-vdW-DF	4.2286
optPBE-vdW-DF	4.1967
RPBE-vdW-DF	4.2747

Table S2. The H-H distance r_b and the H_2-surface distance Z_b at the minimum barrier geometry, and the minimum barrier height E_b are provided for configurations in which H_2 is parallel to the Au(111) surface (θ=90°). Results are provided for the PBE and for the RPBE functional.

	PBE			RPBE		
Configuration	r_b (Å)	Z_b (Å)	E_b (eV)	r_b (Å)	Z_b (Å)	E_b (eV)
top, ϕ=0°	1.504	1.468	1.237	1.497	1.472	1.538
bridge, ϕ=0°	1.516	1.485	1.175	1.526	1.484	1.465
bridge, ϕ=90°	1.187	1.096	1.245	1.187	1.077	1.579
hcp, ϕ=0°	1.354	1.239	1.220	1.364	1.236	1.530
t2h, ϕ=120°	1.335	1.304	1.260	1.373	1.298	1.569
t2h, ϕ=30°	1.685	1.549	1.637	1.692	1.555	1.941

Table S3. The H-H distance r_b and the H_2-surface distance Z_b at the minimum barrier geometry, and the minimum barrier height E_b are provided for configurations in which H_2 is parallel to the Au(111) surface (θ=90°). Results are provided for the PBE-vdW-DF and for the RPBE-vdW-DF functional.

	PBE-vdW-DF			RPBE-vdW-DF		
Configuration	r_b (Å)	Z_b (Å)	E_b (eV)	r_b (Å)	Z_b (Å)	E_b (eV)
top, ϕ=0°	1.466	1.485	1.403	1.479	1.485	1.691
bridge, ϕ=0°	1.419	1.479	1.292	1.416	1.476	1.566
bridge, ϕ=90°	1.210	1.084	1.546	1.221	1.059	1.864
hcp, ϕ=0°	1.309	1.252	1.423	1.314	1.244	1.719
t2h, ϕ=120°	1.365	1.310	1.471	1.379	1.301	1.773
t2h, ϕ=30°	1.655	1.566	1.787	1.657	1.572	2.077

Table S4: Fit parameters for reaction probabilities of H_2, initially in the (ν, J) state, dissociating on Au(111) using the SRP48 functional.

ν	J	A	E_0' (eV)	W' (eV)	E_0'' (eV)	W'' (eV)
0	0	0.870012	1.253869	0.306219	1.141940	0.045706
0	1	0.876761	1.256545	0.298814	1.140102	0.047135
0	2	0.863459	1.245456	0.281118	1.133006	0.047119
0	3	0.881756	1.214835	0.325776	1.146103	0.054658
0	4	0.877897	1.192613	0.311916	1.135634	0.061577
0	5	0.861885	1.190620	0.287415	1.087751	0.069477
0	6	0.880247	1.170150	0.312985	1.042392	0.079144
0	7	0.858403	1.144569	0.290236	0.955146	0.076027
0	8	0.860402	1.105053	0.292517	0.875179	0.063638
0	9	0.867933	1.043478	0.316497	0.836307	0.066705
0	10	0.865676	0.967442	0.329360	0.779495	0.073836
0	11	0.879288	0.907849	0.343597	0.709849	0.059739
1	0	0.885275	0.862460	0.393213	0.764827	0.055309
1	1	0.868835	0.863494	0.345541	0.755142	0.054931
1	2	0.868954	0.815167	0.355915	0.757687	0.056321
1	3	0.877642	0.761554	0.386768	0.761320	0.051578
1	4	0.879837	0.734213	0.371086	0.759893	0.053191
1	5	0.871249	0.743341	0.337645	0.738880	0.061160
1	6	0.892346	0.714770	0.374096	0.722102	0.068884
1	7	0.890422	0.671823	0.381929	0.699879	0.076724

Table S5: Fit parameters for reaction probabilities of D_2, initially in the (ν, J) state, dissociating on Au(111) using the SRP48 functional.

ν	J	A	E_0' (eV)	W' (eV)	E_0'' (eV)	W'' (eV)
0	0	0.870389	1.321275	0.294550	1.214260	0.047377
0	1	0.871409	1.341825	0.278302	1.200420	0.039741
0	2	0.871218	1.310082	0.288646	1.219757	0.047986
0	3	0.856287	1.317178	0.263617	1.200524	0.042454
0	4	0.865550	1.302656	0.282573	1.207599	0.051395
0	5	0.867980	1.293206	0.287584	1.192862	0.050691
0	6	0.879706	1.267753	0.308803	1.197513	0.063177
0	7	0.870682	1.269528	0.288196	1.157517	0.067411
0	8	0.857451	1.262592	0.271441	1.095560	0.072422
0	9	0.860878	1.244761	0.285854	1.059041	0.068354
0	10	0.851979	1.212845	0.276998	1.000280	0.074887
0	11	0.850149	1.190772	0.275678	0.933009	0.051231
0	12	0.871520	1.151762	0.307530	0.916312	0.074698
0	13	0.870030	1.100590	0.317376	0.875442	0.064622
0	14	0.868342	1.049376	0.327227	0.837192	0.072489
0	15	0.864690	1.012599	0.311715	0.765274	0.050212
1	0	0.887632	1.016163	0.372949	0.916685	0.051205
1	1	0.880248	1.030085	0.346263	0.911882	0.052723
1	2	0.863760	0.993398	0.331061	0.914293	0.056063
1	3	0.869220	0.977994	0.340209	0.915855	0.054613
1	4	0.871370	0.965237	0.344695	0.916874	0.053261
1	5	0.875110	0.927704	0.359907	0.920550	0.054556
1	6	0.884060	0.904298	0.375043	0.922172	0.059830
1	7	0.873070	0.909959	0.343103	0.904857	0.067419
1	8	0.870317	0.918987	0.325705	0.865215	0.069894
1	9	0.870734	0.903785	0.332418	0.837490	0.073072
1	10	0.875389	0.887069	0.334409	0.807194	0.075738
1	11	0.870449	0.876340	0.325825	0.750258	0.071364
1	12	0.876758	0.861259	0.327789	0.713385	0.063240
2	0	0.883268	0.784284	0.398970	0.682225	0.056449
2	1	0.870290	0.799398	0.357590	0.675317	0.056087
2	2	0.865609	0.733084	0.368954	0.684361	0.065957
2	3	0.869035	0.710964	0.370001	0.677947	0.058995
2	4	0.879857	0.673082	0.395559	0.676766	0.052908
2	5	0.871037	0.651841	0.369843	0.667677	0.047994
2	6	0.881877	0.613118	0.389527	0.674841	0.052354
2	7	0.882064	0.599147	0.384069	0.676082	0.061625
2	8	0.879582	0.615587	0.356825	0.650582	0.061062
2	9	0.883183	0.599303	0.364607	0.639058	0.067085
2	10	0.884770	0.600416	0.352755	0.613337	0.072296

Table S6: Fit parameters for reaction probabilities of H_2, initially in the (ν, J) state, dissociating on Au(111) using the optPBE-vdW-DF functional.

ν	J	A	E_0' (eV)	W' (eV)	E_0'' (eV)	W'' (eV)
0	0	0.896350	1.284168	0.337951	1.200016	0.058846
0	1	0.874442	1.305263	0.280712	1.177880	0.052073
0	2	0.869986	1.290157	0.279372	1.183035	0.057697
0	3	0.887680	1.284172	0.311082	1.184780	0.058379
0	4	0.885778	1.247035	0.316416	1.177520	0.069912
0	5	0.858358	1.243134	0.278152	1.113651	0.088465
0	6	0.887247	1.216230	0.323547	1.080359	0.085884
0	7	0.865953	1.173121	0.313751	1.017383	0.089853
0	8	0.857841	1.146752	0.290674	0.895431	0.069768
0	9	0.872632	1.088120	0.320857	0.862465	0.064253
0	10	0.875232	1.004701	0.340977	0.808915	0.074876
0	11	0.886937	0.936760	0.354121	0.735466	0.068896
1	0	0.889607	0.870535	0.392887	0.773238	0.052376
1	1	0.878484	0.875061	0.351059	0.772404	0.055917
1	2	0.886670	0.837804	0.371294	0.784504	0.059173
1	3	0.895679	0.801331	0.400520	0.789333	0.055496
1	4	0.884933	0.788623	0.363541	0.781902	0.054791
1	5	0.884392	0.780218	0.354049	0.764181	0.067233
1	6	0.900018	0.773275	0.371327	0.733302	0.064776
1	7	0.897907	0.731495	0.382737	0.701558	0.069264

Table S7: Fit parameters for reaction probabilities of D_2, initially in the (ν, J) state, dissociating on Au(111) using the optPBE-vdW-DF functional.

ν	J	A	E_0' (eV)	W' (eV)	E_0'' (eV)	W'' (eV)
0	0	0.891926	1.362795	0.313404	1.278059	0.055873
0	1	0.859576	1.386019	0.258786	1.245960	0.052340
0	2	0.868443	1.367624	0.266887	1.263530	0.052609
0	3	0.854596	1.375831	0.246989	1.241193	0.048246
0	4	0.889121	1.348647	0.307133	1.278259	0.063623
0	5	0.858925	1.356840	0.262220	1.230237	0.064457
0	6	0.874995	1.330532	0.290633	1.236221	0.073797
0	7	0.878090	1.306793	0.299693	1.222614	0.083015
0	8	0.865349	1.306733	0.282502	1.145406	0.089854
0	9	0.866066	1.288303	0.296045	1.106622	0.083825
0	10	0.848217	1.258105	0.277898	1.029939	0.081015
0	11	0.852353	1.221391	0.288777	0.971822	0.095712
0	12	0.873706	1.200226	0.307221	0.943241	0.066873
0	13	0.876437	1.149221	0.322987	0.903921	0.066144
0	14	0.868226	1.098453	0.321534	0.852171	0.060338
0	15	0.877005	1.037098	0.338862	0.811876	0.077338
1	0	0.886544	1.044776	0.360715	0.933175	0.053757
1	1	0.878302	1.060020	0.333139	0.928228	0.052441
1	2	0.869207	1.026126	0.327198	0.940519	0.058783
1	3	0.876012	1.025173	0.330742	0.944035	0.055598
1	4	0.881098	1.011718	0.344881	0.950151	0.058195
1	5	0.874947	1.003014	0.331971	0.936137	0.054959
1	6	0.882523	0.988530	0.340778	0.937235	0.060919
1	7	0.882005	0.968435	0.341753	0.929589	0.073297
1	8	0.871488	0.994280	0.305421	0.865145	0.067211
1	9	0.885890	0.948179	0.352534	0.868056	0.073739
1	10	0.881612	0.924917	0.346499	0.831161	0.079287
1	11	0.878130	0.904109	0.343514	0.785111	0.077031
1	12	0.885591	0.886947	0.347909	0.740046	0.070972
2	0	0.892408	0.775094	0.412534	0.690710	0.052611
2	1	0.878367	0.787965	0.371923	0.688210	0.055629
2	2	0.881053	0.734321	0.387890	0.700368	0.063472
2	3	0.888064	0.722451	0.392939	0.699646	0.060011
2	4	0.894359	0.704588	0.404573	0.697590	0.056293
2	5	0.881731	0.676973	0.382999	0.695313	0.052401
2	6	0.891506	0.660282	0.392360	0.697675	0.055051
2	7	0.884033	0.673522	0.358256	0.683659	0.057354
2	8	0.891404	0.663823	0.365838	0.664207	0.059542
2	9	0.897235	0.631378	0.390281	0.651365	0.065639
2	10	0.892889	0.635790	0.368672	0.621258	0.065395

Table S8: Fit parameters for reaction probabilities of H_2, initially in the (ν, J) state, dissociating on Au(111) using the PBE functional.

ν	J	A	E_0' (eV)	W' (eV)	E_0'' (eV)	W'' (eV)
0	0	0.878787	1.105231	0.305696	1.003351	0.051525
0	1	0.884582	1.095763	0.300132	1.004770	0.058829
0	2	0.882615	1.085142	0.295785	0.997077	0.051367
0	3	0.889102	1.042242	0.330393	1.010367	0.057084
0	4	0.883310	1.021961	0.313555	1.000979	0.062815
0	5	0.879376	0.999165	0.319516	0.982633	0.078261
0	6	0.893024	1.010810	0.320107	0.918995	0.086563
0	7	0.887247	0.965050	0.332691	0.870308	0.091963
0	8	0.881422	0.956005	0.316998	0.753862	0.071654
0	9	0.884219	0.879531	0.347640	0.729736	0.081536
0	10	0.880604	0.812077	0.350581	0.664246	0.085530
0	11	0.885713	0.766036	0.340232	0.568254	0.069748
1	0	0.872506	0.784093	0.329462	0.590307	0.027351
1	1	0.872660	0.746200	0.327576	0.604846	0.062241
1	2	0.878417	0.673243	0.360466	0.627346	0.057370
1	3	0.886538	0.621169	0.380887	0.622854	0.047166
1	4	0.888531	0.577821	0.374144	0.629648	0.058013
1	5	0.883977	0.586598	0.349855	0.615281	0.067941
1	6	0.894908	0.592015	0.348946	0.588129	0.075427
1	7	0.891743	0.600938	0.329269	0.533371	0.075492

Table S9: Fit parameters for reaction probabilities of D_2, initially in the (ν, J) state, dissociating on Au(111) using the PBE functional.

ν	J	A	E_0' (eV)	W' (eV)	E_0'' (eV)	W'' (eV)
0	0	0.874062	1.169241	0.289206	1.073495	0.049491
0	1	0.891394	1.162141	0.311043	1.081281	0.055213
0	2	0.877136	1.151377	0.283578	1.070922	0.052769
0	3	0.877980	1.151216	0.280643	1.066304	0.048492
0	4	0.879284	1.141464	0.285479	1.069009	0.049689
0	5	0.881715	1.108473	0.306417	1.068025	0.054859
0	6	0.894906	1.069617	0.341199	1.078295	0.064614
0	7	0.878335	1.084616	0.300400	1.044422	0.075109
0	8	0.872986	1.099744	0.282276	0.975148	0.077372
0	9	0.883835	1.076881	0.309241	0.949141	0.082416
0	10	0.885625	1.036169	0.321230	0.916948	0.094211
0	11	0.876469	1.025277	0.310774	0.833401	0.087754
0	12	0.893749	0.986370	0.345001	0.815854	0.088314
0	13	0.887852	0.931168	0.352634	0.778630	0.083412
0	14	0.885193	0.871840	0.361768	0.746128	0.098527
0	15	0.875453	0.869675	0.315507	0.630902	0.051521
1	0	0.881378	0.900350	0.340402	0.758960	0.064016
1	1	0.880721	0.894485	0.333288	0.769000	0.072926
1	2	0.870294	0.850170	0.326344	0.778455	0.065264
1	3	0.872456	0.848353	0.320767	0.767351	0.051289
1	4	0.877438	0.824773	0.331736	0.768675	0.052068
1	5	0.881839	0.776515	0.355015	0.780470	0.058442
1	6	0.895821	0.719483	0.397157	0.795971	0.068370
1	7	0.886744	0.717878	0.371080	0.789106	0.076957
1	8	0.882181	0.745492	0.341629	0.750646	0.082014
1	9	0.885852	0.742730	0.345057	0.722137	0.088198
1	10	0.891705	0.737673	0.343632	0.682735	0.089914
1	11	0.885794	0.747888	0.330355	0.617792	0.070257
1	12	0.900086	0.696351	0.373439	0.613361	0.078017
2	0	0.875309	0.707282	0.344894	0.527369	0.026972
2	1	0.868451	0.706875	0.321731	0.521034	0.037567
2	2	0.870158	0.615938	0.354237	0.547183	0.067665
2	3	0.881401	0.564728	0.383776	0.551890	0.059508
2	4	0.888254	0.521108	0.401668	0.545200	0.048434
2	5	0.884696	0.495998	0.386552	0.537914	0.045092
2	6	0.892387	0.459587	0.399045	0.549433	0.054309
2	7	0.888101	0.465463	0.371961	0.545704	0.062101
2	8	0.887662	0.482489	0.348853	0.525678	0.069714
2	9	0.892603	0.487182	0.347388	0.506468	0.072300
2	10	0.891131	0.524783	0.315218	0.457348	0.065882