Supplementary Information

DFT study of hydrogen interaction with transition metal doped graphene for efficient hydrogen storage: effect of d-orbital occupancy and Kubas interaction

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Table S1: Fermi energy values of PG, GD, TM/PG and TM/GD.

System	Fermi energy (eV)	Fermi energy after H_2 adsorption (eV)
Pristine Graphene (PG)	-0.436	-0.352
Graphene with defect (GD)	-1.100	-0.990
Fe/PG	0.612	-0.340
Ag/GD	0.273	0.511
Au/GD	0.357	0.520
Cu/GD	-0.120	0.076
Fe/GD	0	0.592

Table S2: Binding energy of TM atom on the GD layer (E_b), Bond distance between TM atom and nearest carbon atom (D_{C-TM}), Shortest distance between TM atom and GD layer ($D_{GD\ layer-TM}$).

System	E_b (eV)	D_{TM-C} (Å)	$D_{GD\ layer-TM}$ (Å)
GD/Ag	-1.917	2.200	1.601
GD/Au	-2.554	2.125	1.485
GD/Cu	-3.971	1.875	1.096
GD/Fe	-10.003	1.746	0.889

Table S3: Adsorption energy of H_2 on the different systems using GGA-PPE and after adding DFT-D3 correction.

System	Adsorption Energy of H_2 (eV)	
	GGA-PBE	**DFT-D3**
GD/Ag – H_2	-0.405	-0.404
GD/Au – H_2	-0.341	-0.339
GD/Cu – H_2	-0.282	-0.260
GD/Fe – H_2	-0.580	-0.578
GD/Fe – 2 H_2	-0.606	-0.604
GD/Fe – 3 H_2	-0.866	-0.864
GD/Fe – 4 H_2	-0.902	-0.899
GD/Fe – 5 H_2	-0.921	-0.917

Table S4: Partial charge on the C1, C2, C3, H1, H2, TM before and after H_2 adsorption

System	C1	C2	C3	H1	H2	TM
Ag/GD	-0.148	-0.141	-0.147	-	-	0.779
Ag/GD-H_2	-0.120	-0.192	-0.130	0.037	0.018	0.784
Au/GD	-0.039	-0.088	-0.039	-	-	0.210
Au/GD-H_2	-0.031	-0.032	-0.069	0.105	0.099	0.105
Cu/GD	-0.072	-0.067	-0.072	-	-	0.329
Cu/GD-H_2	-0.088	-0.046	-0.056	0.091	0.101	0.159
Fe/GD	-0.075	-0.075	-0.075	-	-	0.576
Fe/GD-H_2	-0.074	-0.073	-0.076	0.008	0.056	0.545

Table S5: Löwdin charge analysis of TM in TM/GD system before and after H_2 adsorption

Ag/GD system Löwdin charge of Ag

```
Atom #  14: total charge =  10.2193, s =  0.5305, p =  0.0000, d =  9.6888,
            spin up      =   5.1103, s =  0.2651,
            spin up      =   5.1103, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
            spin up      =   5.1103, d =  4.8452, dz2=  0.9960, dxz=  0.9385, dyz=  0.9384, dx2-y2=  0.9862, dxy=  0.9861,
            spin down    =   5.1090, s =  0.2654,
            spin down    =   5.1090, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
            spin down    =   5.1090, d =  4.8436, dz2=  0.9959, dxz=  0.9378, dyz=  0.9377, dx2-y2=  0.9861, dxy=  0.9861,
            polarization =   0.0013, s = -0.0003, p =  0.0000, d =  0.0016,
```

Ag/GD system Löwdin charge of Ag after H_2 adsorption

```
Atom #  14: total charge =  10.2159, s =  0.4927, p =  0.0000, d =  9.7233,
            spin up      =   5.1101, s =  0.2447,
            spin up      =   5.1101, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
            spin up      =   5.1101, d =  4.8654, dz2=  0.9808, dxz=  0.9474, dyz=  0.9586, dx2-y2=  0.9919, dxy=  0.9866,
            spin down    =   5.1058, s =  0.2479,
            spin down    =   5.1058, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
            spin down    =   5.1058, d =  4.8579, dz2=  0.9807, dxz=  0.9422, dyz=  0.9573, dx2-y2=  0.9918, dxy=  0.9860,
            polarization =   0.0043, s = -0.0032, p =  0.0000, d =  0.0075,
```

Au/GD system Löwdin charge of Au

```
Atom #  14: total charge =  10.7904, s =  0.8131, p =  0.5858, d =  9.3915,
            spin up      =   5.3974, s =  0.4077,
            spin up      =   5.3974, p =  0.2937, pz=  0.1336, px=  0.0801, py=  0.0801,
            spin up      =   5.3974, d =  4.6959, dz2=  0.9901, dxz=  0.8836, dyz=  0.8837, dx2-y2=  0.9692, dxy=  0.9692,
            spin down    =   5.3930, s =  0.4054,
            spin down    =   5.3930, p =  0.2920, pz=  0.1321, px=  0.0800, py=  0.0800,
            spin down    =   5.3930, d =  4.6956, dz2=  0.9901, dxz=  0.8835, dyz=  0.8835, dx2-y2=  0.9692, dxy=  0.9692,
            polarization =   0.0044, s =  0.0024, p =  0.0017, d =  0.0003,
```

Au/GD system Löwdin charge of Au after H_2 adsorption

```
Atom #  14: total charge =  10.8950, s =  0.7832, p =  0.6867, d =  9.4250,
            spin up      =   5.4604, s =  0.3926,
            spin up      =   5.4604, p =  0.3497, pz=  0.1515, px=  0.1182, py=  0.0799,
            spin up      =   5.4604, d =  4.7181, dz2=  0.9531, dxz=  0.9000, dyz=  0.9219, dx2-y2=  0.9776, dxy=  0.9656,
            spin down    =   5.4346, s =  0.3906,
            spin down    =   5.4346, p =  0.3371, pz=  0.1492, px=  0.1134, py=  0.0744,
            spin down    =   5.4346, d =  4.7069, dz2=  0.9528, dxz=  0.8941, dyz=  0.9176, dx2-y2=  0.9771, dxy=  0.9652,
            polarization =   0.0258, s =  0.0020, p =  0.0126, d =  0.0112,
```

Cu/GD system Löwdin charge of Cu

```
Atom #  14: total charge =  10.6710, s =  0.5485, p =  0.6288, d =  9.4937,
            spin up      =   5.3363, s =  0.2742,
            spin up      =   5.3363, p =  0.3148, pz=  0.1145, px=  0.0975, py=  0.1027,
            spin up      =   5.3363, d =  4.7474, dz2=  0.9913, dxz=  0.9101, dyz=  0.9148, dx2-y2=  0.9644, dxy=  0.9667,
            spin down    =   5.3347, s =  0.2743,
            spin down    =   5.3347, p =  0.3140, pz=  0.1146, px=  0.0972, py=  0.1022,
            spin down    =   5.3347, d =  4.7463, dz2=  0.9913, dxz=  0.9098, dyz=  0.9143, dx2-y2=  0.9642, dxy=  0.9666,
            polarization =   0.0016, s = -0.0001, p =  0.0007, d =  0.0010,
```

Cu/GD system Löwdin charge of Cu after H_2 adsorption

```
Atom #  14: total charge =  10.8407, s =  0.5124, p =  0.7975, d =  9.5308,
            spin up      =   5.4262, s =  0.2561,
            spin up      =   5.4262, p =  0.4021, pz=  0.1418, px=  0.0925, py=  0.1678,
            spin up      =   5.4262, d =  4.7680, dz2=  0.9815, dxz=  0.9133, dyz=  0.9285, dx2-y2=  0.9658, dxy=  0.9788,
            spin down    =   5.4145, s =  0.2563,
            spin down    =   5.4145, p =  0.3954, pz=  0.1418, px=  0.0884, py=  0.1651,
            spin down    =   5.4145, d =  4.7628, dz2=  0.9814, dxz=  0.9114, dyz=  0.9269, dx2-y2=  0.9650, dxy=  0.9782,
            polarization =   0.0117, s = -0.0003, p =  0.0068, d =  0.0052,
```

Fe/GD system Löwdin charge of Fe

```
Atom #  14: total charge =   7.4241, s =  0.3612, p =  0.0000, d =  7.0629,
           spin up      =   3.7632, s =  0.1885,
           spin up      =   3.7632, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
           spin up      =   3.7632, d =  3.5747, dz2=  0.7023, dxz=  0.7143, dyz=  0.7145, dx2-y2=  0.7217, dxy=  0.7219,
           spin down    =   3.6609, s =  0.1727,
           spin down    =   3.6609, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
           spin down    =   3.6609, d =  3.4882, dz2=  0.6150, dxz=  0.7107, dyz=  0.7109, dx2-y2=  0.7257, dxy=  0.7259,
           polarization =   0.1022, s =  0.0158, p =  0.0000, d =  0.0865,
```

Fe/GD system Löwdin charge of Fe after H_2 adsorption

```
Atom #  14: total charge =   7.4553, s =  0.3487, p =  0.0000, d =  7.1066,
           spin up      =   3.7369, s =  0.1743,
           spin up      =   3.7369, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
           spin up      =   3.7369, d =  3.5626, dz2=  0.6307, dxz=  0.7505, dyz=  0.7197, dx2-y2=  0.7651, dxy=  0.6966,
           spin down    =   3.7184, s =  0.1744,
           spin down    =   3.7184, p =  0.0000, pz=  0.0000, px=  0.0000, py=  0.0000,
           spin down    =   3.7184, d =  3.5440, dz2=  0.6193, dxz=  0.7498, dyz=  0.7190, dx2-y2=  0.7630, dxy=  0.6929,
           polarization =   0.0185, s = -0.0001, p =  0.0000, d =  0.0186,
```

Table S6: Increasing number of H_2, change in total adsorption energy, change in adsorption energy per H_2, Average H-H distance.

Number of H_2	**Adsorption Energy (eV)**	**Average adsorption energy per H_2 (eV / H_2)**	**Average H-H Distance (Å)**
1	-0.580	-0.580	0.843
2	-0.606	-0.303	0.813
3	-0.866	-0.289	0.814
4	-0.902	-0.226	0.799
5	-0.921	-0.184	0.789

Table S7: Desorption Temperature of First adsorbed to last adsorbed H_2 in the Fe/GD system at different atmospheric pressure.

H_2 Number	**Adsorption Energy (eV)**	**Desorption Temperature (K) at Different Pressure (atm)**		
		1 atm	**2 atm**	**3 atm**
1	-0.580	740	801	842
2	-0.300	387	419	440
3	-0.283	368	399	420
4	-0.217	288	312	327
5	-0.178	235	254	267

Table S8: Atomic Cartesian coordinates (in Å) for all optimized geometries. Each structure is labelled by the specific name, followed by the total energy (in Rydberg).

Pristine Graphene (PG)

```
     Final energy   =    -205.1108914153 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C         0.000000000  -0.000000000   1.750025449
C        -1.229996516   2.130420458   1.750000118
C        -2.459999593   4.260848788   1.750000118
C         2.459996922  -0.000002246   1.750000118
C         1.229996516   2.130420458   1.750000118
C         0.000000000   4.260845000   1.749986329
C         4.920003078  -0.000002246   1.750000118
C         3.690000000   2.130422000   1.749986406
C         2.459999593   4.260848788   1.750000118
C         0.000000000   1.420275300   1.750016024
C        -1.230003314   3.550701087   1.749989824
C        -2.460004070   5.681127350   1.750016024
C         2.459998340   1.420279041   1.749994521
C         1.230003314   3.550701087   1.749989824
C         0.000000000   5.681128827   1.749989824
C         4.920001660   1.420279041   1.749994521
C         3.690000000   3.550704917   1.749994521
C         2.460004070   5.681127350   1.750016024
End final coordinates
```

Graphene with Defect (GD)

```
     Final energy   =    -193.1340460413 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C         0.003705404  -0.002141648   1.750025142
C        -1.239189427   2.125109804   1.749991041
C        -2.460008363   4.265127086   1.749949980
C         2.456287969  -0.002135361   1.749949964
C         1.230013150   2.082038573   1.750104375
C        -0.041930814   4.285054088   1.750051607
C         4.920006477   0.010612916   1.749991045
C         3.699204565   2.125108853   1.749943440
C         2.501908122   4.285025494   1.750104380
C        -0.000466681   1.442632936   1.750045970
C        -1.210895376   3.539119776   1.749982968
C        -2.460005382   5.681128003   1.749973324
C         2.460478718   1.442630936   1.749970972
C         0.019585098   5.670371588   1.749982957
C         4.920011438   1.420274598   1.749917873
C         3.670883457   3.539113253   1.749970987
C         2.440408645   5.670353106   1.750045975
End final coordinates
```

PG/Ag

```
     Final energy   =    -278.5707944125 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C         0.000990571   0.000301667   2.876723759
C        -1.229020322   2.130675413   2.876128901
C        -2.458848169   4.261224574   2.876201017
C         2.460994814   0.000135948   2.876057047
C         1.230859825   2.130646762   2.876569411
C         0.000861812   4.261202777   2.875920902
C         4.920991162   0.000274057   2.876107127
C         3.691139383   2.130577629   2.876796163
C         2.460949780   4.261340349   2.876891605
C         0.000857702   1.420508534   2.876266723
C        -1.229008322   3.550999134   2.876022641
C        -2.458991486   5.681422236   2.876370135
C         2.460931545   1.420370583   2.876627742
C         1.230785038   3.551050225   2.876540920
C         0.000952650   5.681396585   2.875973982
C         4.921157821   1.420467554   2.876097291
C         3.691177755   3.551102989   2.877001828
C         2.460990591   5.681625943   2.876370494
Ag        2.599951852   2.910957041   6.589843310
End final coordinates
```

PG/Au

```
     Final energy   =    -306.9421791737 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C         0.027597580   0.003162442   2.863897054
C        -1.202380076   2.133587488   2.864148040
C        -2.432395894   4.263983336   2.863457649
C         2.487649580   0.003134214   2.863715940
C         1.257413755   2.133199764   2.869048550
C         0.027390715   4.264221500   2.864857919
C         4.947593110   0.003186203   2.864075512
C         3.717706042   2.133620594   2.863436737
C         2.487914798   4.264288457   2.864869057
C         0.027338364   1.423241729   2.865149508
C        -1.202368395   3.553882119   2.864155903
C        -2.432408791   5.684299235   2.864062887
C         2.487966531   1.423143934   2.865460528
C         1.257450304   3.554392915   2.868401371
C         0.027594485   5.684324220   2.863744171
C         4.947617868   1.423477275   2.863454384
C         3.717647230   3.553863703   2.863306388
C         2.487672137   5.684334990   2.863720078
Au        1.873352656   2.773072884   6.381009324
End final coordinates
```

PG/Cu

```
     Final energy   =    -326.3613340258 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.050911902  -0.042076927   1.699878064
C       -1.280549940   2.086781557   1.700399761
C       -2.500334051   4.222893993   1.729713196
C        2.409686053  -0.043763199   1.689435618
C        1.177925090   2.086320962   1.699726951
C       -0.051342057   4.218288437   1.692438482
C        4.868127695  -0.043476780   1.692587903
C        3.639636903   2.075093164   1.729650131
C        2.398917214   4.223292725   1.752286442
C       -0.051298478   1.377097564   1.694285709
C       -1.278345050   3.507352670   1.700580827
C       -2.509299310   5.640273928   1.699979678
C        2.407657125   1.374193449   1.699893592
C        1.173747608   3.506492986   1.707044264
C       -0.051088013   5.637793134   1.693637871
C        4.870320485   1.374628596   1.700670009
C        3.641915924   3.505648955   1.805578387
C        2.407108125   5.642712944   1.707210721
Cu       3.404543580   3.643495844   3.900402394
End final coordinates
```

PG/Fe

```
     Final energy   =    -260.6179157026 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.007184232  -0.000894650   1.712852822
C       -1.231915977   2.120391955   1.712857399
C       -2.447797425   4.265068442   1.715734743
C        2.458091332  -0.020239969   1.715598599
C        1.218892030   2.121174475   1.702579352
C       -0.016032593   4.265070486   1.715534707
C        4.923367257  -0.000885037   1.712889140
C        3.697267387   2.121184413   1.702891963
C        2.458078159   4.267564646   1.702738123
C       -0.016044710   1.408188148   1.715509441
C       -1.231918937   3.552851846   1.712882078
C       -2.456643476   5.674145263   1.712877262
C        2.458069963   1.405746726   1.702738249
C        1.218869462   3.552087640   1.702569125
C       -0.007177382   5.674155677   1.712847073
C        4.932194774   1.408208587   1.715708269
C        3.697290519   3.552087864   1.702899645
C        2.458097925   5.693509027   1.715642947
Fe       2.457657924   2.837106460   3.247492062
End final coordinates
```

GD/Ag

```
     Final energy   =    -266.7189655118 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.000693570   0.015366604   1.638657737
C       -1.243050134   2.143341025   1.657117901
C       -2.464743353   4.283249063   1.638658426
C        2.451198843   0.015363557   1.638444214
C        1.225154452   2.060714714   1.812877059
C       -0.080501077   4.322161570   1.813196656
C        4.915262459   0.027153287   1.657486360
C        3.693454190   2.143406446   1.657413922
C        2.531276422   4.322426656   1.815359774
C       -0.012799698   1.442339642   1.683272774
C       -1.235003681   3.559275373   1.683370399
C       -2.464729390   5.698860950   1.644713618
C        2.463196370   1.442401006   1.683348437
Ag       1.224496499   3.567711327   3.416556417
C        0.003046693   5.703485285   1.683494405
C        4.915216711   1.437958519   1.636628264
C        3.685504604   3.559326178   1.684159475
C        2.447503662   5.703572798   1.684198162
End final coordinates
```

GD/Au

```
     Final energy   =    -295.1309478584 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.004035291  -0.002329776   2.834066904
C       -1.237388012   2.126160472   2.883761232
C       -2.459998803   4.265518001   2.834217197
C        2.455953661  -0.002337537   2.834217197
C        1.230008034   2.031209500   3.061319949
C       -0.085989949   4.310491320   3.061988408
C        4.919996586   0.008528971   2.883761232
C        3.697398273   2.126150605   2.883719425
C        2.545924562   4.310444292   3.061319949
C       -0.009715275   1.418431602   2.903609080
C       -1.236477140   3.543222716   2.903928694
C       -2.460014554   5.681133403   2.834838112
C        2.469719856   1.418415187   2.903661821
Au       1.230091305   3.550650285   4.547103155
C        0.003239546   5.690474510   2.903928694
C        4.919998594   1.420280812   2.857288052
C        3.696474904   3.543217764   2.903661821
C        2.456743122   5.690462873   2.903609080
End final coordinates
```

GD/Cu

```
     Final energy   =    -314.6381294996 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.003275292   0.004988753   2.883274967
C       -1.237779787   2.133576508   2.912373903
C       -2.460902983   4.272621749   2.879991811
C        2.454925204   0.004981125   2.883293216
C        1.229103240   2.023154527   3.133508330
C       -0.089963557   4.323738431   3.082056917
C        4.919099525   0.019896878   2.902106549
C        3.695979360   2.133588716   2.912346107
C        2.548139399   4.323742318   3.081987768
C       -0.011867493   1.423727724   2.946287076
C       -1.236587313   3.550370449   2.930772834
C       -2.460903645   5.687390953   2.890800616
C        2.470069554   1.423725625   2.946270078
Cu       1.229157904   3.571715141   4.191614104
C        0.002963267   5.701447587   2.926310647
C        4.919096336   1.429681551   2.882259531
C        3.694775706   3.550384144   2.930731583
C        2.455235991   5.701459821   2.926290961
End final coordinates
```

GD/Fe

```
     Final energy   =    -249.0792117970 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.000797102   0.008427507   2.855564820
C       -1.235048053   2.138876622   2.921689360
C       -2.462981443   4.273000561   2.855379409
C        2.454808445   0.008404537   2.855493615
C        1.227011503   2.056523646   3.155146359
C       -0.075356555   4.312283488   3.154795879
C        4.917009005   0.012044085   2.921540243
C        3.689055960   2.138879153   2.921599411
C        2.529418236   4.312307916   3.154684357
C       -0.008995950   1.417949251   2.944875281
C       -1.246378615   3.561140717   2.944636122
C       -2.463000509   5.690783224   2.844845324
C        2.463001001   1.417940650   2.944769304
Fe       1.227136598   3.560614278   4.043719089
C       -0.010374637   5.702001222   2.944575131
C        4.917001930   1.429938438   2.861423400
C        3.700413382   3.561137772   2.944527751
C        2.464389804   5.702025932   2.944575143
End final coordinates
```

PG – H_2

```
     Final energy   =    -207.4444676858 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.003105178  -0.006092731   1.510572205
C       -1.233102014   2.124370668   1.510676460
C       -2.463133086   4.254770552   1.510551380
C        2.456884726  -0.006099061   1.510896987
C        1.226898513   2.124399221   1.510586769
C       -0.003084585   4.254756647   1.510591646
C        4.916906573  -0.006095200   1.510644937
C        3.686878669   2.124414440   1.510631663
C        2.456895071   4.254765757   1.509419927
C       -0.003096945   1.414214391   1.510711099
C       -1.233113540   3.544667617   1.510730516
C       -2.463103886   5.675027320   1.510729164
C        2.456883986   1.414220775   1.510781626
C        1.226955427   3.544686253   1.510033997
C       -0.003113925   5.675021153   1.510783983
C        4.916886615   1.414221403   1.510787605
C        3.686834739   3.544700789   1.509999938
C        2.456903904   5.674971158   1.510054756
H        2.515409351   4.750691710   5.094842740
H        2.595741585   4.008204138   5.185371603
End final coordinates
```

GD – H_2

```
     Final energy   =    -195.4695737127 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.048140973   0.009546301   1.593388362
C       -1.286720549   2.140481479   1.572832062
C       -2.511258814   4.277606241   1.529529581
C        2.405413321   0.009341749   1.600043708
C        1.176873020   2.088649512   1.772474220
C       -0.076246921   4.298356004   1.493524725
C        4.868511430   0.027719425   1.576393404
C        3.642358779   2.141299207   1.573053536
C        2.435555471   4.301687960   1.463117947
C       -0.051056741   1.451228456   1.639395939
C       -1.255671790   3.555021294   1.521035991
C       -2.511244240   5.692377289   1.574557595
C        2.406779975   1.452147593   1.643447043
C       -0.031757828   5.684812586   1.571106890
C        4.867733022   1.438616007   1.573938118
C        3.612787878   3.555705932   1.513048604
C        2.389625921   5.686258304   1.558083959
H        2.420393019   3.297474992   5.287770718
H        2.077580021   3.207237671   4.623267598
End final coordinates
```

GD/Ag – H_2

```
     Final energy   =    -269.0798710588 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C       -0.024441807   0.050176009   1.536427377
C       -1.252484491   2.182029313   1.614991695
C       -2.487200426   4.316116990   1.584509236
C        2.428765941   0.049771465   1.575020002
C        1.236190240   2.147670751   1.515297133
C       -0.139943313   4.371151030   2.012053593
C        4.893420321   0.040912123   1.626842675
C        3.694026787   2.162123766   1.554685504
C        2.490532164   4.301019846   1.613536148
C       -0.014271591   1.496799923   1.548560487
C       -1.264765369   3.600038170   1.709878764
C       -2.492929666   5.736246142   1.567371212
C        2.452623223   1.476773596   1.533401895
Ag       1.266438794   3.546956721   3.418617522
C       -0.032201622   5.733837612   1.712554228
C        4.909126234   1.458172276   1.583477400
C        3.672614877   3.579523447   1.566536161
C        2.419857069   5.707646814   1.578582033
H        1.389875395   3.278705915   5.402377769
H        2.085429239   2.998713091   5.147580165
End final coordinates
```

GD/Au – H_2

```
     Final energy   =    -297.4872088845 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.100728965   0.055479431   2.877976549
C       -1.163058911   2.172400146   2.870890430
C       -2.364457298   4.325351572   2.878232657
C        2.556535790   0.059096523   2.833441148
C        1.290347694   2.139910727   2.892511757
C        0.035455215   4.313416492   2.892872235
C        5.019531736   0.050719967   2.968233851
C        3.780816158   2.196251364   2.968188297
C        2.686983267   4.395213773   3.423379954
C        0.076585004   1.477830593   2.848526453
C       -1.144761426   3.593250941   2.848871005
C       -2.354320689   5.745485479   2.859669608
C        2.544714287   1.502747592   2.884994140
Au       1.291917723   3.589976614   4.685046133
C        0.110852743   5.718307893   2.885162133
C        5.001283595   1.470529390   2.917752971
C        3.801398861   3.617022189   3.062585335
C        2.570255097   5.749394829   3.062491669
H        0.647462688   3.218842905   6.328456380
H        0.053329500   2.875138582   5.820611296
End final coordinates
```

GD/Cu – H_2

```
     Final energy   =    -316.9900005314 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.041679550  -0.090984860   2.895403768
C       -1.194895030   2.035524390   2.954587880
C       -2.423371791   4.170375397   2.855515464
C        2.489664735  -0.091653381   2.879767333
C        1.247438141   1.901713037   3.409091475
C        0.008329537   4.213426128   2.837168114
C        4.953264862  -0.060131570   2.889890980
C        3.708140344   2.043417675   2.986156514
C        2.544422755   4.245220335   3.075309796
C        0.027258088   1.311409654   3.049851815
C       -1.173034015   3.454533071   2.872976887
C       -2.421731197   5.580195919   2.875076738
C        2.483916514   1.317204161   3.061692704
Cu       1.199428542   3.555940245   4.287412742
C        0.041422462   5.601409380   2.839272369
C        4.946080174   1.345312871   2.916783619
C        3.706090447   3.463582747   2.946248189
C        2.492793593   5.616477955   2.913380133
H        0.784467037   4.071962938   5.897343010
H        0.987207252   4.766684907   5.530175470
End final coordinates
```

GD/Fe – H_2

```
     Final energy   =    -251.4529787559 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.021675225  -0.003412986   2.860666826
C       -1.210496997   2.128118543   2.933836065
C       -2.432747954   4.261649710   2.873208966
C        2.480826943  -0.010356069   2.873181212
C        1.253226679   2.043568737   3.124361892
C       -0.044506054   4.295643498   3.232956001
C        4.939635705  -0.002083753   2.933845520
C        3.715756715   2.124649981   2.914578450
C        2.554707164   4.297811582   3.124445789
C        0.012371637   1.408897281   2.934667420
C       -1.213994778   3.552558983   2.982162786
C       -2.439765052   5.678565799   2.864522047
C        2.491533234   1.401825158   2.932441626
Fe       1.328858827   3.502705604   4.087989889
C        0.014285682   5.679990691   2.982149528
C        4.943616322   1.415749819   2.872917741
C        3.729626519   3.546280010   2.932475183
C        2.483926279   5.689753954   2.934697757
H        1.245121315   3.551399933   5.820318505
H        0.561175591   3.946122524   5.526267797
End final coordinates
```

GD/Fe – $2H_2$

```
     Final energy   =    -253.7921752392 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.091048465   0.119644771   2.818352641
C       -1.144238687   2.251436410   2.932309375
C       -2.367818753   4.385133100   2.869554501
C        2.547627979   0.116411422   2.869517467
C        1.304646362   2.171980676   3.172316954
C        0.023082980   4.419749396   3.259807110
C        5.007258081   0.123556380   2.932248418
C        3.774381671   2.253999197   2.954653389
C        2.610531979   4.433815209   3.172329693
C        0.082559962   1.529183061   2.921398868
C       -1.145367576   3.675196807   2.984353488
C       -2.370421035   5.801683134   2.843305985
C        2.545748846   1.531134651   2.963607194
Fe       1.372729998   3.640620553   4.201905956
C        0.083670594   5.803957735   2.984304770
C        5.003498581   1.544305382   2.878429973
C        3.786113013   3.679461429   2.963658198
C        2.556140414   5.813549325   2.921330162
H        0.657805040   2.564483437   5.413077824
H        0.162070514   3.208826740   5.440637715
H        1.140717938   4.904782406   5.440297019
H        1.946734634   4.798188779   5.412820300
End final coordinates
```

GD/Fe – $3H_2$

```
     Final energy   =    -256.1363897153 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.137216567   0.092311602   2.834267261
C       -1.094394437   2.225204266   2.946158478
C       -2.325473294   4.358275896   2.834112667
C        2.591275646   0.092442857   2.834073765
C        1.364303160   2.146551939   3.244748638
C        0.066971282   4.393951366   3.244868799
C        5.054255338   0.092570237   2.945728839
C        3.823097420   2.225250427   2.945485254
C        2.662067291   4.393841908   3.244007443
C        0.137922745   1.506313339   2.962145118
C       -1.100575366   3.651863386   2.961930462
C       -2.325695103   5.775185247   2.820597693
C        2.590677418   1.506433783   2.961394892
Fe       1.365311036   3.645196121   4.331922506
C        0.125509058   5.776118868   2.961798863
C        5.054336506   1.514357079   2.878822842
C        3.829694430   3.651804513   2.961275807
C        2.602974784   5.776060972   2.961365795
H        0.389721519   2.612596102   5.370097510
H       -0.015763499   3.318660937   5.372025187
H        2.748557433   3.315318148   5.368144419
H        2.341791189   2.609997452   5.366261021
H        0.960399974   5.008922352   5.366213698
H        1.774683904   5.007166202   5.367186043
End final coordinates
```

GD/Fe – $4H_2$

```
     Final energy   =    -258.4701823105 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.133483092   0.289188935   2.737051484
C       -1.098047568   2.421864045   2.849514838
C       -2.329234986   4.554766612   2.736687966
C        2.587933640   0.289139016   2.736820135
C        1.360725755   2.343243312   3.148865686
C        0.063285563   4.590557639   3.148718805
C        5.050726994   0.289300580   2.849255900
C        3.819454192   2.421855532   2.849140322
C        2.658323736   4.590544416   3.148352397
C        0.134258102   1.703092035   2.865590976
C       -1.104326294   3.848494612   2.865325917
C       -2.329294564   5.971874194   2.722562163
C        2.587131277   1.703092993   2.865198215
Fe       1.361124331   3.841647558   4.235130411
C        0.122057701   5.972743199   2.865220300
C        5.050706831   1.711006904   2.782036279
C        3.825854133   3.848461981   2.864949501
C        2.599392323   5.972799998   2.865155353
H        0.384474047   2.809848806   5.274814589
H       -0.022116941   3.514836466   5.273285677
H        2.743473659   3.513003617   5.275941029
H        2.337642876   2.807740397   5.273430658
H        0.955499990   5.204037128   5.275434511
H        1.769128636   5.204898831   5.272567247
H        1.460856507  -0.545557333   5.826047552
H        1.479983966  -0.552978472   6.579414089
End final coordinates
```

GD/Fe – $5H_2$

```
     Final energy   =    -260.8027232080 Ry
Begin final coordinates

ATOMIC_POSITIONS (angstrom)
C        0.128572376   0.274722173   2.521041409
C       -1.102801635   2.407519113   2.632950556
C       -2.334088298   4.540452577   2.520606283
C        2.583151117   0.274664794   2.521008070
C        1.355944404   2.328725948   2.933263293
C        0.058508200   4.576089433   2.933831656
C        5.045838632   0.274880505   2.633514310
C        3.814569063   2.407464358   2.632677943
C        2.653342539   4.576062676   2.933626332
C        0.129432030   1.688728838   2.649340026
C       -1.109064384   3.834205931   2.649403459
C       -2.334155232   5.957533772   2.506739001
C        2.582334634   1.688676610   2.649132042
Fe       1.356018033   3.826146616   4.020875336
C        0.117249847   5.958240402   2.649953664
C        5.045901786   1.696609061   2.565533543
C        3.820850484   3.834161732   2.649026306
C        2.594439875   5.958257975   2.649779826
H        0.379816583   2.793964225   5.061570018
H       -0.029572031   3.497201482   5.054971281
H        2.740306173   3.496613328   5.056732107
H        2.330927934   2.793285504   5.061322892
H        0.948248940   5.186218410   5.061874870
H        1.761816809   5.186288197   5.063333610
H        1.444160507  -0.540085868   5.633036805
H        1.467393020  -0.537337752   6.386327580
H        1.013601307   3.752113132   8.697824110
H        1.740539290   3.940685828   8.746493672
End final coordinates
```

Table S9: Final energy (ground state energy) of free transition metal atom and H_2 (in Rydberg).

Atom/Molecule	Final Energy (Ry)
Ag	-73.4440623648
Au	-101.8092043249
Cu	-121.2143242825
Fe	-55.2099534839
H_2	-2.33116958