Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supporting information

for

Multiscale Modeling Reveals Aluminum Nitride as Efficient Propane Dehydrogenation Catalyst

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1. Microkinetic modeling

Microkinetic modeling was performed using the fp_echem software package available on the Mpourmpakis group Github.[1] A full reaction network was constructed which simultaneously considered all four propane dehydrogenation mechanisms investigated in this work (presented in Fig. 3 of the main manuscript). A total of seventeen elementary steps were included. Reaction networks for the AlN and Ga/AlN surfaces were identical, while only elementary reaction energies, barriers, and state vibrational frequencies differed. The total set of elementary steps is as follows:

1. Concerted C_3H_8 Adsorption: $C_3H_{8,(g)} + X + Y + N \leftrightarrow C_3H_8 - X - Y - N$
2. PS C_3H_8 Adsorption: $C_3H_{8,(g)} + X \leftrightarrow C_3H_{8,v} - X$
3. SP C_3H_8 Adsorption: $C_3H_{8,(g)} + X \leftrightarrow C_3H_{8,h} - X$
4. Concerted C_3H_6 Formation: $C_3H_8 - X - Y - N \leftrightarrow C_3H_6 - Y + H - X + H - N$
5. PS C_3H_7- Formation: $C_3H_{8,v} - X + N \leftrightarrow C_3H_{7,v} - X + H - N$
6. SP C_3H_7- Formation: $C_3H_{8,h} - X + N \leftrightarrow C_3H_{7,h} - X + H - N$
7. PSD C_3H_6 Formation: $C_3H_{7,v} - X + H - N + Y \leftrightarrow C_3H_{6,v} - X + H_2 - Y_a + N$
8. PSI C_3H_6 Formation: $C_3H_{7,v} - X + Y \leftrightarrow C_3H_{6,h} - X + H - Y$
9. SP C_3H_6 Formation: $C_3H_{7,h} - X + Y \leftrightarrow C_3H_{6,h} - X + H - Y$
10. PSD C_3H_6 Desorption: $C_3H_{6,v} - X \leftrightarrow C_3H_{6,(g)} + X$
11. PSI C_3H_6 Desorption: $C_3H_{6,h} - X \leftrightarrow C_3H_{6,(g)} + X$
12. Concerted C_3H_6 Desorption: $C_3H_6 - Y \leftrightarrow C_3H_{6,(g)} + Y$
13. Concerted H_2 Formation: $H - X + H - N \leftrightarrow H_2 - X_b + N$
14. Concerted H_2 Desorption: $H_2 - X_b \leftrightarrow H_{2,(g)} + X$

15. PSD H_2 Desorption: $H_2 - Y_a \leftrightarrow H_{2,(g)} + Y$

16. PSI H_2 Formation: $H - Y + H - N \leftrightarrow H_2 - Y_b + N$

17. PSI H_2 Desorption: $H_2 - Y_b \leftrightarrow H_{2,(g)} + Y$

In the above steps, X and Y indicate either two different Al atoms or a Ga atom and an Al atom, respectively for the AlN and Ga/AlN surfaces. N is a nitrogen site. Each model considers three total surface sites that are involved in adsorption, surface reactions, and desorption. These are two metal sites (Al and Al for AlN or Ga and Al for Ga/AlN) and one nitrogen site. The subscripts *v* and *h* respectively denote vertical and horizontal adsorption configurations of alkanes, carbocations, and alkenes. These help to differentiate activation pathways which start with either the primary or secondary carbons of propane. The subscripts *a* and *b* identify H_2 adsorption states with or without a neighboring adsorbed alkene. We note many of these steps are shared between mechanisms, as shown by the reaction coordinate diagram in Fig. 3 of the main manuscript.

2. Ab initio molecular dynamics

2.1. Methods

Ab initio molecular dynamics (AIMD) and metadynamics simulations were performed using CP2K in the NVT ensemble at 873.15 K (typical dehydrogenation conditions)[2] using a velocity-scaling thermostat with a time constant of 100 fs. A time step of 0.5 fs was used for the integration of the equations of motion. To reduce the computational cost of AIMD and metadynamics simulations, the SCF and cutoff criteria were reduced to 10^{-6} Hartree and 350 Ry, respectively (compared to the density function theory (DFT) reaction profile calculations). Additionally, a reduced Ga/AlN slab model with four layers was used where the bottom two layers were fixed at their optimized bulk position, whereas the top two layers were allowed to relax during the simulations. The system was first thermally equilibrated using the NVT ensemble for 2 ps, starting from a DFT-optimized structure. From the equilibrated structure, a metadynamics simulation was performed to accelerate sampling of the relevant configuration space with a history-dependent biasing potential along the collective variables (CVs). Two CVs were defined for metadynamics simulations of H_2 formation: (i) the sum of the distance between gallium and hydrogen and the distance between nitrogen and the other hydrogen ($CV1 = d_{M\text{-}Ha} + d_{N\text{-}Hb}$) and (ii) the distance between two hydrogen atoms ($CV2 = d_{Ha\text{-}Hb}$). The biasing potential has the form of Gaussian hills of 1.5 kcal/mol height and 0.05 Å width, which were applied at intervals of 50 time steps (25 fs). The free energy surface was constructed from the sum of the Gaussian potentials.[3] Quadratic walls were used to avoid the sampling of non-relevant parts of the configuration space by limiting the range of the CVs.

2.2. Discussion

We performed AIMD simulations to examine molecular H_2 formation ($H^+ + H^- \rightarrow H_2$) under dehydrogenation conditions at 873.15 K. Fig. S1 shows the free energy surface for molecular H_2 formation from two hydrogen atoms adsorbed on the Ga_a-N_a site pair on the Ga/AlN surface. Note that the AIMD simulation was used to compute the minimum activation free energy for a forward reaction. Therefore, the simulation was stopped 2.5 ps after a first reaction event occurred, and the final state basin was not completely sampled. In the initial state (marked as (I) in Fig. S1), the two hydrogen atoms fluctuated while adsorbed on the surface, leading to a relatively larger variation of the hydrogen-hydrogen distance (CV2) than the sum of the hydrogen-metal and hydrogen-nitrogen distances (CV1). At around 20 ps, the two hydrogen atoms were combined on the Ga atom and desorbed from the surface as molecular H_2 with a free energy barrier of 92 kJ mol^{-1}, which is accessible at typical dehydrogenation temperatures. AIMD simulations, therefore, confirmed that the poisoning of the doped active site of Ga/AlN by hydrogen adsorption is unlikely under operating conditions.

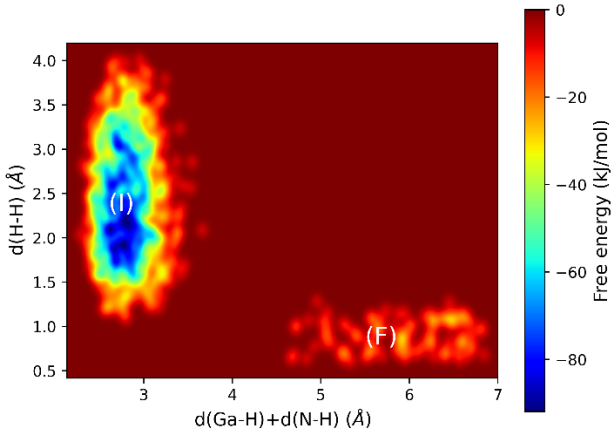

Fig. S1 Free energy surface of metadynamics simulation for molecular H_2 formation from two hydrogen atoms adsorbed on the Ga_a-N_a site pair on Ga/AlN. (I) and (F) indicate initial and final

states, respectively. The final state basin was not completely sampled since reaction events were confirmed.

3. Electronic energy propane dehydrogenation reaction profiles

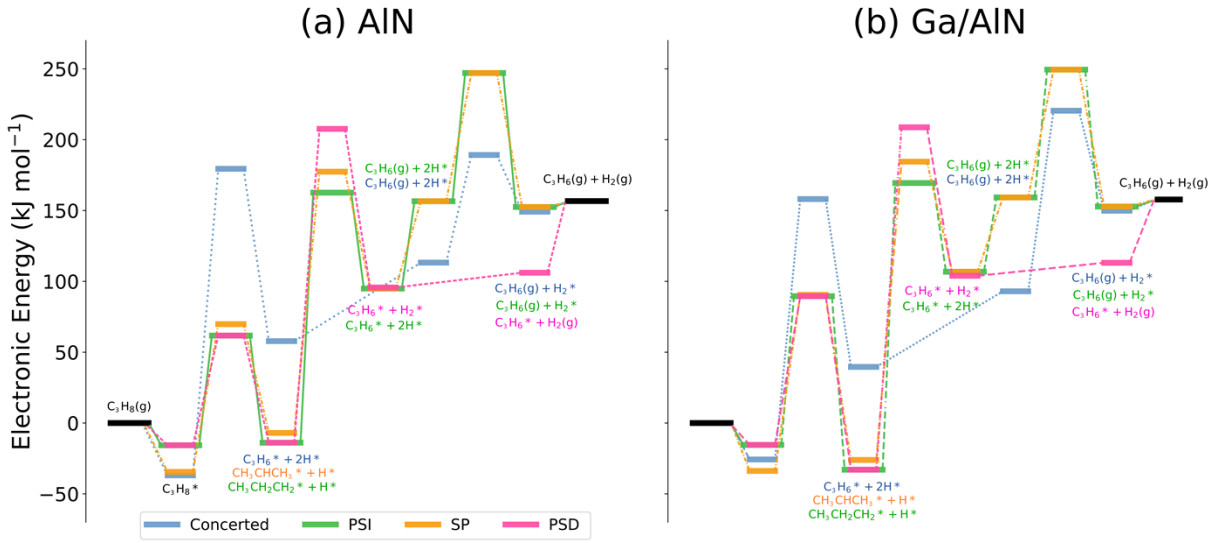

Fig. S2 Electronic energy profiles on (a) undoped AlN and (b) Ga/AlN via the four PDH mechanisms shown in Fig. 3 following the same notation. Concerted, stepwise PSD, stepwise PSI, and stepwise SP mechanisms are depicted in light blue, pink, green, and orange, respectively. Adsorbed states are denoted with asterisks.

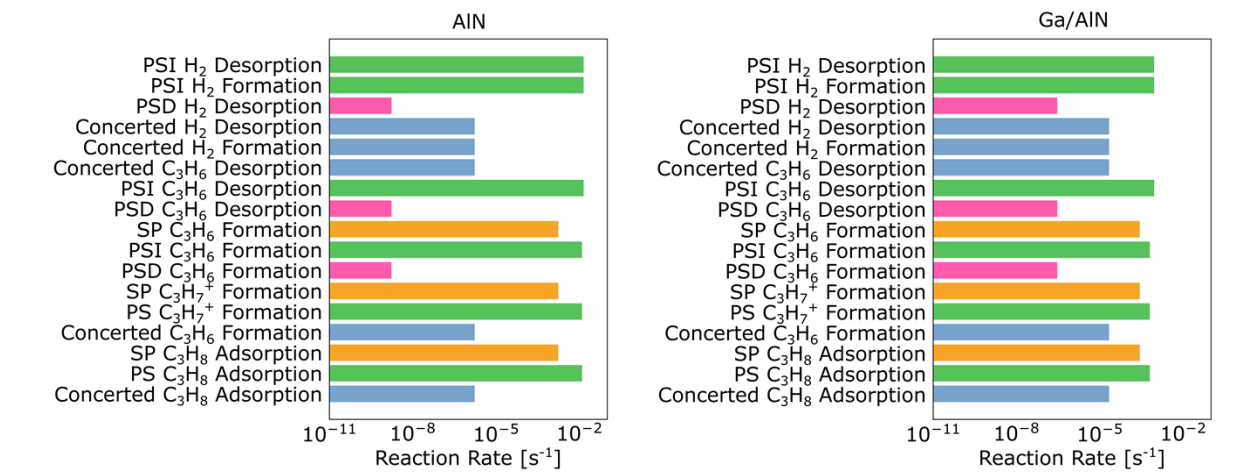

Fig. S3. Reaction rates for each elementary step in the PDH reaction network at 873.15 K on AlN and Ga/AlN. The green bars represent the PSI steps, the pink bars the PSD steps, the blue bars the concerted steps, and the orange bars the SP steps.

4. Propane dehydrogenation on metal nitrides vs metal oxides

Table S1. Comparison between the apparent turnover frequencies (TOFs) of the nitride catalysts and metal oxide catalysts reported in literature. Green, yellow, and red shading indicate catalyst with the highest, second highest, and lowest TOFs, respectively. References 4-7 are based on theoretical results, whereas reference 8 is based on experiments.

Catalyst	Temperature [K]	Pressure [bar]	TOF: Catalyst [s^{-1}]	TOF: AlN [s^{-1}]	TOF: Ga/AlN [s^{-1}]
β-Ga_2O_3[4]	895.00	1.00	1.78×10^{-5}	2.47×10^{-2}	1.72×10^{-3}
Pt/β-Ga_2O_3[4]	895.00	1.00	5.04×10^{-5}	2.47×10^{-2}	1.72×10^{-3}
Cr_2O_3[5]	850.00	0.35	6.28×10^{-5}	3.47×10^{-3}	2.32×10^{-4}
V_2O_3[6]	850.00	0.35	7.03×10^{-3}	3.47×10^{-3}	2.32×10^{-4}
ZnO[5]	850.00	0.35	2.52×10^{-4}	3.47×10^{-3}	2.32×10^{-4}
TiO_2[7]	850.00	1.01	$\sim 5.5 \times 10^{-4}$	7.41×10^{-3}	4.50×10^{-4}
V/TiO_2[7]	850.00	1.01	5.67×10^{-3}	7.41×10^{-3}	4.50×10^{-4}
Al_2O_3[8]	873.15	1.01	$\sim 8.5 \times 10^{-2}$	1.40×10^{-2}	9.20×10^{-4}

5. The relaxed Cartesian coordinates of relevant transition state structures and their total energies

Table S2. transition state (TS) of C-H activation of propane on AlN through the concerted mechanism.

Total energy: -797.137 Hartree

Al	1.77279148	1.96607564	6.71278482
Al	5.75695623	1.96562794	6.71376307
N	3.76447788	1.96518975	6.71040935
N	7.75125460	1.96683206	6.70829006
Al	3.76906638	3.89900517	6.72546978
Al	7.74652181	3.89894823	6.72081709
N	1.77463456	3.90012862	6.71928851
N	5.75501130	3.89893126	6.72798619
Al	1.77124618	7.79716468	6.70731407
Al	5.75754430	7.79620802	6.70749828
N	3.76341208	7.79799002	6.70527967
N	7.75135781	7.79821961	6.70302364
N	1.77142713	9.73108654	6.72048191
N	5.75754467	9.72973926	6.71999577
Al	3.76532147	9.73160121	6.71967723
Al	7.74959123	9.73182448	6.71937344
Al	1.77666862	4.89335876	8.36650012
Al	5.75411203	4.89740721	8.39562173
N	7.74399306	4.88734390	8.37743835
N	3.77215493	4.89051949	8.39791328
N	1.77090313	6.82943897	8.37855246
N	5.75775533	6.83084658	8.37524015
Al	7.74741777	6.82605768	8.36701577
Al	3.76760514	6.82553533	8.36935555
Al	1.77249870	10.74298819	8.36460850
Al	5.75698956	10.74050053	8.36825946
N	3.76734355	10.73484014	8.37101369
N	7.74815956	10.73763550	8.37023275
N	1.77242832	1.01300237	8.39278908
N	5.75822868	1.01724147	8.39742189
Al	3.76748967	1.00596508	8.38451681
Al	7.74921476	1.00848421	8.38304970
Al	1.77195229	7.84787365	10.04551720
Al	5.75536902	7.81943550	10.05330607
N	3.76956838	7.82311595	10.02307361
N	7.74255717	7.84321134	10.00850712
N	1.77452756	9.77356648	10.02610529
N	5.75439479	9.76310736	10.03616390
Al	3.76739783	9.76340842	10.06141718
Al	7.74740624	9.76783247	10.05635866

Al	1.77386394	1.99804546	10.07828943
Al	5.75935808	2.00226494	10.09415861
N	3.76771758	1.98804722	10.05864088
N	7.75085612	2.00141958	10.05115663
N	1.78829682	3.92954727	10.02186833
N	5.74441543	3.94840920	10.10458554
Al	3.77191551	3.91693424	10.10040948
Al	7.75507820	3.93554156	10.06352719
Al	1.77107914	10.80647587	11.63705805
Al	5.75928461	10.80581814	11.63846036
N	3.76344393	10.69732256	11.75252902
N	7.75274019	10.70228347	11.74612582
N	1.77178312	1.04589422	11.76161686
N	5.75969379	1.03853600	11.76602913
Al	3.76628442	0.93329777	11.65553402
Al	7.75142632	0.94204732	11.65320155
Al	1.71341980	5.07000401	11.55571571
Al	5.88327052	4.93226196	11.77764279
N	3.63619177	4.90477323	11.81431306
N	7.82323049	4.94625766	11.68748813
N	1.77018224	6.96298565	11.73539152
N	5.75064700	6.86344162	11.74310632
Al	3.76995180	6.83563235	11.65998826
Al	7.74824317	6.83436669	11.61962050
Al	1.77195765	1.94403754	0.00000000
Al	1.77195765	7.77595756	0.00000000
Al	3.76418364	3.88730355	0.00000000
Al	5.75641122	1.94383752	0.00000000
Al	5.75641122	7.77575753	0.00000000
Al	7.75032740	3.88718396	0.00000000
N	1.77213757	3.88747394	0.00000000
N	3.76478373	1.94379783	0.00000000
N	3.76478373	7.77570726	0.00000000
N	5.75646149	3.88735382	0.00000000
N	7.75006757	1.94370787	0.00000000
N	7.75006757	7.77562735	0.00000000
N	1.77213757	9.71938496	0.00031010
N	5.75646149	9.71926484	0.00031010
Al	3.76418364	9.71921510	0.00031010
Al	7.75032740	9.71910503	0.00031010
Al	1.77195765	4.85982187	1.68307761
Al	5.75641122	4.85962184	1.68307761
N	7.75006757	4.85949166	1.68308767
N	3.76478373	4.85958162	1.68308767
N	1.77213757	6.80325932	1.68315752
N	5.75646149	6.80313920	1.68315752

Al	7.75032740	6.80297939	1.68315752
Al	3.76419370	6.80308946	1.68315752
Al	1.77195765	10.69131325	1.68317763
Al	5.75641122	10.69111322	1.68317763
N	3.76478373	10.69107300	1.68317763
N	7.75006757	10.69098304	1.68317763
N	1.77213757	0.97153828	1.68324801
N	5.75646149	0.97141816	1.68324801
Al	3.76419370	0.97136788	1.68325753
Al	7.75032740	0.97125782	1.68325753
Al	1.77195765	7.77517755	3.36602399
Al	5.75641122	7.77497752	3.36602399
N	3.76478373	7.77493731	3.36602399
N	7.75006757	7.77484735	3.36602399
N	1.77213757	9.71861501	3.36610442
N	5.75646149	9.71849489	3.36610442
Al	3.76418364	9.71844514	3.36610442
Al	7.75032740	9.71832502	3.36610442
Al	1.77195765	1.94389784	3.36636372
Al	5.75641122	1.94369781	3.36636372
N	3.76478373	1.94365760	3.36637430
N	7.75006757	1.94356764	3.36637430
N	1.77213757	3.88732366	3.36644415
N	5.75646149	3.88720354	3.36644415
Al	3.76418364	3.88715379	3.36645368
Al	7.75032740	3.88704372	3.36645368
Al	1.77195765	10.69096293	5.04921167
Al	5.75641122	10.69076290	5.04922172
N	7.75006757	10.69064331	5.04922172
N	3.76478373	10.69072321	5.04922172
N	1.77213757	0.97119802	5.04929157
N	5.75646149	0.97107790	5.04930163
Al	7.75032740	0.97091808	5.04930163
Al	3.76419370	0.97102815	5.04930163
Al	1.77195765	4.85925194	5.04931168
Al	5.75641122	4.85905191	5.04931168
N	3.76478373	4.85901170	5.04932174
N	7.75006757	4.85892174	5.04932174
N	1.77213757	6.80267935	5.04939159
N	5.75646149	6.80255922	5.04939159
Al	3.76419370	6.80251953	5.04940164
Al	7.75032740	6.80239941	5.04940164
C	5.07357917	4.34189917	14.62536881
C	3.72494030	4.74561170	14.60035950
C	3.35439623	6.19611131	14.80673162
H	5.63823085	4.20845930	13.32147360

H	3.65137498	4.68204774	12.94543330
H	5.83205154	5.05614692	14.96653301
H	5.31598984	3.31136462	14.89743115
H	2.95711615	3.99340225	14.79847388
H	4.00360199	6.86974196	14.20447064
H	3.47995698	6.54552028	15.84694505
H	2.31800358	6.41408714	14.51336095

Table S3. TS of H_2 production on AlN through the concerted mechanism.
Total energy: -776.518 Hartree

Al	1.7717867184	1.9455190189	6.7169032186
Al	5.7563445026	1.9448173008	6.7173127648
N	3.7637835227	1.9468985361	6.7170786523
N	7.7504781304	1.9472501994	6.7160211785
Al	3.7669917014	3.8792473212	6.7163291569
Al	7.7469799613	3.8793465742	6.7131720076
N	1.7722831030	3.8789543957	6.7101756854
N	5.7558455118	3.8771198438	6.7189749898
Al	1.7712779297	7.7763907121	6.7171168446
Al	5.7571235960	7.7756001612	6.7174909164
N	3.7638355012	7.7776518949	6.7176215641
N	7.7506425972	7.7779290454	6.7159691629
N	1.7713762745	9.7086798527	6.7125947543
N	5.7571643740	9.7079581499	6.7127034916
Al	3.7649409890	9.7105581755	6.7144302512
Al	7.7494832324	9.7105695410	6.7138916295
Al	1.7729125839	4.8559220451	8.3671976785
Al	5.7554344231	4.8589319009	8.3889680767
N	7.7432676363	4.8534598407	8.3780954673
N	3.7701823042	4.8534936491	8.3888956495
N	1.7708308898	6.7919517099	8.3823211208
N	5.7575143195	6.7927372052	8.3812768510
Al	7.7480417702	6.7888977940	8.3721643315
Al	3.7664984541	6.7878541294	8.3736387092
Al	1.7722534176	10.6919603861	8.3714561734
Al	5.7560919358	10.6903834615	8.3745452293
N	3.7662188818	10.6857737920	8.3819799590
N	7.7482015979	10.6867730619	8.3809128119
N	1.7717556907	0.9603836924	8.3797114554
N	5.7565572251	0.9647759595	8.3831956538
Al	3.7655382780	0.9585121299	8.3747459528
Al	7.7487327257	0.9575061716	8.3730312765
Al	1.7739094740	7.7788360431	10.0692662194
Al	5.7560749863	7.7576674968	10.0763543501
N	3.7700459199	7.7597461581	10.0451864085
N	7.7440605996	7.7738675122	10.0360507817

N	1.7730766652	9.7072054393	10.0318128012
N	5.7551746562	9.7007260212	10.0417248053
Al	3.7666730080	9.7007725190	10.0687005359
Al	7.7476458944	9.7046882742	10.0644253633
Al	1.7726943418	1.9394173676	10.0655018262
Al	5.7567172097	1.9407831081	10.0790106049
N	3.7666005732	1.9373829167	10.0453745667
N	7.7478916904	1.9359823704	10.0407086628
N	1.7698782543	3.8654223079	10.0096947338
N	5.7543499610	3.8825274891	10.0851446907
Al	3.7606793547	3.8755186137	10.0788010510
Al	7.7626233025	3.8687438425	10.0511332526
Al	1.7707831904	10.7491308515	11.6399989594
Al	5.7573220141	10.7512135027	11.6426313757
N	3.7632319349	10.6433739075	11.7553476942
N	7.7510335446	10.6447453342	11.7514190845
N	1.7708760760	0.9889012226	11.7497518353
N	5.7574986524	0.9871164641	11.7576175277
Al	3.7628072072	0.8794492876	11.6409440160
Al	7.7509648017	0.8834977671	11.6416337857
Al	1.7444109445	4.9516290842	11.5986673530
Al	5.8827095208	4.8895864273	11.7082293043
N	3.6617512220	4.8161638185	11.7614779307
N	7.8061422463	4.8418656872	11.7165224383
N	1.7694654625	6.8602540327	11.7546553361
N	5.7569869167	6.7843058431	11.7670496915
Al	3.7630679683	6.7106202167	11.6454051674
Al	7.7672181603	6.7446801885	11.6401223829
Al	1.7719600000	1.9440400000	0.0000000000
Al	1.7719600000	7.7759700000	0.0000000000
Al	3.7641900000	3.8873100000	0.0000000000
Al	5.7564200000	1.9438400000	0.0000000000
Al	5.7564200000	7.7757700000	0.0000000000
Al	7.7503400000	3.8871900000	0.0000000000
N	1.7721400000	3.8874800000	0.0000000000
N	3.7647900000	1.9438000000	0.0000000000
N	3.7647900000	7.7757200000	0.0000000000
N	5.7564700000	3.8873600000	0.0000000000
N	7.7500800000	1.9437100000	0.0000000000
N	7.7500800000	7.7756400000	0.0000000000
N	1.7721400000	9.7194000000	0.0003100000
N	5.7564700000	9.7192800000	0.0003100000
Al	3.7641900000	9.7192300000	0.0003100000
Al	7.7503400000	9.7191200000	0.0003100000
Al	1.7719600000	4.8598300000	1.6830800000
Al	5.7564200000	4.8596300000	1.6830800000

N	7.7500800000	4.8595000000	1.6830900000
N	3.7647900000	4.8595900000	1.6830900000
N	1.7721400000	6.8032700000	1.6831600000
N	5.7564700000	6.8031500000	1.6831600000
Al	7.7503400000	6.8029900000	1.6831600000
Al	3.7642000000	6.8031000000	1.6831600000
Al	1.7719600000	10.6913300000	1.6831800000
Al	5.7564200000	10.6911300000	1.6831800000
N	3.7647900000	10.6910900000	1.6831800000
N	7.7500800000	10.6910000000	1.6831800000
N	1.7721400000	0.9715400000	1.6832500000
N	5.7564700000	0.9714200000	1.6832500000
Al	3.7642000000	0.9713700000	1.6832600000
Al	7.7503400000	0.9712600000	1.6832600000
Al	1.7719600000	7.7751900000	3.3660300000
Al	5.7564200000	7.7749900000	3.3660300000
N	3.7647900000	7.7749500000	3.3660300000
N	7.7500800000	7.7748600000	3.3660300000
N	1.7721400000	9.7186300000	3.3661100000
N	5.7564700000	9.7185100000	3.3661100000
Al	3.7641900000	9.7184600000	3.3661100000
Al	7.7503400000	9.7183400000	3.3661100000
Al	1.7719600000	1.9439000000	3.3663700000
Al	5.7564200000	1.9437000000	3.3663700000
N	3.7647900000	1.9436600000	3.3663800000
N	7.7500800000	1.9435700000	3.3663800000
N	1.7721400000	3.8873300000	3.3664500000
N	5.7564700000	3.8872100000	3.3664500000
Al	3.7641900000	3.8871600000	3.3664600000
Al	7.7503400000	3.8870500000	3.3664600000
Al	1.7719600000	10.6909800000	5.0492200000
Al	5.7564200000	10.6907800000	5.0492300000
N	7.7500800000	10.6906600000	5.0492300000
N	3.7647900000	10.6907400000	5.0492300000
N	1.7721400000	0.9712000000	5.0493000000
N	5.7564700000	0.9710800000	5.0493100000
Al	7.7503400000	0.9709200000	5.0493100000
Al	3.7642000000	0.9710300000	5.0493100000
Al	1.7719600000	4.8592600000	5.0493200000
Al	5.7564200000	4.8590600000	5.0493200000
N	3.7647900000	4.8590200000	5.0493300000
N	7.7500800000	4.8589300000	5.0493300000
N	1.7721400000	6.8026900000	5.0494000000
N	5.7564700000	6.8025700000	5.0494000000
Al	3.7642000000	6.8025300000	5.0494100000
Al	7.7503400000	6.8024100000	5.0494100000

H	4.5995559234	4.2488422604	12.8225246000
H	5.4030821236	4.0200970694	13.2308970660

Table S4. TS of first C-H activation on AlN through the stepwise PS mechanism.
Total energy: -797.182 Hartree

Al	1.76931830	1.95513877	6.71662797
Al	5.75413890	1.95421813	6.71745757
N	3.76117857	1.95433466	6.71465462
N	7.74834630	1.95476002	6.71420700
Al	3.76608935	3.88872558	6.72198995
Al	7.74277769	3.88885610	6.71891180
N	1.76990357	3.89028577	6.71573541
N	5.75304002	3.88768470	6.72597573
Al	1.76889084	7.78510687	6.71522375
Al	5.75486200	7.78417826	6.71586162
N	3.76108798	7.78568978	6.71362905
N	7.74866783	7.78604705	6.71230859
N	1.76920195	9.72032416	6.71843650
N	5.75503933	9.71916527	6.71820192
Al	3.76307786	9.72073354	6.71894508
Al	7.74709467	9.72080511	6.71860714
Al	1.76720298	4.87029442	8.37071995
Al	5.75110759	4.87514029	8.40245618
N	7.73435212	4.86773745	8.38265440
N	3.76868806	4.86730342	8.39456299
N	1.76616146	6.80837731	8.38556805
N	5.75271166	6.81056264	8.38394433
Al	7.74211238	6.80475424	8.37366337
Al	3.76314924	6.80369180	8.37481294
Al	1.76821132	10.71644994	8.37175405
Al	5.75238752	10.71418713	8.37589742
N	3.76317724	10.70873604	8.38118095
N	7.74338705	10.70969445	8.38047321
N	1.76749912	0.98609130	8.38914084
N	5.75247100	0.99198340	8.39450052
Al	3.76219467	0.98270191	8.38275717
Al	7.74383138	0.98171480	8.38200182
Al	1.76685770	7.81108169	10.06389394
Al	5.75032079	7.78226700	10.07402132
N	3.76543998	7.78935754	10.03686727
N	7.73577038	7.80256117	10.02892411
N	1.76762598	9.73717524	10.03169649
N	5.74988428	9.72720418	10.04345254
Al	3.76209561	9.72679291	10.06720596
Al	7.74141651	9.73073870	10.06413242
Al	1.76593692	1.96796326	10.07469151

Al	5.75106839	1.97079320	10.09385546
N	3.76136595	1.96414329	10.05604559
N	7.74077220	1.96134132	10.05359438
N	1.75975009	3.89068703	10.01353550
N	5.74814037	3.91597742	10.12222901
Al	3.75426535	3.90577417	10.09030897
Al	7.75436804	3.89954447	10.06640184
Al	1.76518056	10.76663126	11.64391160
Al	5.75117904	10.76830657	11.64626513
N	3.75674589	10.66094686	11.75928579
N	7.74596095	10.66178126	11.75788508
N	1.76520219	1.00671399	11.75691290
N	5.75111541	1.00077258	11.76446162
Al	3.75468368	0.89533331	11.64187865
Al	7.74617356	0.89858245	11.64551515
Al	1.72886012	5.00739518	11.58729950
Al	5.89640575	4.91428517	11.77279292
N	3.61988458	4.86864584	11.74809150
N	7.81479262	4.89090476	11.72111744
N	1.75937009	6.91966319	11.75829792
N	5.74856298	6.81765970	11.77217648
Al	3.75374188	6.75721861	11.64307179
Al	7.75973784	6.79138329	11.64043122
Al	1.77195924	1.94403913	0.00000000
Al	1.77195924	7.77596708	0.00000000
Al	3.76418840	3.88730831	0.00000000
Al	5.75641757	1.94383910	0.00000000
Al	5.75641757	7.77576706	0.00000000
Al	7.75033693	3.88718872	0.00000000
N	1.77213916	3.88747871	0.00000000
N	3.76478849	1.94379942	0.00000000
N	3.76478849	7.77571678	0.00000000
N	5.75646784	3.88735858	0.00000000
N	7.75007710	1.94370946	0.00000000
N	7.75007710	7.77563688	0.00000000
N	1.77213916	9.71939608	0.00031010
N	5.75646784	9.71927595	0.00031010
Al	3.76418840	9.71922621	0.00031010
Al	7.75033693	9.71911614	0.00031010
Al	1.77195924	4.85982822	1.68307920
Al	5.75641757	4.85962819	1.68307920
N	7.75007710	4.85949801	1.68308925
N	3.76478849	4.85958797	1.68308925
N	1.77213916	6.80326726	1.68315910
N	5.75646784	6.80314714	1.68315910
Al	7.75033693	6.80298733	1.68315910

Al	3.76419846	6.80309740	1.68315910
Al	1.77195924	10.69132595	1.68317921
Al	5.75641757	10.69112592	1.68317921
N	3.76478849	10.69108570	1.68317921
N	7.75007710	10.69099574	1.68317921
N	1.77213916	0.97153987	1.68324959
N	5.75646784	0.97141974	1.68324959
Al	3.76419846	0.97136947	1.68325912
Al	7.75033693	0.97125940	1.68325912
Al	1.77195924	7.77518708	3.36602875
Al	5.75641757	7.77498705	3.36602875
N	3.76478849	7.77494683	3.36602875
N	7.75007710	7.77485687	3.36602875
N	1.77213916	9.71862612	3.36610866
N	5.75646784	9.71850600	3.36610866
Al	3.76418840	9.71845626	3.36610866
Al	7.75033693	9.71833613	3.36610866
Al	1.77195924	1.94389943	3.36636848
Al	5.75641757	1.94369940	3.36636848
N	3.76478849	1.94365918	3.36637854
N	7.75007710	1.94356922	3.36637854
N	1.77213916	3.88732842	3.36644892
N	5.75646784	3.88720830	3.36644892
Al	3.76418840	3.88715856	3.36645844
Al	7.75033693	3.88704849	3.36645844
Al	1.77195924	10.69097563	5.04921802
Al	5.75641757	10.69077560	5.04922807
N	7.75007710	10.69065601	5.04922807
N	3.76478849	10.69073591	5.04922807
N	1.77213916	0.97119961	5.04929792
N	5.75646784	0.97107948	5.04930798
Al	7.75033693	0.97091967	5.04930798
Al	3.76419846	0.97102974	5.04930798
Al	1.77195924	4.85925829	5.04931803
Al	5.75641757	4.85905826	5.04931803
N	3.76478849	4.85901805	5.04932809
N	7.75007710	4.85892809	5.04932809
N	1.77213916	6.80268728	5.04939794
N	5.75646784	6.80256716	5.04939794
Al	3.76419846	6.80252747	5.04940799
Al	7.75033693	6.80240735	5.04940799
C	5.35374294	3.97734196	13.69334015
C	4.67101119	4.65300695	14.89453067
C	4.83469713	3.86304399	16.19579076
H	4.46498934	4.37794415	12.74066270
H	6.45085711	4.04483938	13.81202144

H	5.14580206	2.89637433	13.66217143
H	5.07268422	5.67158868	15.01710935
H	3.59509264	4.78129882	14.68347525
H	4.36168591	2.87298693	16.10958872
H	5.89707032	3.69975204	16.42968116
H	4.37524445	4.38050244	17.05100381

Table S5. TS of second C-H activation on AlN through the stepwise PSI mechanism.
Total energy: -797.143 Hartree

Al	1.78428964	1.92525470	6.72212108
Al	5.76809690	1.92490609	6.72223368
N	3.77452601	1.92511095	6.72612154
N	7.76170044	1.92683146	6.72348780
Al	3.78237763	3.85963939	6.70489667
Al	7.75692162	3.85978670	6.70018982
N	1.78478877	3.85934834	6.69394417
N	5.76657062	3.85733719	6.70661268
Al	1.78973664	7.75525823	6.72719877
Al	5.76579959	7.75536495	6.72571323
N	3.77542108	7.75541364	6.73435578
N	7.76411295	7.75464523	6.72245835
N	1.78265768	9.68980541	6.70457554
N	5.76948730	9.68910367	6.70271371
Al	3.77761299	9.68761416	6.70929291
Al	7.76258120	9.68757550	6.70742686
Al	1.79853678	4.82558043	8.34650131
Al	5.77951767	4.83234627	8.38432363
N	7.76042069	4.81945358	8.36889880
N	3.80104230	4.81476925	8.38860287
N	1.80738680	6.75484939	8.38437977
N	5.77375109	6.76391346	8.37903764
Al	7.77499552	6.76345133	8.37263029
Al	3.79358172	6.75722489	8.40971159
Al	1.79850979	10.64236103	8.37983254
Al	5.77730739	10.64084106	8.38168596
N	3.78958285	10.63326404	8.39168641
N	7.76978070	10.64094321	8.38605057
N	1.79376227	0.91291304	8.37004692
N	5.77725746	0.92049814	8.37512040
Al	3.78861256	0.90174672	8.36370425
Al	7.77081934	0.90578384	8.36092587
Al	1.83044696	7.71586864	10.10017784
Al	5.78286767	7.69023852	10.09239399
N	3.80703164	7.67593451	10.14013356
N	7.79354496	7.73658650	10.01193067
N	1.81508736	9.65624829	10.03888825

N	5.78437524	9.64527377	10.04520240
Al	3.80289270	9.62519755	10.08684307
Al	7.78396894	9.64185322	10.05929172
Al	1.80264759	1.87418344	10.05588756
Al	5.79079614	1.87678777	10.07619885
N	3.79745036	1.85997444	10.04327538
N	7.77863975	1.87202580	10.03298256
N	1.81579972	3.79804957	9.95616744
N	5.77389461	3.81862992	10.08073535
Al	3.79868403	3.77987424	10.02089575
Al	7.79144713	3.79861625	10.01236596
Al	1.80734709	10.68337792	11.64666628
Al	5.79811276	10.68436021	11.64782059
N	3.80183705	10.57584503	11.77660158
N	7.78823316	10.58315574	11.75613180
N	1.80777113	0.93002783	11.74828680
N	5.79379002	0.92798287	11.76090211
Al	3.79933112	0.80138332	11.63092925
Al	7.78989997	0.81803242	11.63136439
Al	1.74089432	4.93229641	11.49789290
Al	5.95163026	4.83682761	11.71229644
N	3.65819041	4.72180046	11.72377661
N	7.86935762	4.75440049	11.65055572
N	1.77040979	6.79393414	11.75943426
N	5.87685993	6.70777103	11.70936013
Al	3.83310332	6.71376373	11.93738760
Al	7.80684535	6.67943351	11.59358725
Al	1.77195964	1.94403958	0.00000000
Al	1.77195964	7.77596341	0.00000000
Al	3.76418923	3.88730919	0.00000000
Al	5.75641883	1.94383957	0.00000000
Al	5.75641883	7.77576340	0.00000000
Al	7.75033334	3.88718938	0.00000000
N	1.77213960	3.88747938	0.00000000
N	3.76478927	1.94379972	0.00000000
N	3.76478927	7.77571326	0.00000000
N	5.75646896	3.88735931	0.00000000
N	7.75007342	1.94370974	0.00000000
N	7.75007342	7.77563331	0.00000000
N	1.77213960	9.71939293	0.00031005
N	5.75646896	9.71927287	0.00031005
Al	3.76418923	9.71922300	0.00031005
Al	7.75033334	9.71911296	0.00031005
Al	1.77195964	4.85982914	1.68307962
Al	5.75641883	4.85962913	1.68307962
N	7.75007342	4.85949904	1.68308964

N	3.76478927	4.85958902	1.68308964
N	1.77213960	6.80326350	1.68315957
N	5.75646896	6.80314344	1.68315957
Al	7.75033334	6.80298353	1.68315957
Al	3.76419925	6.80309357	1.68315957
Al	1.77195964	10.69132287	1.68317962
Al	5.75641883	10.69112285	1.68317962
N	3.76478927	10.69108275	1.68317962
N	7.75007342	10.69099276	1.68317962
N	1.77213960	0.97153994	1.68324980
N	5.75646896	0.97141987	1.68324980
Al	3.76419925	0.97136974	1.68325958
Al	7.75033334	0.97125971	1.68325958
Al	1.77195964	7.77518341	3.36602940
Al	5.75641883	7.77498340	3.36602940
N	3.76478927	7.77494329	3.36602940
N	7.75007342	7.77485331	3.36602940
N	1.77213960	9.71862295	3.36610935
N	5.75646896	9.71850289	3.36610935
Al	3.76418923	9.71845302	3.36610935
Al	7.75033334	9.71833296	3.36610935
Al	1.77195964	1.94389972	3.36636926
Al	5.75641883	1.94369971	3.36636926
N	3.76478927	1.94365961	3.36637929
N	7.75007342	1.94356963	3.36637929
N	1.77213960	3.88732924	3.36644948
N	5.75646896	3.88720918	3.36644948
Al	3.76418923	3.88715930	3.36645924
Al	7.75033334	3.88704927	3.36645924
Al	1.77195964	10.69097271	5.04921905
Al	5.75641883	10.69077270	5.04922907
N	7.75007342	10.69065290	5.04922907
N	3.76478927	10.69073285	5.04922907
N	1.77213960	0.97119981	5.04929900
N	5.75646896	0.97107974	5.04930903
Al	7.75033334	0.97091984	5.04930903
Al	3.76419925	0.97102987	5.04930903
Al	1.77195964	4.85925918	5.04931905
Al	5.75641883	4.85905916	5.04931905
N	3.76478927	4.85901906	5.04932908
N	7.75007342	4.85892908	5.04932908
N	1.77213960	6.80268351	5.04939901
N	5.75646896	6.80256345	5.04939901
Al	3.76419925	6.80252360	5.04940903
Al	7.75033334	6.80240354	5.04940903
C	5.77832721	4.22357417	13.82524795

C	5.36350132	5.31589803	14.58866623
C	4.45665212	5.16812796	15.76586944
H	6.85288907	4.16812178	13.59057629
H	5.30861659	3.24865069	13.99923561
H	3.83886535	4.21607571	12.60013704
H	5.98556454	6.21382104	14.55383176
H	4.10421040	6.54383099	13.63668256
H	3.98952517	6.11183442	16.06465127
H	5.07457215	4.80270905	16.60829810
H	3.68172995	4.40979447	15.59232637

Table S6. TS of H_2 production on AlN through the stepwise PSD mechanism.
Total energy: -797.126 Hartree

Al	1.77735278	1.95730066	6.71544145
Al	5.76240084	1.95671353	6.71600768
N	3.76936845	1.95579754	6.71375820
N	7.75628180	1.95668966	6.71184776
Al	3.77468283	3.89039029	6.72238027
Al	7.75129928	3.89011721	6.71974190
N	1.77867794	3.89104499	6.71672392
N	5.76131416	3.88919763	6.72559537
Al	1.77628952	7.78541426	6.71365492
Al	5.76311333	7.78458268	6.71380384
N	3.76875737	7.78692685	6.71075412
N	7.75651167	7.78681852	6.70993102
N	1.77656264	9.72040998	6.71745691
N	5.76247723	9.71932960	6.71688912
Al	3.77028074	9.72081194	6.71716256
Al	7.75484028	9.72088807	6.71665971
Al	1.78433937	4.87052655	8.37329528
Al	5.76564179	4.87475437	8.40457493
N	7.75223389	4.86532577	8.38397280
N	3.78373552	4.86944108	8.39444361
N	1.78026773	6.80839467	8.38246709
N	5.76831046	6.81068541	8.38010901
Al	7.75690489	6.80327672	8.37034608
Al	3.77783499	6.80427368	8.36923133
Al	1.78086847	10.72155963	8.36709501
Al	5.76690686	10.71950417	8.36995626
N	3.77611572	10.71418502	8.37522382
N	7.75759575	10.71498518	8.37493912
N	1.78078093	0.99134574	8.38950999
N	5.76775982	0.99552880	8.39389207
Al	3.77599524	0.98535534	8.38109987
Al	7.75862742	0.98746143	8.38046344
Al	1.78461056	7.81645818	10.05662016

Al	5.77094217	7.79142505	10.06442038
N	3.78453060	7.80376966	10.01949815
N	7.75652011	7.81016500	10.01988906
N	1.78543325	9.74428650	10.02785756
N	5.76880263	9.73659645	10.03709813
Al	3.78044772	9.73562132	10.05929265
Al	7.76017345	9.73651742	10.05778159
Al	1.78579257	1.97333469	10.07640994
Al	5.77198957	1.97529490	10.09177468
N	3.78058938	1.95871046	10.05625783
N	7.76283744	1.96993197	10.05314424
N	1.79969039	3.89694230	10.02289884
N	5.76214692	3.91498998	10.11890860
Al	3.78608288	3.89715737	10.09321982
Al	7.76183869	3.91429029	10.07822049
Al	1.78553380	10.77078038	11.63907425
Al	5.77309629	10.77272371	11.64099564
N	3.77788360	10.66483429	11.75686514
N	7.76613553	10.66750993	11.75315711
N	1.78606488	1.01119001	11.76130580
N	5.77343543	1.00739774	11.77025003
Al	3.77638269	0.89950509	11.64255256
Al	7.76990291	0.90376317	11.64177969
Al	1.75974614	5.02417546	11.57363140
Al	5.81375432	4.89701022	11.82472582
N	3.69958700	4.88620934	11.77111565
N	7.83662824	4.90932600	11.69238155
N	1.79385090	6.92740839	11.75203214
N	5.76579649	6.83813693	11.76647474
Al	3.78544768	6.79168295	11.62988687
Al	7.76129237	6.78317931	11.62137246
Al	1.77195924	1.94403913	0.00000000
Al	1.77195924	7.77596708	0.00000000
Al	3.76418840	3.88730831	0.00000000
Al	5.75641757	1.94383910	0.00000000
Al	5.75641757	7.77576706	0.00000000
Al	7.75033693	3.88718872	0.00000000
N	1.77213916	3.88747871	0.00000000
N	3.76478849	1.94379942	0.00000000
N	3.76478849	7.77571678	0.00000000
N	5.75646784	3.88735858	0.00000000
N	7.75007710	1.94370946	0.00000000
N	7.75007710	7.77563688	0.00000000
N	1.77213916	9.71939608	0.00031010
N	5.75646784	9.71927595	0.00031010
Al	3.76418840	9.71922621	0.00031010

Al	7.75033693	9.71911614	0.00031010
Al	1.77195924	4.85982822	1.68307920
Al	5.75641757	4.85962819	1.68307920
N	7.75007710	4.85949801	1.68308925
N	3.76478849	4.85958797	1.68308925
N	1.77213916	6.80326726	1.68315910
N	5.75646784	6.80314714	1.68315910
Al	7.75033693	6.80298733	1.68315910
Al	3.76419846	6.80309740	1.68315910
Al	1.77195924	10.69132595	1.68317921
Al	5.75641757	10.69112592	1.68317921
N	3.76478849	10.69108570	1.68317921
N	7.75007710	10.69099574	1.68317921
N	1.77213916	0.97153987	1.68324959
N	5.75646784	0.97141974	1.68324959
Al	3.76419846	0.97136947	1.68325912
Al	7.75033693	0.97125940	1.68325912
Al	1.77195924	7.77518708	3.36602875
Al	5.75641757	7.77498705	3.36602875
N	3.76478849	7.77494683	3.36602875
N	7.75007710	7.77485687	3.36602875
N	1.77213916	9.71862612	3.36610866
N	5.75646784	9.71850600	3.36610866
Al	3.76418840	9.71845626	3.36610866
Al	7.75033693	9.71833613	3.36610866
Al	1.77195924	1.94389943	3.36636848
Al	5.75641757	1.94369940	3.36636848
N	3.76478849	1.94365918	3.36637854
N	7.75007710	1.94356922	3.36637854
N	1.77213916	3.88732842	3.36644892
N	5.75646784	3.88720830	3.36644892
Al	3.76418840	3.88715856	3.36645844
Al	7.75033693	3.88704849	3.36645844
Al	1.77195924	10.69097563	5.04921802
Al	5.75641757	10.69077560	5.04922807
N	7.75007710	10.69065601	5.04922807
N	3.76478849	10.69073591	5.04922807
N	1.77213916	0.97119961	5.04929792
N	5.75646784	0.97107948	5.04930798
Al	7.75033693	0.97091967	5.04930798
Al	3.76419846	0.97102974	5.04930798
Al	1.77195924	4.85925829	5.04931803
Al	5.75641757	4.85905826	5.04931803
N	3.76478849	4.85901805	5.04932809
N	7.75007710	4.85892809	5.04932809
N	1.77213916	6.80268728	5.04939794

N	5.75646784	6.80256716	5.04939794
Al	3.76419846	6.80252747	5.04940799
Al	7.75033693	6.80240735	5.04940799
C	5.98242994	3.98054475	13.83160224
C	5.14764784	4.42869893	14.86027351
C	4.33023840	3.50677922	15.71993549
H	3.61354314	5.01939445	13.37566459
H	3.77611169	5.14226817	14.21944489
H	6.97486499	4.43585703	13.79232030
H	5.95227338	2.91430849	13.57727417
H	5.40368998	5.39322593	15.31413913
H	5.00499560	3.04060979	16.45809226
H	3.88073169	2.69790522	15.12935284
H	3.54007407	4.03353748	16.27134535

Table S7. TS of H_2 production on AlN through the stepwise PSI/SP mechanism.
Total energy: -776.496 Hartree

Al	1.77097467	1.94156441	6.71548525
Al	5.75544469	1.94136887	6.71553245
N	3.76332726	1.94384086	6.71645996
N	7.74940596	1.94446628	6.71485706
Al	3.76321326	3.87527869	6.71304399
Al	7.74900138	3.87470596	6.71050880
N	1.77128589	3.87388100	6.70994680
N	5.75533058	3.87376798	6.71045304
Al	1.77296257	7.77105559	6.71669778
Al	5.75328679	7.77091137	6.71669867
N	3.76326948	7.77254768	6.72266546
N	7.74931118	7.77130622	6.71469949
N	1.77003344	9.70288655	6.70689816
N	5.75670136	9.70271821	6.70727111
Al	3.76330516	9.70370990	6.71075152
Al	7.74903371	9.70367488	6.71004356
Al	1.77370894	4.85362917	8.36893022
Al	5.75040723	4.85358884	8.36936033
N	7.74826878	4.84127752	8.38282288
N	3.76234535	4.84267500	8.39105975
N	1.77410207	6.78302550	8.37433243
N	5.75062776	6.78300470	8.37464904
Al	7.74797343	6.78460904	8.36916168
Al	3.76212097	6.78254404	8.39111951
Al	1.77026295	10.68025974	8.36995815
Al	5.75447896	10.68020748	8.37029853
N	3.76258282	10.67497070	8.37766738
N	7.74861762	10.67931616	8.37532109
N	1.77028891	0.95348753	8.37530007

N	5.75511491	0.95355229	8.37554994
Al	3.76247299	0.94408161	8.36735390
Al	7.74833696	0.94605865	8.36532957
Al	1.77178681	7.75404053	10.06531459
Al	5.75018296	7.75422636	10.06591685
N	3.76139226	7.75101660	10.08309302
N	7.74725900	7.77132121	10.01211686
N	1.77147288	9.69605064	10.03027605
N	5.75225831	9.69614423	10.03076040
Al	3.76152489	9.68828927	10.06759237
Al	7.74747309	9.69084147	10.05643626
Al	1.77066790	1.91837472	10.06764137
Al	5.75239633	1.91832819	10.06801874
N	3.76191417	1.91410951	10.04190393
N	7.74782463	1.92449236	10.03345973
N	1.78578421	3.86098544	10.03448151
N	5.73660883	3.86110272	10.03523944
Al	3.76098337	3.84366915	10.06939060
Al	7.74697074	3.84782356	10.05121399
Al	1.76588730	10.73405700	11.63822175
Al	5.75661642	10.73402062	11.63863996
N	3.76154368	10.62944469	11.75698090
N	7.74766979	10.63343171	11.74395110
N	1.76735143	0.97307938	11.74760939
N	5.75589569	0.97315434	11.74816867
Al	3.76121920	0.86441346	11.64463897
Al	7.74731919	0.86957586	11.63931888
Al	1.72306367	4.92804924	11.60230225
Al	5.79694494	4.92808804	11.60351365
N	3.76046381	4.78611022	11.83164922
N	7.74651039	4.79421041	11.72115906
N	1.72961873	6.80215821	11.71601299
N	5.78957612	6.80226756	11.71727588
Al	3.75954008	6.81706983	11.80374415
Al	7.74598411	6.71833099	11.61239031
Al	1.77196000	1.94404000	0.00000000
Al	1.77196000	7.77597000	0.00000000
Al	3.76419000	3.88731000	0.00000000
Al	5.75642000	1.94384000	0.00000000
Al	5.75642000	7.77577000	0.00000000
Al	7.75034000	3.88719000	0.00000000
N	1.77214000	3.88748000	0.00000000
N	3.76479000	1.94380000	0.00000000
N	3.76479000	7.77572000	0.00000000
N	5.75647000	3.88736000	0.00000000
N	7.75008000	1.94371000	0.00000000

N	7.75008000	7.77564000	0.00000000
N	1.77214000	9.71940000	0.00031000
N	5.75647000	9.71928000	0.00031000
Al	3.76419000	9.71923000	0.00031000
Al	7.75034000	9.71912000	0.00031000
Al	1.77196000	4.85983000	1.68308000
Al	5.75642000	4.85963000	1.68308000
N	7.75008000	4.85950000	1.68309000
N	3.76479000	4.85959000	1.68309000
N	1.77214000	6.80327000	1.68316000
N	5.75647000	6.80315000	1.68316000
Al	7.75034000	6.80299000	1.68316000
Al	3.76420000	6.80310000	1.68316000
Al	1.77196000	10.69133000	1.68318000
Al	5.75642000	10.69113000	1.68318000
N	3.76479000	10.69109000	1.68318000
N	7.75008000	10.69100000	1.68318000
N	1.77214000	0.97154000	1.68325000
N	5.75647000	0.97142000	1.68325000
Al	3.76420000	0.97137000	1.68326000
Al	7.75034000	0.97126000	1.68326000
Al	1.77196000	7.77519000	3.36603000
Al	5.75642000	7.77499000	3.36603000
N	3.76479000	7.77495000	3.36603000
N	7.75008000	7.77486000	3.36603000
N	1.77214000	9.71863000	3.36611000
N	5.75647000	9.71851000	3.36611000
Al	3.76419000	9.71846000	3.36611000
Al	7.75034000	9.71834000	3.36611000
Al	1.77196000	1.94390000	3.36637000
Al	5.75642000	1.94370000	3.36637000
N	3.76479000	1.94366000	3.36638000
N	7.75008000	1.94357000	3.36638000
N	1.77214000	3.88733000	3.36645000
N	5.75647000	3.88721000	3.36645000
Al	3.76419000	3.88716000	3.36646000
Al	7.75034000	3.88705000	3.36646000
Al	1.77196000	10.69098000	5.04922000
Al	5.75642000	10.69078000	5.04923000
N	7.75008000	10.69066000	5.04923000
N	3.76479000	10.69074000	5.04923000
N	1.77214000	0.97120000	5.04930000
N	5.75647000	0.97108000	5.04931000
Al	7.75034000	0.97092000	5.04931000
Al	3.76420000	0.97103000	5.04931000
Al	1.77196000	4.85926000	5.04932000

Al	5.75642000	4.85906000	5.04932000
N	3.76479000	4.85902000	5.04933000
N	7.75008000	4.85893000	5.04933000
N	1.77214000	6.80269000	5.04940000
N	5.75647000	6.80257000	5.04940000
Al	3.76420000	6.80253000	5.04941000
Al	7.75034000	6.80241000	5.04941000
H	3.75302447	6.38893039	13.52691686
H	3.75605646	5.42824171	12.97527269

Table S8. TS of first C-H activation on AlN through the stepwise SP mechanism.
Total energy: -797.178 Hartree

Al	1.7666406528	1.9366805335	6.7179128062
Al	5.7513919207	1.9355814213	6.7188808394
N	3.7590966533	1.9371104552	6.7190595313
N	7.7462501528	1.9369271105	6.7183456538
Al	3.7631484162	3.8687195003	6.7134245563
Al	7.7392578414	3.8688701703	6.7118989052
N	1.7666961990	3.8702833052	6.7056682467
N	5.7510121855	3.8677240897	6.7183015443
Al	1.7659094967	7.7639604241	6.7204985453
Al	5.7517866117	7.7629734015	6.7215869222
N	3.7589582666	7.7644651143	6.7223922580
N	7.7459812813	7.7644297044	6.7219252682
N	1.7670316451	9.6971241020	6.7055411810
N	5.7530247263	9.6962306238	6.7058636056
Al	3.7604532829	9.6981023033	6.7092983406
Al	7.7444146668	9.6980513621	6.7093767897
Al	1.7592735781	4.8277517507	8.3711627296
Al	5.7453938875	4.8323143470	8.4052400425
N	7.7283133588	4.8239063537	8.3879550743
N	3.7630422366	4.8226081260	8.3953861073
N	1.7617246230	6.7646792791	8.3773406929
N	5.7470116314	6.7664170445	8.3764890118
Al	7.7359113242	6.7594572932	8.3690573005
Al	3.7568914533	6.7590872420	8.3694645017
Al	1.7636509778	10.6714009079	8.3684920811
Al	5.7480363923	10.6686585239	8.3728551967
N	3.7593983150	10.6632858503	8.3808797759
N	7.7400893655	10.6636950946	8.3810758592
N	1.7647519280	0.9405798453	8.3726009320
N	5.7483514517	0.9456867380	8.3768827084
Al	3.7570583626	0.9372566190	8.3706047776
Al	7.7398847402	0.9364332144	8.3682403487
Al	1.7560519110	7.7459287570	10.0648209428
Al	5.7422679892	7.7151111851	10.0749687440

N	3.7573455674	7.7262313583	10.0389762604
N	7.7281177683	7.7332248620	10.0332582604
N	1.7624182842	9.6713562261	10.0169716063
N	5.7444768893	9.6583082168	10.0275209388
Al	3.7550670363	9.6627743988	10.0565601403
Al	7.7353252413	9.6627661272	10.0545359927
Al	1.7575376435	1.9086986673	10.0624052405
Al	5.7454934873	1.9086887788	10.0789335134
N	3.7544353575	1.9056853221	10.0481002086
N	7.7367780460	1.9030678067	10.0386776542
N	1.7483227192	3.8339492630	10.0063413470
N	5.7396642669	3.8541481116	10.1217260506
Al	3.7454369541	3.8429648792	10.0779069029
Al	7.7403762887	3.8375936848	10.0655326249
Al	1.7581376330	10.6947354066	11.6334596900
Al	5.7454521354	10.6930659291	11.6320257763
N	3.7515361841	10.5902147069	11.7526984242
N	7.7416035224	10.5825148449	11.7506485636
N	1.7566307968	0.9317476078	11.7280656462
N	5.7532821030	0.9184088195	11.7230074361
Al	3.7462822853	0.8256741515	11.6258415069
Al	7.7398939294	0.8194717077	11.6156086576
Al	1.7118758098	4.9336463961	11.5875449658
Al	5.8713396901	4.8259169698	11.7972787667
N	3.6051346106	4.8022463047	11.7303786219
N	7.7949873092	4.8053787204	11.7427247812
N	1.7444018127	6.8434554658	11.7536511744
N	5.7366554154	6.7372458139	11.7690017483
Al	3.7415404390	6.6895327662	11.6360936248
Al	7.7408912042	6.7112363363	11.6406737485
Al	1.7719592400	1.9440391300	0.0000000000
Al	1.7719592400	7.7759670800	0.0000000000
Al	3.7641884000	3.8873083100	0.0000000000
Al	5.7564175700	1.9438391000	0.0000000000
Al	5.7564175700	7.7757670600	0.0000000000
Al	7.7503369300	3.8871887200	0.0000000000
N	1.7721391600	3.8874787100	0.0000000000
N	3.7647884900	1.9437994200	0.0000000000
N	3.7647884900	7.7757167800	0.0000000000
N	5.7564678400	3.8873585800	0.0000000000
N	7.7500771000	1.9437094600	0.0000000000
N	7.7500771000	7.7756368800	0.0000000000
N	1.7721391600	9.7193960800	0.0003101000
N	5.7564678400	9.7192759500	0.0003101000
Al	3.7641884000	9.7192262100	0.0003101000
Al	7.7503369300	9.7191161400	0.0003101000

Al	1.7719592400	4.8598282200	1.6830792000
Al	5.7564175700	4.8596281900	1.6830792000
N	7.7500771000	4.8594980100	1.6830892500
N	3.7647884900	4.8595879700	1.6830892500
N	1.7721391600	6.8032672600	1.6831591000
N	5.7564678400	6.8031471400	1.6831591000
Al	7.7503369300	6.8029873300	1.6831591000
Al	3.7641984600	6.8030974000	1.6831591000
Al	1.7719592400	10.6913259500	1.6831792100
Al	5.7564175700	10.6911259200	1.6831792100
N	3.7647884900	10.6910857000	1.6831792100
N	7.7500771000	10.6909957400	1.6831792100
N	1.7721391600	0.9715398700	1.6832495900
N	5.7564678400	0.9714197400	1.6832495900
Al	3.7641984600	0.9713694700	1.6832591200
Al	7.7503369300	0.9712594000	1.6832591200
Al	1.7719592400	7.7751870800	3.3660287500
Al	5.7564175700	7.7749870500	3.3660287500
N	3.7647884900	7.7749468300	3.3660287500
N	7.7500771000	7.7748568700	3.3660287500
N	1.7721391600	9.7186261200	3.3661086600
N	5.7564678400	9.7185060000	3.3661086600
Al	3.7641884000	9.7184562600	3.3661086600
Al	7.7503369300	9.7183361300	3.3661086600
Al	1.7719592400	1.9438994300	3.3663684800
Al	5.7564175700	1.9436994000	3.3663684800
N	3.7647884900	1.9436591800	3.3663785400
N	7.7500771000	1.9435692200	3.3663785400
N	1.7721391600	3.8873284200	3.3664489200
N	5.7564678400	3.8872083000	3.3664489200
Al	3.7641884000	3.8871585600	3.3664584400
Al	7.7503369300	3.8870484900	3.3664584400
Al	1.7719592400	10.6909756300	5.0492180200
Al	5.7564175700	10.6907756000	5.0492280700
N	7.7500771000	10.6906560100	5.0492280700
N	3.7647884900	10.6907359100	5.0492280700
N	1.7721391600	0.9711996100	5.0492979200
N	5.7564678400	0.9710794800	5.0493079800
Al	7.7503369300	0.9709196700	5.0493079800
Al	3.7641984600	0.9710297400	5.0493079800
Al	1.7719592400	4.8592582900	5.0493180300
Al	5.7564175700	4.8590582600	5.0493180300
N	3.7647884900	4.8590180500	5.0493280900
N	7.7500771000	4.8589280900	5.0493280900
N	1.7721391600	6.8026872800	5.0493979400
N	5.7564678400	6.8025671600	5.0493979400

Al	3.7641984600	6.8025274700	5.0494079900
Al	7.7503369300	6.8024073500	5.0494079900
C	4.7520461206	5.1678553336	14.7778971232
C	5.1577589289	4.0197313429	13.8406625138
C	4.3397029470	2.7561453549	14.1717410587
H	5.3711201615	6.0648195485	14.6436753631
H	4.3870656338	4.3483609369	12.7333991481
H	3.7037635428	5.4601861860	14.5902638593
H	4.8075778982	4.8637131614	15.8353844372
H	6.2273195910	3.7846060164	13.9784003392
H	4.7934362370	1.8652731595	13.7143738579
H	3.2959094026	2.8456704512	13.8205513870
H	4.2878305285	2.5777945110	15.2582083724

Table S9. TS of second C-H activation on AlN through the stepwise SP mechanism.
Total energy: -797.137 Hartree

Al	1.77028176	1.92423764	6.72246259
Al	5.75489016	1.92406223	6.72389063
N	3.76245636	1.92427203	6.72668405
N	7.74914435	1.92565625	6.72544031
Al	3.76792168	3.85902534	6.70410645
Al	7.74227296	3.85926514	6.70162358
N	1.77123709	3.85845042	6.69305091
N	5.75354714	3.85748575	6.70852450
Al	1.77438748	7.75568683	6.72571626
Al	5.75150004	7.75550101	6.72603698
N	3.76226199	7.75542097	6.73389795
N	7.75011267	7.75499447	6.72266532
N	1.76991841	9.68988749	6.70421179
N	5.75651103	9.68972230	6.70424750
Al	3.76373993	9.68782203	6.70970732
Al	7.74824361	9.68809924	6.70867291
Al	1.76693843	4.82494754	8.34600577
Al	5.75297111	4.83248595	8.38725793
N	7.73539550	4.81932967	8.37150800
N	3.77072879	4.81445194	8.38650151
N	1.77802862	6.75567270	8.38116897
N	5.74676836	6.76345703	8.37997518
Al	7.74519845	6.76321544	8.37312756
Al	3.76471290	6.75716429	8.40767743
Al	1.77189939	10.64121956	8.38084145
Al	5.75225620	10.64040713	8.38446540
N	3.76460542	10.63211150	8.39307956
N	7.74626130	10.63997873	8.38791757
N	1.77004866	0.91238784	8.37078097
N	5.75335583	0.92015970	8.37774455

Al	3.76349433	0.90099278	8.36486714
Al	7.74533605	0.90472766	8.36411671
Al	1.78345868	7.71391211	10.09339208
Al	5.74372618	7.69181736	10.09452910
N	3.76536592	7.67691514	10.13756091
N	7.74899996	7.73375828	10.01410855
N	1.77547268	9.65454383	10.04085774
N	5.74957653	9.64726727	10.04971489
Al	3.76485184	9.62623746	10.08821493
Al	7.74535715	9.64213857	10.06424283
Al	1.76418312	1.87308744	10.05710408
Al	5.75446458	1.87750867	10.08019993
N	3.76197686	1.86103030	10.04409852
N	7.74328403	1.86794755	10.03770649
N	1.77020174	3.79662162	9.95675092
N	5.73815050	3.82195106	10.08592935
Al	3.76262008	3.77955492	10.01153115
Al	7.74045192	3.79538054	10.02015180
Al	1.76532281	10.68335520	11.64981129
Al	5.75621226	10.68481305	11.65536866
N	3.76082981	10.57589707	11.77623965
N	7.74764124	10.58537447	11.76115616
N	1.76919280	0.93118265	11.75045323
N	5.75187719	0.93200377	11.76872011
Al	3.75688731	0.80062863	11.63334192
Al	7.74946627	0.82028558	11.63965970
Al	1.68703410	4.92820324	11.48803314
Al	5.89428682	4.85163288	11.74292883
N	3.62857981	4.71757047	11.70945305
N	7.80811060	4.73593448	11.67107661
N	1.71373035	6.78215081	11.74001153
N	5.82709432	6.71521228	11.71664299
Al	3.77281461	6.73072564	11.92620352
Al	7.75404580	6.67427550	11.60213964
Al	1.77195919	1.94403914	0.00000000
Al	1.77195919	7.77596588	0.00000000
Al	3.76418788	3.88730784	0.00000000
Al	5.75641704	1.94383913	0.00000000
Al	5.75641704	7.77576587	0.00000000
Al	7.75033581	3.88718802	0.00000000
N	1.77213916	3.88747801	0.00000000
N	3.76478792	1.94379927	0.00000000
N	3.76478792	7.77571575	0.00000000
N	5.75646716	3.88735796	0.00000000
N	7.75007589	1.94370929	0.00000000
N	7.75007589	7.77563579	0.00000000

N	1.77213916	9.71939496	0.00031004
N	5.75646716	9.71927491	0.00031004
Al	3.76418788	9.71922502	0.00031004
Al	7.75033581	9.71911499	0.00031004
Al	1.77195919	4.85982733	1.68307917
Al	5.75641704	4.85962731	1.68307917
N	7.75007589	4.85949723	1.68308920
N	3.76478792	4.85958722	1.68308920
N	1.77213916	6.80326643	1.68315913
N	5.75646716	6.80314638	1.68315913
Al	7.75033581	6.80298646	1.68315913
Al	3.76419791	6.80309649	1.68315913
Al	1.77195919	10.69132443	1.68317918
Al	5.75641704	10.69112442	1.68317918
N	3.76478792	10.69108433	1.68317918
N	7.75007589	10.69099434	1.68317918
N	1.77213916	0.97153947	1.68324935
N	5.75646716	0.97141942	1.68324935
Al	3.76419791	0.97136929	1.68325914
Al	7.75033581	0.97125926	1.68325914
Al	1.77195919	7.77518588	3.36602803
Al	5.75641704	7.77498586	3.36602803
N	3.76478792	7.77494577	3.36602803
N	7.75007589	7.77485579	3.36602803
N	1.77213916	9.71862498	3.36610799
N	5.75646716	9.71850493	3.36610799
Al	3.76418788	9.71845505	3.36610799
Al	7.75033581	9.71833499	3.36610799
Al	1.77195919	1.94389927	3.36636792
Al	5.75641704	1.94369926	3.36636792
N	3.76478792	1.94365917	3.36637794
N	7.75007589	1.94356918	3.36637794
N	1.77213916	3.88732789	3.36644810
N	5.75646716	3.88720783	3.36644810
Al	3.76418788	3.88715795	3.36645790
Al	7.75033581	3.88704792	3.36645790
Al	1.77195919	10.69097430	5.04921723
Al	5.75641704	10.69077429	5.04922726
N	7.75007589	10.69065446	5.04922726
N	3.76478792	10.69073442	5.04922726
N	1.77213916	0.97119936	5.04929720
N	5.75646716	0.97107930	5.04930722
Al	7.75033581	0.97091938	5.04930722
Al	3.76419791	0.97102941	5.04930722
Al	1.77195919	4.85925736	5.04931724
Al	5.75641704	4.85905735	5.04931724

N	3.76478792	4.85901725	5.04932726
N	7.75007589	4.85892726	5.04932726
N	1.77213916	6.80268644	5.04939720
N	5.75646716	6.80256639	5.04939720
Al	3.76419791	6.80252653	5.04940723
Al	7.75033581	6.80240648	5.04940723
C	6.95824276	4.42625763	14.69740721
C	5.59173914	4.32382503	14.04362558
C	4.53770036	5.07631017	14.55074903
H	7.76036527	4.12067077	14.01806252
H	3.72052435	4.14615649	12.55330514
H	7.15941447	5.46200278	15.00761534
H	7.00212350	3.79426430	15.59849599
H	5.33832044	3.32387689	13.64850680
H	3.95988971	6.63537584	13.61037815
H	4.71896660	5.80198892	15.34380159
H	3.49873261	4.76917752	14.45338891

Table S10. TS of C-H activation of propane on Ga/AlN through the concerted mechanism.
Total energy: -869.591 Hartree

Al	1.77662527	1.96384425	6.71263057
Al	5.76090129	1.96346251	6.71373424
N	3.76855731	1.96293879	6.71119818
N	7.75485170	1.96456100	6.70881638
Al	3.77295982	3.89683135	6.72310223
Al	7.75068988	3.89678774	6.71866729
N	1.77831793	3.89709617	6.71570192
N	5.75925092	3.89632434	6.72420182
Al	1.77514499	7.79572970	6.70804991
Al	5.76169385	7.79454300	6.70819956
N	3.76764660	7.79622810	6.70612467
N	7.75509438	7.79654198	6.70403610
N	1.77521760	9.72887687	6.71906274
N	5.76130577	9.72745565	6.71852405
Al	3.76907003	9.72915829	6.71853966
Al	7.75350835	9.72942238	6.71800128
Al	1.78477409	4.89204934	8.36097788
Al	5.76114323	4.89693144	8.38877580
N	7.75206268	4.88751418	8.37322907
N	3.77905675	4.89133863	8.39147252
N	1.77772154	6.82947880	8.38008264
N	5.76602329	6.82932266	8.37740041
Al	7.75542869	6.82373488	8.36822485
Al	3.77475117	6.82319721	8.36956333
Al	1.77934714	10.73661696	8.36478258

Al	5.76386876	10.73390734	8.36786611
N	3.77404115	10.72774441	8.37239560
N	7.75515699	10.73033369	8.37152138
N	1.77898413	1.00564501	8.38946575
N	5.76532585	1.00843601	8.39401690
Al	3.77334167	0.99892164	8.38217052
Al	7.75714201	1.00183234	8.38057689
Al	1.78436091	7.84321342	10.04952979
Al	5.76469224	7.81445342	10.05599116
N	3.78198880	7.81382585	10.02625445
N	7.75125574	7.83371360	10.01567342
N	1.78396289	9.76739783	10.02695496
N	5.76382806	9.75802541	10.03700462
Al	3.77751620	9.75620817	10.06293113
Al	7.75648832	9.76058201	10.05776747
Al	1.78381081	1.99266577	10.07476990
Al	5.76862940	1.98983889	10.08773926
N	3.77467708	1.97794798	10.05673472
N	7.76266453	1.99386352	10.04938637
N	1.80052684	3.92350464	10.00716003
N	5.75267537	3.92434160	10.07389383
Al	3.78127129	3.91033809	10.09418158
Al	7.76794152	3.93107056	10.05647771
Al	1.78153798	10.80005542	11.63764571
Al	5.76960912	10.80005455	11.63971534
N	3.77402434	10.69163614	11.75375987
N	7.76266249	10.69643369	11.74693093
N	1.78178751	1.03983725	11.76149184
N	5.77024505	1.03473468	11.76637897
Al	3.77457293	0.92654207	11.65183515
Al	7.76394511	0.93539721	11.64898774
Al	1.73625990	5.06412425	11.55403031
Ga	5.89427644	4.88312124	11.85728773
N	3.60593097	4.87865952	11.83848314
N	7.87699877	4.93790911	11.68927670
N	1.78959588	6.96727768	11.74174008
N	5.75864761	6.89720664	11.77654162
Al	3.78165890	6.81994371	11.66508908
Al	7.76538256	6.82974499	11.62969577
Al	1.77195871	1.94403860	0.00000000
Al	1.77195871	7.77596391	0.00000000
Al	3.76418682	3.88730672	0.00000000
Al	5.75641545	1.94383857	0.00000000
Al	5.75641545	7.77576388	0.00000000
Al	7.75033375	3.88718713	0.00000000
N	1.77213863	3.88747712	0.00000000

N	3.76478690	1.94379889	0.00000000
N	3.76478690	7.77571361	0.00000000
N	5.75646572	3.88735700	0.00000000
N	7.75007392	1.94370893	0.00000000
N	7.75007392	7.77563370	0.00000000
N	1.77213863	9.71939237	0.00031010
N	5.75646572	9.71927225	0.00031010
Al	3.76418682	9.71922251	0.00031010
Al	7.75033375	9.71911244	0.00031010
Al	1.77195871	4.85982610	1.68307867
Al	5.75641545	4.85962607	1.68307867
N	7.75007392	4.85949589	1.68308872
N	3.76478690	4.85958585	1.68308872
N	1.77213863	6.80326462	1.68315858
N	5.75646572	6.80314449	1.68315858
Al	7.75033375	6.80298468	1.68315858
Al	3.76419687	6.80309475	1.68315858
Al	1.77195871	10.69132171	1.68317868
Al	5.75641545	10.69112168	1.68317868
N	3.76478690	10.69108147	1.68317868
N	7.75007392	10.69099151	1.68317868
N	1.77213863	0.97153934	1.68324906
N	5.75646572	0.97141921	1.68324906
Al	3.76419687	0.97136894	1.68325859
Al	7.75033375	0.97125887	1.68325859
Al	1.77195871	7.77518390	3.36602716
Al	5.75641545	7.77498387	3.36602716
N	3.76478690	7.77494366	3.36602716
N	7.75007392	7.77485370	3.36602716
N	1.77213863	9.71862242	3.36610760
N	5.75646572	9.71850230	3.36610760
Al	3.76418682	9.71845255	3.36610760
Al	7.75033375	9.71833243	3.36610760
Al	1.77195871	1.94389890	3.36636689
Al	5.75641545	1.94369887	3.36636689
N	3.76478690	1.94365865	3.36637748
N	7.75007392	1.94356869	3.36637748
N	1.77213863	3.88732683	3.36644733
N	5.75646572	3.88720671	3.36644733
Al	3.76418682	3.88715697	3.36645685
Al	7.75033375	3.88704690	3.36645685
Al	1.77195871	10.69097140	5.04921590
Al	5.75641545	10.69077137	5.04922596
N	7.75007392	10.69065177	5.04922596
N	3.76478690	10.69073168	5.04922596
N	1.77213863	0.97119908	5.04929581

N	5.75646572	0.97107895	5.04930586
Al	7.75033375	0.97091914	5.04930586
Al	3.76419687	0.97102921	5.04930586
Al	1.77195871	4.85925618	5.04931592
Al	5.75641545	4.85905615	5.04931592
N	3.76478690	4.85901593	5.04932597
N	7.75007392	4.85892597	5.04932597
N	1.77213863	6.80268464	5.04939582
N	5.75646572	6.80256451	5.04939582
Al	3.76419687	6.80252483	5.04940588
Al	7.75033375	6.80240470	5.04940588
C	5.08198237	4.06037226	14.60416619
C	3.72997713	4.47900961	14.57634304
C	3.37763156	5.90854753	14.91467356
H	5.63027891	4.13468058	13.38790391
H	3.63820950	4.58308642	12.96860117
H	5.82139156	4.71347640	15.08525357
H	5.29729159	2.99947830	14.76575326
H	2.95357867	3.71771256	14.69317334
H	4.09334942	6.61017803	14.44253027
H	3.41879442	6.12886784	15.99569512
H	2.37420233	6.19026958	14.56359112

Table S11. TS of H_2 production on Ga/AlN through the concerted mechanism.
Total energy: -848.952 Hartree

Al	1.77325994	1.94443645	6.71637526
Al	5.75782900	1.94378011	6.71691886
N	3.76523562	1.94567136	6.71630098
N	7.75149446	1.94645259	6.71520783
Al	3.76797218	3.87732070	6.71499783
Al	7.74838523	3.87777266	6.71125489
N	1.77335919	3.87675029	6.70669588
N	5.75671972	3.87544989	6.71623864
Al	1.77262856	7.77493068	6.71646038
Al	5.75852290	7.77423618	6.71673377
N	3.76525699	7.77595913	6.71727084
N	7.75151089	7.77656575	6.71527879
N	1.77264603	9.70663651	6.71030598
N	5.75849062	9.70595179	6.71023153
Al	3.76646276	9.70862587	6.71268198
Al	7.75101353	9.70885513	6.71200303
Al	1.77487843	4.85196632	8.36380308
Al	5.75612623	4.85803126	8.38407029
N	7.74435107	4.85199455	8.37478205
N	3.77102053	4.85106566	8.38750162
N	1.77218367	6.78896069	8.38004003

N	5.75911189	6.78929893	8.38015977
Al	7.75072365	6.78537066	8.37003972
Al	3.76740402	6.78345990	8.37162204
Al	1.77489846	10.69195766	8.36745629
Al	5.75854458	10.69035126	8.37024991
N	3.76862734	10.68541388	8.37781926
N	7.75015474	10.68691437	8.37670239
N	1.77354134	0.95888287	8.37736440
N	5.75883120	0.96322743	8.38144397
Al	3.76757555	0.95760413	8.37398754
Al	7.75158173	0.95693868	8.37215597
Al	1.77713498	7.78425110	10.06016072
Al	5.75702428	7.76367773	10.06585342
N	3.77306081	7.75785866	10.03833042
N	7.74417743	7.77558003	10.02788236
N	1.77604039	9.70779851	10.02381936
N	5.75743042	9.70145227	10.03410384
Al	3.76978017	9.70006901	10.06262460
Al	7.75071524	9.70506235	10.05790881
Al	1.77533003	1.93597113	10.06528045
Al	5.75891740	1.93167061	10.08053261
N	3.76741739	1.93334431	10.04693148
N	7.75091582	1.93302427	10.04263479
N	1.76928257	3.86198613	10.00215723
N	5.75366264	3.86839981	10.06837134
Al	3.75556230	3.87524047	10.08412883
Al	7.76873532	3.86859027	10.04911355
Al	1.77519563	10.74018483	11.63726077
Al	5.76073167	10.74247436	11.64058485
N	3.76659888	10.63563981	11.75138318
N	7.75440023	10.63624941	11.74789393
N	1.77386835	0.97824073	11.74596299
N	5.76099881	0.97614877	11.75484475
Al	3.76550411	0.87137582	11.63868779
Al	7.75600616	0.87427590	11.63876401
Al	1.74800135	4.97225779	11.59066181
Ga	5.87778123	4.90450297	11.75479036
N	3.61831060	4.82597427	11.77169936
N	7.84472722	4.86420995	11.71344175
N	1.76977998	6.89570423	11.75700369
N	5.75525947	6.85536194	11.79839808
Al	3.75032992	6.72913614	11.65529199
Al	7.78299865	6.77570021	11.64762908
Al	1.77196000	1.94404000	0.00000000
Al	1.77196000	7.77597000	0.00000000
Al	3.76419000	3.88731000	0.00000000

Al	5.75642000	1.94384000	0.00000000
Al	5.75642000	7.77577000	0.00000000
Al	7.75034000	3.88719000	0.00000000
N	1.77214000	3.88748000	0.00000000
N	3.76479000	1.94380000	0.00000000
N	3.76479000	7.77572000	0.00000000
N	5.75647000	3.88736000	0.00000000
N	7.75008000	1.94371000	0.00000000
N	7.75008000	7.77564000	0.00000000
N	1.77214000	9.71940000	0.00031000
N	5.75647000	9.71928000	0.00031000
Al	3.76419000	9.71923000	0.00031000
Al	7.75034000	9.71912000	0.00031000
Al	1.77196000	4.85983000	1.68308000
Al	5.75642000	4.85963000	1.68308000
N	7.75008000	4.85950000	1.68309000
N	3.76479000	4.85959000	1.68309000
N	1.77214000	6.80327000	1.68316000
N	5.75647000	6.80315000	1.68316000
Al	7.75034000	6.80299000	1.68316000
Al	3.76420000	6.80310000	1.68316000
Al	1.77196000	10.69133000	1.68318000
Al	5.75642000	10.69113000	1.68318000
N	3.76479000	10.69109000	1.68318000
N	7.75008000	10.69100000	1.68318000
N	1.77214000	0.97154000	1.68325000
N	5.75647000	0.97142000	1.68325000
Al	3.76420000	0.97137000	1.68326000
Al	7.75034000	0.97126000	1.68326000
Al	1.77196000	7.77519000	3.36603000
Al	5.75642000	7.77499000	3.36603000
N	3.76479000	7.77495000	3.36603000
N	7.75008000	7.77486000	3.36603000
N	1.77214000	9.71863000	3.36611000
N	5.75647000	9.71851000	3.36611000
Al	3.76419000	9.71846000	3.36611000
Al	7.75034000	9.71834000	3.36611000
Al	1.77196000	1.94390000	3.36637000
Al	5.75642000	1.94370000	3.36637000
N	3.76479000	1.94366000	3.36638000
N	7.75008000	1.94357000	3.36638000
N	1.77214000	3.88733000	3.36645000
N	5.75647000	3.88721000	3.36645000
Al	3.76419000	3.88716000	3.36646000
Al	7.75034000	3.88705000	3.36646000
Al	1.77196000	10.69098000	5.04922000

Al	5.75642000	10.69078000	5.04923000
N	7.75008000	10.69066000	5.04923000
N	3.76479000	10.69074000	5.04923000
N	1.77214000	0.97120000	5.04930000
N	5.75647000	0.97108000	5.04931000
Al	7.75034000	0.97092000	5.04931000
Al	3.76420000	0.97103000	5.04931000
Al	1.77196000	4.85926000	5.04932000
Al	5.75642000	4.85906000	5.04932000
N	3.76479000	4.85902000	5.04933000
N	7.75008000	4.85893000	5.04933000
N	1.77214000	6.80269000	5.04940000
N	5.75647000	6.80257000	5.04940000
Al	3.76420000	6.80253000	5.04941000
Al	7.75034000	6.80241000	5.04941000
H	4.53328393	4.23497177	12.85752717
H	5.32137481	3.99115928	13.29412990

Table S12. TS of first C-H activation on Ga/AlN through the stepwise PS mechanism.
Total energy: -869.617 Hartree

Al	1.77128933	1.95476582	6.71383116
Al	5.75568048	1.95392190	6.71482561
N	3.76318076	1.95430778	6.71248134
N	7.74980407	1.95510440	6.71181777
Al	3.76750501	3.88721488	6.71832342
Al	7.74527069	3.88751247	6.71409579
N	1.77203203	3.88767702	6.70949973
N	5.75476674	3.88578669	6.72044847
Al	1.77073245	7.78509991	6.71292490
Al	5.75639471	7.78416961	6.71355273
N	3.76308318	7.78544103	6.71193298
N	7.75007812	7.78609769	6.70999537
N	1.77062030	9.71820278	6.71466636
N	5.75644529	9.71709542	6.71447061
Al	3.76446710	9.71888395	6.71557811
Al	7.74847987	9.71905776	6.71499195
Al	1.77282748	4.87068518	8.36015204
Al	5.75367659	4.87787425	8.38843054
N	7.73899353	4.87097687	8.37179267
N	3.77172455	4.86983708	8.38681121
N	1.76978764	6.80948888	8.38226826
N	5.75649156	6.81031442	8.38180666
Al	7.74672935	6.80479823	8.36971193
Al	3.76591809	6.80302671	8.37215825
Al	1.77119275	10.71511253	8.36526813
Al	5.75462980	10.71279816	8.36935401

N	3.76599871	10.70690361	8.37561215
N	7.74588554	10.70826123	8.37445900
N	1.77038931	0.98224225	8.38210281
N	5.75552693	0.98780868	8.38810921
Al	3.76448493	0.97995129	8.37734365
Al	7.74751616	0.97886684	8.37596370
Al	1.77428335	7.81499224	10.05724984
Al	5.75435479	7.78677263	10.06605983
N	3.77258328	7.78647258	10.03460900
N	7.73915125	7.80358517	10.02261791
N	1.77196982	9.73799932	10.02384227
N	5.75332142	9.72847939	10.03667408
Al	3.76625099	9.72585863	10.06132850
Al	7.74510001	9.73104671	10.05653504
Al	1.77154514	1.96405078	10.06689699
Al	5.75470655	1.96039117	10.08578755
N	3.76437873	1.95893674	10.05105986
N	7.74658419	1.95678473	10.04777194
N	1.76583466	3.88420421	9.99247587
N	5.75290834	3.89688072	10.08406116
Al	3.75396701	3.90493941	10.08691928
Al	7.76956672	3.89518959	10.04834915
Al	1.76933300	10.76299293	11.63798732
Al	5.75498331	10.76587413	11.64109498
N	3.76097442	10.65845105	11.75386915
N	7.74979598	10.65889477	11.75117896
N	1.76916625	1.00276562	11.75118216
N	5.75583692	0.99882645	11.75979860
Al	3.75711710	0.89234923	11.63540230
Al	7.75226190	0.89559017	11.63855941
Al	1.74500949	5.01611249	11.57139612
Ga	5.91524675	4.90818125	11.79412374
N	3.59424394	4.86514690	11.75439445
N	7.86450088	4.89805011	11.70693313
N	1.76707644	6.94137404	11.75685562
N	5.75639465	6.87150832	11.79946375
Al	3.75250485	6.75940590	11.65061492
Al	7.78188950	6.80967617	11.64314239
Al	1.77195924	1.94403913	0.00000000
Al	1.77195924	7.77596708	0.00000000
Al	3.76418840	3.88730831	0.00000000
Al	5.75641757	1.94383910	0.00000000
Al	5.75641757	7.77576706	0.00000000
Al	7.75033693	3.88718872	0.00000000
N	1.77213916	3.88747871	0.00000000
N	3.76478849	1.94379942	0.00000000

N	3.76478849	7.77571678	0.00000000
N	5.75646784	3.88735858	0.00000000
N	7.75007710	1.94370946	0.00000000
N	7.75007710	7.77563688	0.00000000
N	1.77213916	9.71939608	0.00031010
N	5.75646784	9.71927595	0.00031010
Al	3.76418840	9.71922621	0.00031010
Al	7.75033693	9.71911614	0.00031010
Al	1.77195924	4.85982822	1.68307920
Al	5.75641757	4.85962819	1.68307920
N	7.75007710	4.85949801	1.68308925
N	3.76478849	4.85958797	1.68308925
N	1.77213916	6.80326726	1.68315910
N	5.75646784	6.80314714	1.68315910
Al	7.75033693	6.80298733	1.68315910
Al	3.76419846	6.80309740	1.68315910
Al	1.77195924	10.69132595	1.68317921
Al	5.75641757	10.69112592	1.68317921
N	3.76478849	10.69108570	1.68317921
N	7.75007710	10.69099574	1.68317921
N	1.77213916	0.97153987	1.68324959
N	5.75646784	0.97141974	1.68324959
Al	3.76419846	0.97136947	1.68325912
Al	7.75033693	0.97125940	1.68325912
Al	1.77195924	7.77518708	3.36602875
Al	5.75641757	7.77498705	3.36602875
N	3.76478849	7.77494683	3.36602875
N	7.75007710	7.77485687	3.36602875
N	1.77213916	9.71862612	3.36610866
N	5.75646784	9.71850600	3.36610866
Al	3.76418840	9.71845626	3.36610866
Al	7.75033693	9.71833613	3.36610866
Al	1.77195924	1.94389943	3.36636848
Al	5.75641757	1.94369940	3.36636848
N	3.76478849	1.94365918	3.36637854
N	7.75007710	1.94356922	3.36637854
N	1.77213916	3.88732842	3.36644892
N	5.75646784	3.88720830	3.36644892
Al	3.76418840	3.88715856	3.36645844
Al	7.75033693	3.88704849	3.36645844
Al	1.77195924	10.69097563	5.04921802
Al	5.75641757	10.69077560	5.04922807
N	7.75007710	10.69065601	5.04922807
N	3.76478849	10.69073591	5.04922807
N	1.77213916	0.97119961	5.04929792
N	5.75646784	0.97107948	5.04930798

Al	7.75033693	0.97091967	5.04930798
Al	3.76419846	0.97102974	5.04930798
Al	1.77195924	4.85925829	5.04931803
Al	5.75641757	4.85905826	5.04931803
N	3.76478849	4.85901805	5.04932809
N	7.75007710	4.85892809	5.04932809
N	1.77213916	6.80268728	5.04939794
N	5.75646784	6.80256716	5.04939794
Al	3.76419846	6.80252747	5.04940799
Al	7.75033693	6.80240735	5.04940799
C	5.31748633	3.92856789	13.73483400
C	4.65399833	4.64300040	14.91945052
C	4.83108273	3.88160050	16.23483753
H	4.47305439	4.33926236	12.77151123
H	6.41546277	3.96409822	13.85119615
H	5.07952219	2.85424220	13.69187055
H	5.06905251	5.66017336	15.00828135
H	3.57864138	4.77424865	14.71244776
H	4.37728572	2.88170119	16.16569313
H	5.89564676	3.74356676	16.47283847
H	4.35957153	4.40766075	17.07714670

Table S13. TS of second C-H activation on Ga/AlN through the stepwise SPI mechanism.
Total energy: -869.587 Hartree

Al	1.7686495406	1.9033978427	6.7296202766
Al	5.7539202585	1.9025720012	6.7310278962
N	3.7614815891	1.9027690458	6.7362310992
N	7.7479287368	1.9038709232	6.7354490574
Al	3.7662636731	3.8389013534	6.6959727949
Al	7.7404840647	3.8394136655	6.6945571363
N	1.7692207149	3.8379581715	6.6813819795
N	5.7523950397	3.8362901203	6.6967313968
Al	1.7732619637	7.7336661272	6.7359604438
Al	5.7504864900	7.7330585717	6.7369691051
N	3.7617233477	7.7324328548	6.7451309141
N	7.7487810166	7.7324154997	6.7362520423
N	1.7692029337	9.6689203464	6.6960783020
N	5.7557436752	9.6684967272	6.6963438258
Al	3.7630860449	9.6659071243	6.7044641519
Al	7.7472992319	9.6664769781	6.7035280959
Al	1.7619501815	4.7838571016	8.3426553318
Al	5.7503675376	4.7925370291	8.3843026091
N	7.7308332455	4.7835458183	8.3720131363
N	3.7676715574	4.7758253660	8.3839333147
N	1.7752515941	6.7156284717	8.3858736786
N	5.7442128651	6.7215321481	8.3869989297

Al	7.7422571624	6.7219284266	8.3802574778
Al	3.7620670700	6.7148701067	8.4129401868
Al	1.7704030711	10.5955447154	8.3853736881
Al	5.7506816629	10.5938047878	8.3895438179
N	3.7635482998	10.5836144942	8.4020192567
N	7.7444518519	10.5913421447	8.3972237980
N	1.7677462556	0.8660933281	8.3652357599
N	5.7519749771	0.8725241889	8.3728073516
Al	3.7609213663	0.8551536875	8.3618946923
Al	7.7437145711	0.8593017582	8.3625299030
Al	1.7849600046	7.6563786875	10.1093683193
Al	5.7356349444	7.6299515108	10.1140835698
N	3.7659368458	7.6101786187	10.1549737006
N	7.7420586381	7.6674471248	10.0377168927
N	1.7732995448	9.5939952511	10.0388581340
N	5.7476399537	9.5861082338	10.0507236122
Al	3.7635136984	9.5619159330	10.0904879459
Al	7.7422984386	9.5791846322	10.0671548090
Al	1.7597150087	1.8168884769	10.0570789196
Al	5.7514845630	1.8143818779	10.0816011327
N	3.7571795674	1.8017958947	10.0462063537
N	7.7409886546	1.8081339838	10.0443613026
N	1.7591770445	3.7365995753	9.9335891490
N	5.7345117627	3.7504090545	10.0590171320
Al	3.7530782903	3.7228422453	9.9972301019
Al	7.7395130543	3.7423732567	10.0175350989
Al	1.7629689153	10.6137076510	11.6535098878
Al	5.7537951699	10.6148094657	11.6599225443
N	3.7578811055	10.5047203902	11.7824095815
N	7.7463042652	10.5150233463	11.7688231370
N	1.7675028997	0.8614188182	11.7476348793
N	5.7480633667	0.8587267347	11.7649568659
Al	3.7524761004	0.7296488904	11.6243071148
Al	7.7478499826	0.7496335901	11.6343593588
Al	1.6861438534	4.8750312499	11.4825657555
Ga	5.8738140054	4.7628579877	11.8166089742
N	3.5646439750	4.6522765217	11.6837675183
N	7.8414499337	4.6803055730	11.6818420690
N	1.7165074789	6.7451368607	11.7686006414
N	5.8215493613	6.7043763693	11.7908668429
Al	3.7493639005	6.6405325383	11.9626870191
Al	7.7600721531	6.6210899161	11.6339277070
Al	1.7719592389	1.9440391324	0.0000000000
Al	1.7719592389	7.7759670844	0.0000000000
Al	3.7641884040	3.8873083123	0.0000000000
Al	5.7564175691	1.9438391035	0.0000000000

Al	5.7564175691	7.7757670555	0.0000000000
Al	7.7503369255	3.8871887183	0.0000000000
N	1.7721391590	3.8874787073	0.0000000000
N	3.7647884907	1.9437994152	0.0000000000
N	3.7647884907	7.7757167836	0.0000000000
N	5.7564678409	3.8873585841	0.0000000000
N	7.7500770996	1.9437094552	0.0000000000
N	7.7500770996	7.7756368779	0.0000000000
N	1.7721391590	9.7193960757	0.0003100977
N	5.7564678409	9.7192759525	0.0003100977
Al	3.7641884040	9.7192262099	0.0003100977
Al	7.7503369255	9.7191161411	0.0003100977
Al	1.7719592389	4.8598282154	1.6830791991
Al	5.7564175691	4.8596281865	1.6830791991
N	7.7500770996	4.8594980089	1.6830892535
N	3.7647884907	4.8595879690	1.6830892535
N	1.7721391590	6.8032672611	1.6831591048
N	5.7564678409	6.8031471379	1.6831591048
Al	7.7503369255	6.8029873265	1.6831591048
Al	3.7641984583	6.8030973953	1.6831591048
Al	1.7719592389	10.6913259464	1.6831792136
Al	5.7564175691	10.6911259175	1.6831792136
N	3.7647884907	10.6910857001	1.6831792136
N	7.7500770996	10.6909957400	1.6831792136
N	1.7721391590	0.9715398673	1.6832495941
N	5.7564678409	0.9714197441	1.6832495941
Al	3.7641984583	0.9713694723	1.6832591193
Al	7.7503369255	0.9712594035	1.6832591193
Al	1.7719592389	7.7751870775	3.3660287499
Al	5.7564175691	7.7749870486	3.3660287499
N	3.7647884907	7.7749468311	3.3660287499
N	7.7500770996	7.7748568710	3.3660287499
N	1.7721391590	9.7186261232	3.3661086556
N	5.7564678409	9.7185060000	3.3661086556
Al	3.7641884040	9.7184562574	3.3661086556
Al	7.7503369255	9.7183361342	3.3661086556
Al	1.7719592389	1.9438994297	3.3663684815
Al	5.7564175691	1.9436994008	3.3663684815
N	3.7647884907	1.9436591833	3.3663785359
N	7.7500770996	1.9435692233	3.3663785359
N	1.7721391590	3.8873284211	3.3664489164
N	5.7564678409	3.8872082979	3.3664489164
Al	3.7641884040	3.8871585552	3.3664584416
Al	7.7503369255	3.8870484864	3.3664584416
Al	1.7719592389	10.6909756312	5.0492180178
Al	5.7564175691	10.6907756023	5.0492280722

N	7.7500770996	10.6906560083	5.0492280722
N	3.7647884907	10.6907359141	5.0492280722
N	1.7721391590	0.9711996065	5.0492979235
N	5.7564678409	0.9710794833	5.0493079779
Al	7.7503369255	0.9709196718	5.0493079779
Al	3.7641984583	0.9710297406	5.0493079779
Al	1.7719592389	4.8592582918	5.0493180323
Al	5.7564175691	4.8590582629	5.0493180323
N	3.7647884907	4.8590180454	5.0493280866
N	7.7500770996	4.8589280853	5.0493280866
N	1.7721391590	6.8026872831	5.0493979380
N	5.7564678409	6.8025671599	5.0493979380
Al	3.7641984583	6.8025274717	5.0494079924
Al	7.7503369255	6.8024073485	5.0494079924
C	5.6110772593	3.9732290177	13.8860459853
C	4.6620108665	4.6745352898	14.6374209907
C	3.3382925344	4.0704299827	14.9964049902
H	3.7068841253	4.0908613722	12.5235594006
H	3.9242647549	6.3210491378	13.6540896764
H	5.3861199422	2.9343947948	13.6035177545
H	6.6672852408	4.1701313135	14.0788761348
H	4.9951182779	5.5116952814	15.2478359976
H	2.9815024762	3.3411335254	14.2515806666
H	2.5562560039	4.8185579214	15.1666408288
H	3.4696513865	3.4948863907	15.9312104065

Table S14. TS of H_2 production on Ga/AlN through the stepwise PSD mechanism.
Total energy: -869.572 Hartree

Al	1.7765268084	1.9606839510	6.7150152247
Al	5.7617435917	1.9602713125	6.7162043112
N	3.7689644366	1.9592263093	6.7133966876
N	7.7552074699	1.9599583421	6.7116350083
Al	3.7736258675	3.8940439066	6.7230672523
Al	7.7506810914	3.8939071364	6.7212610109
N	1.7775387648	3.8942219315	6.7177256350
N	5.7607668168	3.8932594104	6.7260662856
Al	1.7757779462	7.7895684242	6.7116779887
Al	5.7623409206	7.7888289700	6.7120918855
N	3.7684437097	7.7909660440	6.7089416722
N	7.7555456332	7.7908716314	6.7080202367
N	1.7759602405	9.7240630128	6.7185384041
N	5.7619304715	9.7232108253	6.7182122246
Al	3.7695958564	9.7245582887	6.7179852304
Al	7.7543912101	9.7246724510	6.7175208267
Al	1.7818331890	4.8796766548	8.3717333642
Al	5.7645927823	4.8855032937	8.4007034438

N	7.7516376360	4.8765464372	8.3825480895
N	3.7809344878	4.8799905960	8.3905274186
N	1.7791042276	6.8183005627	8.3826763312
N	5.7667751245	6.8208079063	8.3824869831
Al	7.7563016793	6.8123180382	8.3703443041
Al	3.7757401883	6.8130343992	8.3699219727
Al	1.7798478096	10.7294460399	8.3659755649
Al	5.7657731330	10.7276641739	8.3683404630
N	3.7745140602	10.7212441292	8.3740614977
N	7.7570613536	10.7221612278	8.3737866241
N	1.7795952122	0.9989143043	8.3913066464
N	5.7662760408	1.0014192606	8.3962501195
Al	3.7740659676	0.9927705726	8.3833940922
Al	7.7579246552	0.9950389114	8.3827499905
Al	1.7844044208	7.8291512650	10.0538209887
Al	5.7681215938	7.8093255530	10.0602159616
N	3.7821583822	7.8137807883	10.0203737310
N	7.7557187192	7.8210689167	10.0186850099
N	1.7841175837	9.7569466836	10.0298175286
N	5.7673355388	9.7519900369	10.0376435839
Al	3.7788636545	9.7483168533	10.0608345457
Al	7.7589018368	9.7493215740	10.0590936683
Al	1.7832337572	1.9846128008	10.0783805628
Al	5.7699747976	1.9816766565	10.0932483236
N	3.7762013000	1.9677674509	10.0589591079
N	7.7627079971	1.9785341733	10.0556736229
N	1.7956979931	3.9094168364	10.0205700451
N	5.7599448703	3.9119127741	10.0972643881
Al	3.7851846222	3.9078831726	10.0882500373
Al	7.7556854365	3.9252577701	10.0802418904
Al	1.7837324494	10.7866307795	11.6390975964
Al	5.7712042991	10.7874453049	11.6430139942
N	3.7764483573	10.6797945624	11.7574658819
N	7.7639097876	10.6822276170	11.7532992964
N	1.7838099502	1.0262686329	11.7674402443
N	5.7712463997	1.0253360533	11.7797303006
Al	3.7740651880	0.9155104126	11.6498727471
Al	7.7677190857	0.9190323249	11.6467581638
Al	1.7582130448	5.0345477804	11.5810794899
Ga	5.8030370787	4.8990650840	11.9138476947
N	3.6685067385	4.8935793356	11.7765799885
N	7.8595016864	4.9179629397	11.7070030552
N	1.7926645198	6.9452576761	11.7529055624
N	5.7638733735	6.9129886635	11.7908200247
Al	3.7815729631	6.8115479722	11.6426878267
Al	7.7622043203	6.8033097487	11.6255490082

Al	1.7719587097	1.9440386033	0.0000000000
Al	1.7719587097	7.7759639093	0.0000000000
Al	3.7641868164	3.8873067248	0.0000000000
Al	5.7564154524	1.9438385743	0.0000000000
Al	5.7564154524	7.7757638804	0.0000000000
Al	7.7503337505	3.8871871308	0.0000000000
N	1.7721386299	3.8874771198	0.0000000000
N	3.7647869032	1.9437988861	0.0000000000
N	3.7647869032	7.7757136086	0.0000000000
N	5.7564657242	3.8873569966	0.0000000000
N	7.7500739246	1.9437089260	0.0000000000
N	7.7500739246	7.7756337028	0.0000000000
N	1.7721386299	9.7193923715	0.0003100977
N	5.7564657242	9.7192722483	0.0003100977
Al	3.7641868164	9.7192225057	0.0003100977
Al	7.7503337505	9.7191124368	0.0003100977
Al	1.7719587097	4.8598260987	1.6830786699
Al	5.7564154524	4.8596260698	1.6830786699
N	7.7500739246	4.8594958922	1.6830887243
N	3.7647869032	4.8595858523	1.6830887243
N	1.7721386299	6.8032646152	1.6831585757
N	5.7564657242	6.8031444920	1.6831585757
Al	7.7503337505	6.8029846806	1.6831585757
Al	3.7641968708	6.8030947494	1.6831585757
Al	1.7719587097	10.6913217130	1.6831786844
Al	5.7564154524	10.6911216841	1.6831786844
N	3.7647869032	10.6910814666	1.6831786844
N	7.7500739246	10.6909915066	1.6831786844
N	1.7721386299	0.9715393381	1.6832490649
N	5.7564657242	0.9714192149	1.6832490649
Al	3.7641968708	0.9713689431	1.6832585901
Al	7.7503337505	0.9712588743	1.6832585901
Al	1.7719587097	7.7751839024	3.3660271623
Al	5.7564154524	7.7749838735	3.3660271623
N	3.7647869032	7.7749436560	3.3660271623
N	7.7500739246	7.7748536960	3.3660271623
N	1.7721386299	9.7186224189	3.3661075972
N	5.7564657242	9.7185022958	3.3661075972
Al	3.7641868164	9.7184525531	3.3661075972
Al	7.7503337505	9.7183324299	3.3661075972
Al	1.7719587097	1.9438989005	3.3663668940
Al	5.7564154524	1.9436988716	3.3663668940
N	3.7647869032	1.9436586542	3.3663774775
N	7.7500739246	1.9435686941	3.3663774775
N	1.7721386299	3.8873268335	3.3664473289
N	5.7564657242	3.8872067103	3.3664473289

Al	3.7641868164	3.8871569677	3.3664568541
Al	7.7503337505	3.8870468989	3.3664568541
Al	1.7719587097	10.6909713978	5.0492159011
Al	5.7564154524	10.6907713689	5.0492259555
N	7.7500739246	10.6906517749	5.0492259555
N	3.7647869032	10.6907316807	5.0492259555
N	1.7721386299	0.9711990773	5.0492958068
N	5.7564657242	0.9710789541	5.0493058612
Al	7.7503337505	0.9709191426	5.0493058612
Al	3.7641968708	0.9710292115	5.0493058612
Al	1.7719587097	4.8592561751	5.0493159156
Al	5.7564154524	4.8590561462	5.0493159156
N	3.7647869032	4.8590159287	5.0493259699
N	7.7500739246	4.8589259686	5.0493259699
N	1.7721386299	6.8026846372	5.0493958213
N	5.7564657242	6.8025645141	5.0493958213
Al	3.7641968708	6.8025248258	5.0494058756
Al	7.7503337505	6.8024047026	5.0494058756
C	6.1304957112	4.3377909525	14.1077303276
C	5.0697772493	4.3670170378	15.0134526859
C	4.6118753751	3.1526319969	15.7672355746
H	3.4865301274	4.5986344949	13.3627270414
H	3.5812488556	4.5190332946	14.2094177829
H	6.7980791181	5.2010846829	14.1043384836
H	6.6093664966	3.3745439274	13.9104218287
H	4.8352300417	5.3329934854	15.4716919834
H	5.2494587404	3.0438097166	16.6615445864
H	4.7281755606	2.2416904262	15.1658080713
H	3.5720016430	3.2314357715	16.1091819169

Table S15. TS of H_2 production on Ga/AlN through the stepwise PSI/SP mechanism.
Total energy: -848.941 Hartree

Al	1.77109847	1.94121852	6.71336666
Al	5.75565999	1.94110280	6.71378697
N	3.76364431	1.94396955	6.71428008
N	7.74940171	1.94473973	6.71269203
Al	3.76362390	3.87433547	6.70967399
Al	7.74894622	3.87391852	6.70726032
N	1.77140100	3.87242324	6.70519166
N	5.75549090	3.87252137	6.70620304
Al	1.77338746	7.77172417	6.71411017
Al	5.75353066	7.77140600	6.71463644
N	3.76388514	7.77303104	6.72069438
N	7.74930565	7.77189204	6.71226398
N	1.77030504	9.70162964	6.70364783
N	5.75693342	9.70151465	6.70420535

Al	3.76361120	9.70281420	6.70824346
Al	7.74930505	9.70282177	6.70748663
Al	1.77389921	4.85510524	8.36036628
Al	5.75093600	4.85653775	8.36164880
N	7.74829007	4.84569165	8.37523334
N	3.76279311	4.84649092	8.38344306
N	1.77506100	6.78460260	8.37065588
N	5.75095795	6.78379790	8.37318866
Al	7.74887547	6.78513127	8.36511051
Al	3.76261446	6.78259806	8.38760236
Al	1.77071229	10.67877348	8.36542336
Al	5.75494655	10.67857935	8.36575762
N	3.76307386	10.67232373	8.37400255
N	7.74903790	10.67698292	8.37151538
N	1.77040994	0.95001556	8.36920497
N	5.75549589	0.94942387	8.37000913
Al	3.76220960	0.94074940	8.36186992
Al	7.74922117	0.94309322	8.36011211
Al	1.77351352	7.75659713	10.05730717
Al	5.75132174	7.75601632	10.05936590
N	3.76497134	7.74828395	10.08072347
N	7.74708218	7.77050274	10.00718702
N	1.77262584	9.69455314	10.02369529
N	5.75284374	9.69526416	10.02488587
Al	3.76283388	9.68702629	10.06247510
Al	7.74800897	9.68989235	10.05074214
Al	1.77071806	1.91639246	10.05960932
Al	5.75293587	1.91014421	10.06091469
N	3.76031882	1.91007995	10.03450533
N	7.74986314	1.92100931	10.02725440
N	1.78533581	3.85885822	10.01557795
N	5.73649355	3.84342306	10.00860061
Al	3.75926267	3.83932939	10.05522585
Al	7.75035793	3.84562632	10.03863189
Al	1.76646721	10.73196776	11.63171876
Al	5.75735527	10.73086394	11.63327571
N	3.76179720	10.62666009	11.75054773
N	7.74866275	10.63124151	11.73684723
N	1.76796514	0.96964318	11.73877569
N	5.75616511	0.96780659	11.73906887
Al	3.76066340	0.86159071	11.63653394
Al	7.74881649	0.86681964	11.63130118
Al	1.73429198	4.92956139	11.59488552
Ga	5.79474251	4.91135846	11.67283867
N	3.70166666	4.78261189	11.83279708
N	7.79301110	4.79659849	11.71608111

N	1.73262769	6.81920200	11.71341123
N	5.79761507	6.85592201	11.74855354
Al	3.74771228	6.83171249	11.81139934
Al	7.75871615	6.73096627	11.61440591
Al	1.77196000	1.94404000	0.00000000
Al	1.77196000	7.77597000	0.00000000
Al	3.76419000	3.88731000	0.00000000
Al	5.75642000	1.94384000	0.00000000
Al	5.75642000	7.77577000	0.00000000
Al	7.75034000	3.88719000	0.00000000
N	1.77214000	3.88748000	0.00000000
N	3.76479000	1.94380000	0.00000000
N	3.76479000	7.77572000	0.00000000
N	5.75647000	3.88736000	0.00000000
N	7.75008000	1.94371000	0.00000000
N	7.75008000	7.77564000	0.00000000
N	1.77214000	9.71940000	0.00031000
N	5.75647000	9.71928000	0.00031000
Al	3.76419000	9.71923000	0.00031000
Al	7.75034000	9.71912000	0.00031000
Al	1.77196000	4.85983000	1.68308000
Al	5.75642000	4.85963000	1.68308000
N	7.75008000	4.85950000	1.68309000
N	3.76479000	4.85959000	1.68309000
N	1.77214000	6.80327000	1.68316000
N	5.75647000	6.80315000	1.68316000
Al	7.75034000	6.80299000	1.68316000
Al	3.76420000	6.80310000	1.68316000
Al	1.77196000	10.69133000	1.68318000
Al	5.75642000	10.69113000	1.68318000
N	3.76479000	10.69109000	1.68318000
N	7.75008000	10.69100000	1.68318000
N	1.77214000	0.97154000	1.68325000
N	5.75647000	0.97142000	1.68325000
Al	3.76420000	0.97137000	1.68326000
Al	7.75034000	0.97126000	1.68326000
Al	1.77196000	7.77519000	3.36603000
Al	5.75642000	7.77499000	3.36603000
N	3.76479000	7.77495000	3.36603000
N	7.75008000	7.77486000	3.36603000
N	1.77214000	9.71863000	3.36611000
N	5.75647000	9.71851000	3.36611000
Al	3.76419000	9.71846000	3.36611000
Al	7.75034000	9.71834000	3.36611000
Al	1.77196000	1.94390000	3.36637000
Al	5.75642000	1.94370000	3.36637000

N	3.76479000	1.94366000	3.36638000
N	7.75008000	1.94357000	3.36638000
N	1.77214000	3.88733000	3.36645000
N	5.75647000	3.88721000	3.36645000
Al	3.76419000	3.88716000	3.36646000
Al	7.75034000	3.88705000	3.36646000
Al	1.77196000	10.69098000	5.04922000
Al	5.75642000	10.69078000	5.04923000
N	7.75008000	10.69066000	5.04923000
N	3.76479000	10.69074000	5.04923000
N	1.77214000	0.97120000	5.04930000
N	5.75647000	0.97108000	5.04931000
Al	7.75034000	0.97092000	5.04931000
Al	3.76420000	0.97103000	5.04931000
Al	1.77196000	4.85926000	5.04932000
Al	5.75642000	4.85906000	5.04932000
N	3.76479000	4.85902000	5.04933000
N	7.75008000	4.85893000	5.04933000
N	1.77214000	6.80269000	5.04940000
N	5.75647000	6.80257000	5.04940000
Al	3.76420000	6.80253000	5.04941000
Al	7.75034000	6.80241000	5.04941000
H	3.75293248	6.38016024	13.52457447
H	3.72661249	5.42107207	12.96814164

Table S16. TS of first C-H activation on Ga/AlN through the stepwise SP mechanism.
Total energy: -869.617 Hartree

Al	1.76872544	1.93563021	6.71711980
Al	5.75305273	1.93475629	6.71805622
N	3.76099197	1.93594595	6.71873419
N	7.74762032	1.93618095	6.71760349
Al	3.76430213	3.86686751	6.71056530
Al	7.74190895	3.86711134	6.70769693
N	1.76863747	3.86755777	6.69991548
N	5.75243986	3.86593910	6.71275845
Al	1.76786080	7.76446615	6.71857842
Al	5.75346595	7.76360925	6.71940165
N	3.76078986	7.76488757	6.72085366
N	7.74735199	7.76505317	6.71957806
N	1.76861974	9.69655633	6.70382774
N	5.75453378	9.69569709	6.70385101
Al	3.76208753	9.69764642	6.70774002
Al	7.74627997	9.69761258	6.70741180
Al	1.76437621	4.82635882	8.36352222
Al	5.74715401	4.83412227	8.39295619
N	7.73300017	4.82532269	8.37910191

N	3.76449973	4.82382412	8.38977193
N	1.76459029	6.76495566	8.37415882
N	5.75003846	6.76590002	8.37397389
Al	7.74045459	6.75842399	8.36465309
Al	3.75884026	6.75753180	8.36640453
Al	1.76705640	10.67195951	8.36535681
Al	5.75075209	10.66933619	8.36903448
N	3.76217738	10.66297280	8.37818975
N	7.74279336	10.66375466	8.37760479
N	1.76743928	0.93930109	8.37064726
N	5.75121240	0.94404991	8.37490650
Al	3.75965242	0.93658575	8.36985816
Al	7.74362254	0.93552863	8.36666759
Al	1.76203097	7.75146491	10.05667361
Al	5.74523281	7.72267440	10.06430271
N	3.76220963	7.72549854	10.03425478
N	7.73108800	7.73627319	10.02431562
N	1.76658790	9.67426466	10.01308588
N	5.74781676	9.66196588	10.02328792
Al	3.75916437	9.66368438	10.05362737
Al	7.73954474	9.66494716	10.04954262
Al	1.76284617	1.90498976	10.06188888
Al	5.74862570	1.90006585	10.07785884
N	3.75720586	1.90099928	10.05024410
N	7.74142399	1.89924654	10.03960684
N	1.75323066	3.82821130	9.99242492
N	5.74211426	3.83931733	10.08632210
Al	3.74458170	3.84379874	10.07994343
Al	7.75102395	3.83489578	10.05188684
Al	1.76276563	10.68904147	11.63438538
Al	5.75021919	10.68786101	11.63323778
N	3.75607396	10.58466920	11.75367694
N	7.74553594	10.57788927	11.74948367
N	1.76084116	0.92608114	11.72925107
N	5.75734688	0.91366141	11.72508172
Al	3.74988139	0.82034797	11.62542371
Al	7.74571701	0.81539965	11.61544720
Al	1.72256477	4.94498887	11.57524644
Ga	5.87448603	4.83356801	11.82467827
N	3.57516905	4.80543540	11.74176311
N	7.83671292	4.81854868	11.72594412
N	1.74878217	6.86676971	11.75224979
N	5.74110489	6.80054782	11.79405631
Al	3.73494840	6.69885753	11.64354961
Al	7.76001357	6.73414783	11.64186242
Al	1.77195924	1.94403913	0.00000000

Al	1.77195924	7.77596708	0.00000000
Al	3.76418840	3.88730831	0.00000000
Al	5.75641757	1.94383910	0.00000000
Al	5.75641757	7.77576706	0.00000000
Al	7.75033693	3.88718872	0.00000000
N	1.77213916	3.88747871	0.00000000
N	3.76478849	1.94379942	0.00000000
N	3.76478849	7.77571678	0.00000000
N	5.75646784	3.88735858	0.00000000
N	7.75007710	1.94370946	0.00000000
N	7.75007710	7.77563688	0.00000000
N	1.77213916	9.71939608	0.00031010
N	5.75646784	9.71927595	0.00031010
Al	3.76418840	9.71922621	0.00031010
Al	7.75033693	9.71911614	0.00031010
Al	1.77195924	4.85982822	1.68307920
Al	5.75641757	4.85962819	1.68307920
N	7.75007710	4.85949801	1.68308925
N	3.76478849	4.85958797	1.68308925
N	1.77213916	6.80326726	1.68315910
N	5.75646784	6.80314714	1.68315910
Al	7.75033693	6.80298733	1.68315910
Al	3.76419846	6.80309740	1.68315910
Al	1.77195924	10.69132595	1.68317921
Al	5.75641757	10.69112592	1.68317921
N	3.76478849	10.69108570	1.68317921
N	7.75007710	10.69099574	1.68317921
N	1.77213916	0.97153987	1.68324959
N	5.75646784	0.97141974	1.68324959
Al	3.76419846	0.97136947	1.68325912
Al	7.75033693	0.97125940	1.68325912
Al	1.77195924	7.77518708	3.36602875
Al	5.75641757	7.77498705	3.36602875
N	3.76478849	7.77494683	3.36602875
N	7.75007710	7.77485687	3.36602875
N	1.77213916	9.71862612	3.36610866
N	5.75646784	9.71850600	3.36610866
Al	3.76418840	9.71845626	3.36610866
Al	7.75033693	9.71833613	3.36610866
Al	1.77195924	1.94389943	3.36636848
Al	5.75641757	1.94369940	3.36636848
N	3.76478849	1.94365918	3.36637854
N	7.75007710	1.94356922	3.36637854
N	1.77213916	3.88732842	3.36644892
N	5.75646784	3.88720830	3.36644892
Al	3.76418840	3.88715856	3.36645844

Al	7.75033693	3.88704849	3.36645844
Al	1.77195924	10.69097563	5.04921802
Al	5.75641757	10.69077560	5.04922807
N	7.75007710	10.69065601	5.04922807
N	3.76478849	10.69073591	5.04922807
N	1.77213916	0.97119961	5.04929792
N	5.75646784	0.97107948	5.04930798
Al	7.75033693	0.97091967	5.04930798
Al	3.76419846	0.97102974	5.04930798
Al	1.77195924	4.85925829	5.04931803
Al	5.75641757	4.85905826	5.04931803
N	3.76478849	4.85901805	5.04932809
N	7.75007710	4.85892809	5.04932809
N	1.77213916	6.80268728	5.04939794
N	5.75646784	6.80256716	5.04939794
Al	3.76419846	6.80252747	5.04940799
Al	7.75033693	6.80240735	5.04940799
C	4.72022393	5.15813121	14.80740930
C	5.12029777	4.00005264	13.89003590
C	4.31538466	2.72626981	14.19118056
H	5.35383699	6.04370339	14.66152801
H	4.38998303	4.33155899	12.77628797
H	3.67764884	5.46191915	14.60968446
H	4.77201579	4.86619385	15.86827180
H	6.19228349	3.77088953	14.01425569
H	4.79368569	1.85044091	13.72839399
H	3.27624670	2.80390484	13.82568772
H	4.25697045	2.53613994	15.27545962

Table S17. TS of second C-H activation on Ga/AlN through the stepwise SP mechanism.
Total energy: -869.581 Hartree

Al	1.75692767	1.93068017	6.72131121
Al	5.74188975	1.92991068	6.72335167
N	3.75038818	1.93080581	6.72495409
N	7.73680487	1.93186357	6.72461542
Al	3.75417628	3.86500807	6.70700698
Al	7.72815008	3.86531443	6.70595587
N	1.75794283	3.86473807	6.69628844
N	5.74068004	3.86307418	6.71087805
Al	1.76228420	7.76131102	6.72356739
Al	5.73899634	7.76057496	6.72480156
N	3.75099335	7.76152229	6.73154932
N	7.73846135	7.75971533	6.72189133
N	1.75879688	9.69497521	6.70533644
N	5.74599879	9.69426064	6.70585736
Al	3.75228497	9.69344763	6.71039628

Al	7.73640419	9.69338049	6.71020802
Al	1.73951554	4.83514439	8.34743383
Al	5.72534859	4.84291758	8.38697620
N	7.70585404	4.83107511	8.37568904
N	3.74489584	4.82462043	8.38706765
N	1.75584389	6.76721529	8.38214245
N	5.71972101	6.77260638	8.38309660
Al	7.71988606	6.77224109	8.37462579
Al	3.73950273	6.76676703	8.40770828
Al	1.74942431	10.65561119	8.37688834
Al	5.72995763	10.65361882	8.38141130
N	3.74351970	10.64476478	8.38915511
N	7.72417096	10.65214592	8.38574128
N	1.74635194	0.92686240	8.37362434
N	5.72941131	0.93213694	8.38094183
Al	3.73845156	0.91507623	8.36824692
Al	7.72170431	0.91862131	8.36875592
Al	1.74770118	7.73384071	10.08777826
Al	5.70994897	7.70923840	10.09174427
N	3.73447302	7.69327501	10.13566519
N	7.71211406	7.74650260	10.01293247
N	1.74509530	9.67149718	10.03502382
N	5.71664058	9.66369140	10.04664334
Al	3.73303767	9.64327420	10.08448513
Al	7.71273327	9.65731918	10.06216129
Al	1.72708909	1.88868902	10.06334123
Al	5.71728186	1.88427842	10.08567124
N	3.72203098	1.87476391	10.05007488
N	7.70817488	1.88110158	10.04586922
N	1.72656877	3.81467539	9.95816765
N	5.69435578	3.81954221	10.07140970
Al	3.71838223	3.79536455	10.01776475
Al	7.70019051	3.81160891	10.03538996
Al	1.72809222	10.69527518	11.64812934
Al	5.71858662	10.69463500	11.65510830
N	3.72201458	10.58602347	11.77481623
N	7.71144953	10.59590934	11.75957883
N	1.73013708	0.94199116	11.75683149
N	5.71150653	0.93817024	11.77082777
Al	3.71701098	0.81191928	11.63747881
Al	7.71052847	0.83075600	11.64470790
Al	1.64369292	4.95754936	11.49673007
Ga	5.80606821	4.84684735	11.82122904
N	3.53774359	4.74704312	11.71590466
N	7.78761999	4.74828686	11.70278244
N	1.67080426	6.82279171	11.74203428

N	5.79263254	6.78441384	11.75670236
Al	3.71331480	6.76993528	11.92807120
Al	7.71511944	6.69856618	11.61517324
Al	1.77196000	1.94404000	0.00000000
Al	1.77196000	7.77596000	0.00000000
Al	3.76419000	3.88731000	0.00000000
Al	5.75642000	1.94384000	0.00000000
Al	5.75642000	7.77576000	0.00000000
Al	7.75033000	3.88719000	0.00000000
N	1.77214000	3.88748000	0.00000000
N	3.76479000	1.94380000	0.00000000
N	3.76479000	7.77571000	0.00000000
N	5.75647000	3.88736000	0.00000000
N	7.75007000	1.94371000	0.00000000
N	7.75007000	7.77563000	0.00000000
N	1.77214000	9.71939000	0.00031000
N	5.75647000	9.71927000	0.00031000
Al	3.76419000	9.71922000	0.00031000
Al	7.75033000	9.71911000	0.00031000
Al	1.77196000	4.85983000	1.68308000
Al	5.75642000	4.85963000	1.68308000
N	7.75007000	4.85950000	1.68309000
N	3.76479000	4.85959000	1.68309000
N	1.77214000	6.80326000	1.68316000
N	5.75647000	6.80314000	1.68316000
Al	7.75033000	6.80298000	1.68316000
Al	3.76420000	6.80309000	1.68316000
Al	1.77196000	10.69132000	1.68318000
Al	5.75642000	10.69112000	1.68318000
N	3.76479000	10.69108000	1.68318000
N	7.75007000	10.69099000	1.68318000
N	1.77214000	0.97154000	1.68325000
N	5.75647000	0.97142000	1.68325000
Al	3.76420000	0.97137000	1.68326000
Al	7.75033000	0.97126000	1.68326000
Al	1.77196000	7.77518000	3.36603000
Al	5.75642000	7.77498000	3.36603000
N	3.76479000	7.77494000	3.36603000
N	7.75007000	7.77485000	3.36603000
N	1.77214000	9.71862000	3.36611000
N	5.75647000	9.71850000	3.36611000
Al	3.76419000	9.71845000	3.36611000
Al	7.75033000	9.71833000	3.36611000
Al	1.77196000	1.94390000	3.36637000
Al	5.75642000	1.94370000	3.36637000
N	3.76479000	1.94366000	3.36638000

N	7.75007000	1.94357000	3.36638000
N	1.77214000	3.88733000	3.36645000
N	5.75647000	3.88721000	3.36645000
Al	3.76419000	3.88716000	3.36646000
Al	7.75033000	3.88705000	3.36646000
Al	1.77196000	10.69097000	5.04922000
Al	5.75642000	10.69077000	5.04923000
N	7.75007000	10.69065000	5.04923000
N	3.76479000	10.69073000	5.04923000
N	1.77214000	0.97120000	5.04930000
N	5.75647000	0.97108000	5.04931000
Al	7.75033000	0.97092000	5.04931000
Al	3.76420000	0.97103000	5.04931000
Al	1.77196000	4.85926000	5.04932000
Al	5.75642000	4.85906000	5.04932000
N	3.76479000	4.85902000	5.04933000
N	7.75007000	4.85893000	5.04933000
N	1.77214000	6.80268000	5.04940000
N	5.75647000	6.80256000	5.04940000
Al	3.76420000	6.80252000	5.04941000
Al	7.75033000	6.80240000	5.04941000
C	6.71655623	3.88145213	14.63688026
C	5.32498032	4.13414045	14.09321613
C	4.49965982	5.11602113	14.63720518
H	7.20703317	4.81819951	14.93347606
H	6.65121196	3.23276630	15.52479100
H	7.36497655	3.39112463	13.90001848
H	3.64123762	4.19090086	12.56635799
H	4.82402853	3.22170248	13.73463172
H	3.93508644	6.68088710	13.60415911
H	4.91048770	5.86610837	15.31295398
H	3.41779968	5.01205611	14.66252186

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