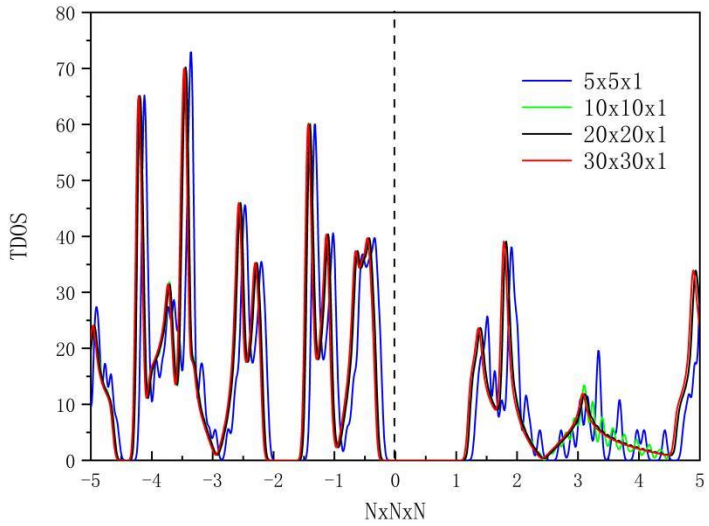

Fig. S1 The convergence tests of total density of states with an increasing k-point grids of N×N×N for pure g-CN monolayer.

The optimized structure coordination of pure g-CN monolayer:

1.00000000000000
14.2329998016000001 0.0000000000000000 0.0000000000000000
-7.1164999008000001 12.3261394002000007 0.0000000000000000
0.0000000000000000 0.0000000000000000 25.0000000000000000
C N
24 24
Direct
0.2197116730115027 0.2800998494882521 0.5008686762526153
0.2197115434903196 0.7800998115366440 0.5008686734222962
0.7197116701909039 0.2800998435081468 0.5008686679990504
0.7197115342653433 0.7800998163032560 0.5008686737948693
0.2808806973986719 0.2189272842200793 0.5008738470132101
0.2808805747539296 0.7189272565823899 0.5008738440063709
0.7808807193828358 0.2189273439438324 0.5008738373976840
0.7808805848730671 0.7189272965227360 0.5008738418101260
0.2196022495707552 0.4382849885421436 0.5008794060852040
0.2196021857468224 0.9382849670347326 0.5008793999394427
0.7196022109722477 0.4382849715442418 0.5008794024264347
0.7196021853584469 0.9382850150392272 0.5008794061770487
0.2807695263466385 0.0606354622896603 0.5008792785952707
0.2807695311090939 0.5606355027063472 0.5008792810175891

0.7807695468904612 0.0606355214792345 0.5008792813449574
0.7807694922896715 0.5606355000243823 0.5008792812077800
0.0614209034673934 0.2800145027687861 0.5008784785916092
0.0614208064079733 0.7800145188641565 0.5008784748226134
0.5614208894607771 0.2800144683426211 0.5008784790148582
0.5614208070069253 0.7800145874664040 0.5008784787106322
0.4390691237184043 0.2188363197214542 0.5008771191429560
0.4390690459615385 0.7188364236616209 0.5008771195402417
0.9390691502727719 0.2188363829130537 0.5008771217033683
0.9390690456356339 0.7188363921963870 0.5008771150786018
0.2786050686979564 0.3886892676098057 0.5008700020105792
0.2786049723780479 0.8886892162052078 0.5008699937629615
0.7786050360514167 0.3886892734653600 0.5008699882343919
0.7786049288062102 0.8886892526551051 0.5008699986813028
0.2218751454728789 0.1103229410246556 0.5008815044008230
0.2218750653867616 0.6103229108770165 0.5008815012931009
0.7218751735597735 0.1103230036228666 0.5008814925066076
0.7218750854970111 0.6103229537271062 0.5008815036398673
0.1110177055853185 0.3886119958957579 0.5008848273304238
0.1110176372718996 0.8886120080183275 0.5008848212356085
0.6110176360739459 0.3886119671732260 0.5008848291827519
0.6110175955886419 0.8886120749107249 0.5008848280181653
0.3893772606606092 0.1102479385638375 0.5008769563232391
0.3893772536328015 0.6102480522540716 0.5008769606334127
0.8893772735547216 0.1102479906292828 0.5008769689251409
0.8893772153328712 0.6102480043219387 0.5008769608223531
0.1111198018431523 0.2211170994591711 0.5008610970804241
0.1111196782809500 0.7211170935232474 0.5008610999902388
0.6111198115304575 0.2211170946649545 0.5008610985347701
0.6111196907762491 0.7211171371797960 0.5008610984292190
0.3894708151325688 0.2778325312195591 0.5008641731358665
0.3894706984795064 0.7778325665766488 0.5008641716308375
0.8894708248717649 0.2778325889891207 0.5008641685924928
0.8894706910751609 0.7778325728305475 0.5008641632480320

The optimized structure coordination of NLi_4 decorated g-CN monolayer with 36 adsorbed hydrogen molecules:

1.00000000000000
14.2329998016000001 0.0000000000000000 0.0000000000000000
-7.1164999008000001 12.3261394003000007 0.0000000000000000
0.0000000000000000 0.0000000000000000 35.0000000000000000
C N Li H
24 28 16 72
Direct

0.2130840283236869	0.2612783452726728	0.5031658773518349
0.2126467238033266	0.7607435307093184	0.4907121709830309
0.7127696738241056	0.2607982761977834	0.4906292320444902
0.7133877640555996	0.7615706450878532	0.5037043040427523
0.2718905164436049	0.2012707072390189	0.5030573458129781
0.2725345514750607	0.7019474547352575	0.4909045196560534
0.7727345040718865	0.2021593389587933	0.4901522014797667
0.7720876450279924	0.7017141849768299	0.5036095512779784
0.2092549993894185	0.4203425238575929	0.4984451877370947
0.2077302095290377	0.9193287731846557	0.4953575189323023
0.7077327539834188	0.4191777815425290	0.4964395276298517
0.7094364291567028	0.9203359332511376	0.4977142397610964
0.2671850856213867	0.0383491214032719	0.4963958866531968
0.2687560686087763	0.5394857322240888	0.4978711024872889
0.7690625501110506	0.0394859361292846	0.4964700959538602
0.7671932793263980	0.5384498369248016	0.4974444569980957
0.0507564017835302	0.2576368609307565	0.4961145043192154
0.0495317277589170	0.7561322874846974	0.4967869574667680
0.5496929990123367	0.2558249429779293	0.4975218613007102
0.5506111910477567	0.7577370119644082	0.4975298668427494
0.4304944907756141	0.1963259549699709	0.4983387329694510
0.4313908198056390	0.6981531277670757	0.4967287142998303
0.9316694462326822	0.1981582297406695	0.4950016639581858
0.9304769346637982	0.6967968164380626	0.4979648898338036
0.2683779333937351	0.3709991464272325	0.5049361581186024
0.2675389732613650	0.8703653035895745	0.4896015621887090
0.7673901146326545	0.3705705214747373	0.4899598708564955
0.7689395448513539	0.8714920853248179	0.5040070547519238
0.2113915390848280	0.0912082326263652	0.5003618535722129
0.2128788961616810	0.5918421459602948	0.4930897240025913
0.7131044626534342	0.0919328171891279	0.4922930124190755
0.7116757312140625	0.5912235891259480	0.5020129597738473
0.1009804985626682	0.3676030582448503	0.4925447778470788
0.0991640965233485	0.8661375996339556	0.5001736749002923
0.5992538079979693	0.3656558125972321	0.5016886625292111
0.6006771199119725	0.8673779893139488	0.4927101068826037
0.3772288313621158	0.0879332698449827	0.4930290100197740
0.3784446912195328	0.5897054036325245	0.5024306963073780
0.8789829320361426	0.0895832952561943	0.5002190272865467
0.8769904553254984	0.5880333283878668	0.4932131697477007
0.1031263151667363	0.2015420873126642	0.5003664221426659
0.1024079368884938	0.7003068628703903	0.4928900422778168
0.6024890874589991	0.2003002921441830	0.4929056530832922
0.6029865200335532	0.7019803309680686	0.5023915762949899

0.3814877393464053	0.2558922059742768	0.5044493268572131
0.3823898906697785	0.7573536257151627	0.4899394624197966
0.8825717050500348	0.2575988033548793	0.4888892798117909
0.8818172125369242	0.7566282864564826	0.5039262180041092
0.4986393384774723	0.4848670175724967	0.5677632903587064
0.0006508411917807	0.9883758810578651	0.5654897343943659
0.4986881355708488	0.9886305336954790	0.4274336995031073
0.9984036628067733	0.4899793978628773	0.4276807080956093
0.4907743564728265	0.3744043107975740	0.5366502323264045
0.5691367414527222	0.5589422014708099	0.5213764850101937
0.3855052603667817	0.4812437239973927	0.5391615345870187
0.5296746658123970	0.4935770386924770	0.6175461903875744
0.9927883030558766	0.8748155516189284	0.5364199490175641
0.0641930109981255	0.0544275545051289	0.5166365898948837
0.8858372558256453	0.9838291458806485	0.5377671674078086
0.0377782039701494	0.0013431483907830	0.6148897156144718
0.3861713508259722	0.9806289296033779	0.4572451789190442
0.5674453241725246	0.0548458268295105	0.4756570115987495
0.4933979734689700	0.8745144724979728	0.4560674555519257
0.5125912661574747	0.0207892979429388	0.3775113162292582
0.8855427940468474	0.4813404692678843	0.4577564150762424
0.0669401613748102	0.5543577201435290	0.4761419955152030
0.9942338654579784	0.3749354740663992	0.4553565965173397
0.0113156552096828	0.5321768244825507	0.3787750271616960
0.5567823671236625	0.5110034748964989	0.7180026816122453
0.5247187037123251	0.4501980263194594	0.7170630365139723
0.5063133332576560	0.3113465722120376	0.5846044708042326
0.5058073334567614	0.2614329359729094	0.5765292313624094
0.2794794073134546	0.4964782770099690	0.5785053910260113
0.5158708187897522	0.6709576503644976	0.5774185282797255
0.5151316854828379	0.7226741740395120	0.5824802011731226
0.3283692241516796	0.4972189315835410	0.5875938947619735
0.7403888797010684	0.5067647681181137	0.5799679176911743
0.3192521877514151	0.3090497447982805	0.5791108433772623
0.2653991871632673	0.2565343722761622	0.5820003980806078
0.6416726804981318	0.4607329224568394	0.6341877005045405
0.6170936734283468	0.4586124009049900	0.6535459992072417
0.3946249337719447	0.3718947503126473	0.6319831595933163
0.4138006858586108	0.3819638382033363	0.6526998027649832
0.5093927088337468	0.6108452665219904	0.6340270171426537
0.5173483129593814	0.5955241259575900	0.6540701815540482
0.6876519916390316	0.5060790348042697	0.5752046749824609
0.0623037356226178	0.0137192997637449	0.7164836105596396
0.0292952682588185	0.9530007373836030	0.7152731184734342

0.0115619215402920	0.8146916327611394	0.5838758148931603
0.0091790699064192	0.7642484263126779	0.5753958602081324
0.7823062395023508	0.0027357759266516	0.5768611379872106
0.0178778589266888	0.1745094370750819	0.5747047281070303
0.0186340018879612	0.2272156162639404	0.5795023696467527
0.8322425168729369	0.0054500336388587	0.5858939307509740
0.2426213864106336	0.0131851924853470	0.5780713009732468
0.8273896132947897	0.8186013821437965	0.5781589212258954
0.7735513854724703	0.7661317525598611	0.5814643122477621
0.1488738574034633	0.9682949202088106	0.6327654976653960
0.1234750563752050	0.9667494939342540	0.6518925136595439
0.9024241234612276	0.8826478746258127	0.6306719909687292
0.9227646682449316	0.8933448797839032	0.6512534346062633
0.0201624426814046	0.1201468729045850	0.6313048046496312
0.0308529973336520	0.1068640316664785	0.6513099211877902
0.1887563800507371	0.0101442354005172	0.5731395183999402
0.5259612703288129	0.0435737176083322	0.2760275122541688
0.4654165662941679	0.0094185934427765	0.2777058811084055
0.3247826015334248	0.9976622232781643	0.4094651967929346
0.2749880132493696	0.9968249126092240	0.4178274364188367
0.5088570843984567	0.7672797025799536	0.4170643069436143
0.6828698498207900	0.0046995841190933	0.4177527859500654
0.7346977021703327	0.0039662231254078	0.4127181396835807
0.5108163449175103	0.8165125517890371	0.4078022575559609
0.5196234007546752	0.2295377090349973	0.4149950191133891
0.3221778221461638	0.8090876916443150	0.4152615652798451
0.2700731338952064	0.7550244548131020	0.4121406960002963
0.4798550762144254	0.1322928539865397	0.3604358579471776
0.4791783065379148	0.1083534183441335	0.3410759592419542
0.3897454502363771	0.8862997078131377	0.3630166646704268
0.4005707597789774	0.9040819540998352	0.3421500102730078
0.6305336065715402	0.0019790141756917	0.3602610941665271
0.6134263748973492	0.0094270304621116	0.3404684485823719
0.5188242101572534	0.1768113434723318	0.4198431222115599
0.0251528689507431	0.5571675325024835	0.2763450726873842
0.9644112309061297	0.5233746010512726	0.2769197027647922
0.8234704799319995	0.4987698688595004	0.4101145954137390
0.7733076587952556	0.4971778546650452	0.4184479348059608
0.0121342239375456	0.2729857460804534	0.4150600346559979
0.1855124799075770	0.5075100709883270	0.4181847428238463
0.2378374878449500	0.5079789308549095	0.4131184031417204
0.0143896923412307	0.3231579054087099	0.4064583137090191
0.0252209757488641	0.7301399865557736	0.4143738653734304
0.8271415407113243	0.3164813606078626	0.4151472265353882

0.7745074934317596	0.2627802933623760	0.4120755168366941
0.9723662176503757	0.6389047096400000	0.3596384485505940
0.9701786231072608	0.6116108613347344	0.3408868276282576
0.8956901476899315	0.3945957640654099	0.3631330242066562
0.9039423954547465	0.4143223243112706	0.3424689004046442
0.1293088127504179	0.5138835525934042	0.3621223508509498
0.1172487162582820	0.5264949426797819	0.3422131110558836
0.0214957936484762	0.6762345176779210	0.4197760636630585