Supplementary information

Enhanced Photocatalytic Mechanism for the Hybrid g-C_3N_4/MoS_2 Nanocomposite

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When g-C_3N_4/MoS_2 nanocomposite is illuminated with light, the photogenerated electrons in g-C_3N_4 layer can easily move to the CB of MoS_2 sheet due to the observed CBO, as shown in Fig. 4. At the same time, the polarized field between C_3N_4 and MoS_2 sheets in the nanocomposite prevents the photogenerated electrons migrating from C_3N_4 to MoS_2 sheet. Clearly, there is a competitive role in electron-hole separation between the band alignment and electric polarized field. To evaluate which one gives the dominative role in migration of the photogenerated electrons, we estimate their electric field strengths. The field strength (E) coming from the band alignment is estimated to be about 2.8×10^{9} V/m ($E=U/d_0$, U is the CBO, d_0 is the separation between g-C_3N_4 and MoS_2 sheets in the nanocomposite, U=0.83 V, $d_0=2.97\times10^{-10}$ m), which is about three times larger than the dipole-induced polarized field strength of 9.8×10^{8} V/m (here, $E=P/\varepsilon_r\varepsilon_0Sd_0$, P is the dipole and P=qd, ε_r is the relative dielectric constant, ε_0 is the permittivity of free space, S is the surface area of the nanocomposite, q=0.06 *e*, $d=2.0\times10^{-10}$ m, $\varepsilon_r=1.0$, $\varepsilon_0=8.85\times10^{-12}$ F/m, $S=7.5\times10^{-19}$ m^2, $d_0=2.97\times10^{-10}$ m). This result indicates that the migration of photogenerated electrons in g-C_3N_4/MoS_2 nanocomposite is dominated by the strong driving force provided by the type II band alignment.

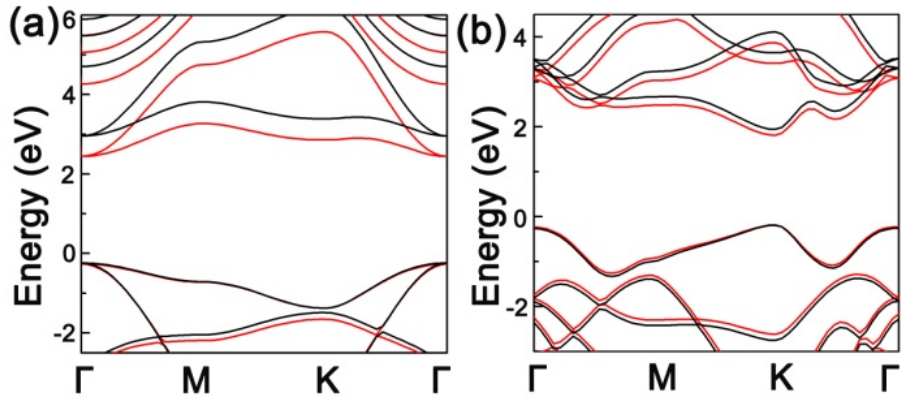

Figure S1 The calculated band structures of isolated g-C_3N_4 (a) and MoS_2 (b) sheets at the HSE06 level. The results of the Hartree-Fock exchange mixing parameter of 0.175 and 0.25 are plotted with the red and black lines, respectively.

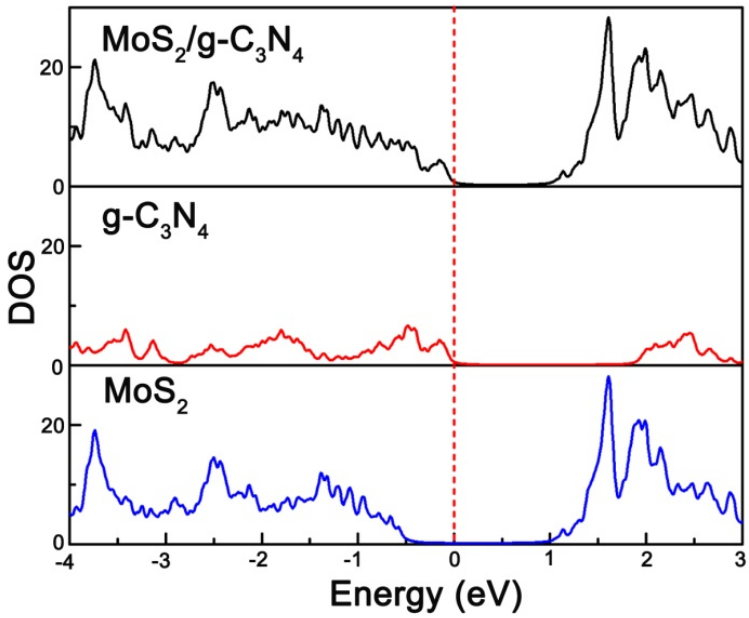

Figure S2 The calculated TDOS and PDOS of g-C_3N_4/MoS_2 nanocomposite at the PBE level.

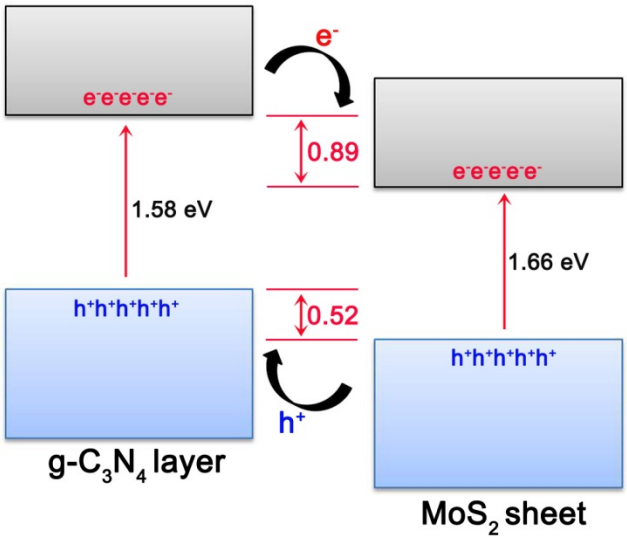

Figure S3 Schematic illustration of the carriers transfer and separation in the proposed g-C_3N_4/MoS_2 nanocomposite at the PBE level.

INCAR input file for structure optimization, energy calculation, band and dos

SYSTEM = Mo S C N
PREC = Normal
ISTART = 0 (1 for band and dos)
ICHARG = 2
ISPIN = 1
ENCUT = 520
NELM = 60; NELMIN= 6; NELMDL= -5
EDIFF = 1E-5
LREAL = Auto
ALGO = Normal
EDIFFG = -1E-2
NSW = 400 (0 for energy calculation, band and dos)
IBRION = 2 (-1 for energy calculation, band and dos)
ISIF = 2
ISYM = 0
POTIM = 0.2
ISMEAR = 0 (-5 for energy calculation and dos)
SIGMA = 0.05
LWAVE = F
LCHARG = F
(LORBIT=11 for dos)
NWRITE = 1
#vdW
LVDW=.TRUE.
#HSE06
LHFCALC=.TRUE.
AEXX=0.175
AGGAX=0.825
PRECFOCK=Fast (Normal for energy calculation, band and dos)
IMIX=1
AMIX=0.2

Optimized geometries by employing PBE functional (9×9×1 k-points)

POSCAR

g-C3N4 monolayer

1.000000

4.790000 0.000000 0.000000

-2.395000 4.148262 0.000000

0.000000 0.000000 15.000000

3 4

Direct

0.353083 0.176542 0.500000

0.823458 0.176542 0.500000

0.823459 0.646917 0.500000

0.000000 0.000000 0.500000

0.503635 0.496365 0.500000

0.503634 0.007269 0.500000

0.992731 0.496366 0.500000

Optimized geometries by employing PBE functional (13×13×1 k-points)

MoS2 sheet

1.000000

3.190000 0.000000 0.000000

-1.595000 2.762621 0.000000

0.000000 0.000000 15.000000

1 2

Direct

0.666666 0.333334 0.501417

0.333333 0.666667 0.397078

0.333333 0.666667 0.605756

Optimized geometries by employing HSE06 hybrid functional (4×4×1 k-points)

POSCAR

g-C3N4 monolayer

1.000000

4.790000 0.000000 0.000000

-2.395000 4.148262 0.000000

0.000000 0.000000 15.000000

3 4

Direct

0.353428 0.176714 0.500000

0.823286 0.176714 0.500000

0.823286 0.646572 0.500000

0.000000 0.000000 0.500000

0.504561 0.495440 0.500000

0.504559 0.009119 0.500000

0.990881 0.495441 0.500000

Optimized geometries by employing HSE06 hybrid functional (6×6×1 k-points)

POSCAR

MoS2 sheet

1.000000

3.190000 0.000000 0.000000

-1.595000 2.762621 0.000000

0.000000 0.000000 15.000000

1 2

Direct

0.666666 0.333334 0.501417

0.333333 0.666667 0.398339

0.333333 0.666667 0.604495

Geometry optimization by employing PBE functional (2×2×1 k-points)
g-C3N4/MoS2 nanocomposite
POSCAR
Mo S C N
1.000000
9.570000 0.000000 0.000000
-4.785000 8.287863 0.000000
0.000000 0.000000 20.000000
9 18 12 16
Direct
0.222180 0.111088 0.336690
0.555504 0.110998 0.337306
0.888912 0.111089 0.336691
0.222232 0.444446 0.337068
0.555504 0.444497 0.337307
0.889003 0.444497 0.337307
0.222227 0.777774 0.337066
0.555555 0.777769 0.337068
0.888913 0.777821 0.336690
0.111237 0.222470 0.258553
0.111021 0.222036 0.414492
0.444970 0.222482 0.259020
0.444127 0.222060 0.415307
0.777519 0.222482 0.259020
0.777938 0.222063 0.415308
0.110951 0.555472 0.259041
0.111443 0.555711 0.415452
0.444531 0.555470 0.259041
0.444285 0.555716 0.415454
0.777519 0.555031 0.259020
0.777941 0.555875 0.415307
0.111235 0.888766 0.258551
0.111022 0.888979 0.414490
0.444528 0.889050 0.259040
0.444290 0.888558 0.415452
0.777531 0.888764 0.258553
0.777965 0.888980 0.414492
0.179565 0.089788 0.564100
0.410645 0.087756 0.587343
0.410642 0.322901 0.587344
0.176039 0.588021 0.597070
0.411976 0.588021 0.597069
0.411980 0.823963 0.597073

0.677098	0.589355	0.587344
0.912244	0.589353	0.587344
0.910210	0.820432	0.564100
0.677098	0.087748	0.587340
0.910216	0.089784	0.564098
0.912253	0.322902	0.587340
0.999997	0.000000	0.560126
0.257999	0.250957	0.565438
0.258006	0.007051	0.565434
0.485652	0.242829	0.606236
0.999867	0.499935	0.591768
0.251212	0.748788	0.598849
0.251211	0.502439	0.598838
0.497564	0.748795	0.598850
0.500062	0.499939	0.591768
0.749040	0.741998	0.565436
0.757170	0.514346	0.606237
0.992947	0.741995	0.565436
0.500064	0.000135	0.591769
0.757170	0.242827	0.606233
0.749036	0.007047	0.565432
0.992950	0.250959	0.565435

Geometry optimization by employing HSE06 hybird functional (single Γ points)
g-C3N4/MoS2 nanocomposite
POSCAR
Mo S C N
1.000000
9.570000 0.000000 0.000000
-4.785000 8.287863 0.000000
0.000000 0.000000 20.000000
9 18 12 16
Direct
0.222205 0.111097 0.336870
0.555489 0.110979 0.337503
0.888899 0.111101 0.336865
0.222250 0.444496 0.337380
0.555489 0.444512 0.337510
0.889023 0.444511 0.337503
0.222255 0.777745 0.337380
0.555504 0.777750 0.337380
0.888904 0.777796 0.336870
0.111361 0.222712 0.260139
0.110804 0.221646 0.413482
0.445069 0.222539 0.260469
0.444028 0.222010 0.414214
0.777467 0.222534 0.260465
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0.111309 0.555651 0.414474
0.444457 0.555543 0.260566
0.444344 0.555657 0.414482
0.777462 0.554932 0.260469
0.777991 0.555972 0.414213
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0.110833 0.889168 0.413485
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0.444349 0.888692 0.414474
0.777288 0.888639 0.260139
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0.410897 0.322889 0.586623
0.176234 0.588116 0.595415
0.411884 0.588111 0.595415
0.411885 0.823767 0.595412

0.677106	0.589109	0.586624
0.911992	0.589098	0.586631
0.910482	0.820962	0.566328
0.677103	0.088006	0.586623
0.910479	0.089532	0.566329
0.911993	0.322900	0.586624
0.999999	0.000008	0.563525
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0.257378	0.007620	0.567254
0.486698	0.243351	0.603761
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0.251992	0.748004	0.596797
0.251984	0.503973	0.596807
0.496014	0.748004	0.596803
0.500090	0.499904	0.590230
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0.756648	0.513308	0.603763
0.992376	0.742620	0.567250
0.500095	0.000189	0.590230
0.756645	0.243356	0.603755
0.750234	0.007625	0.567242
0.992375	0.249771	0.567246