Electronic Supplementary Material (ESI) for Nanoscale.

Hybrid MoS_2 nanosheet-CdSe nanocrystals phototransistors with fast photoresponse

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Contents

1. Absorption and photoluminescence spectra of CdSe NCs for optical bandgap.

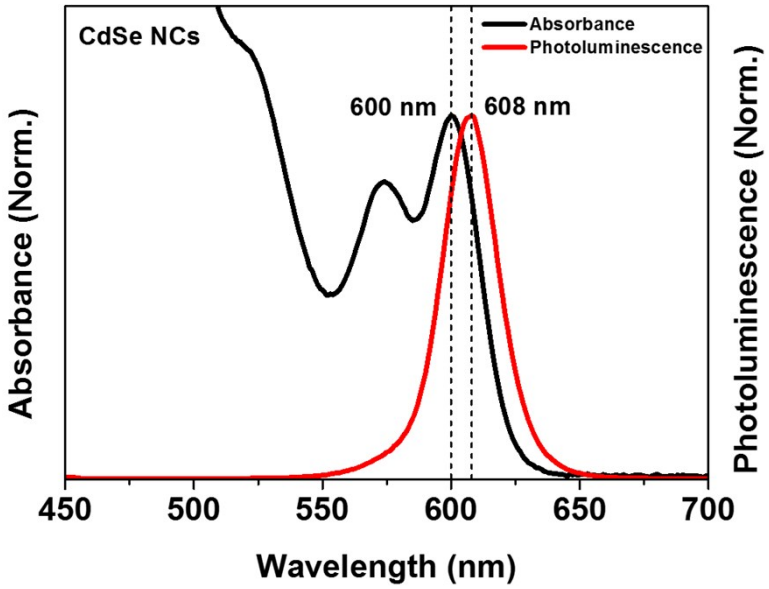

Fig. S1 Absorbance (Norm.) and photoluminescence (Norm.) of CdSe NCs.

2. Transfer curves of pristine MoS_2 and EDT treated MoS_2 FET

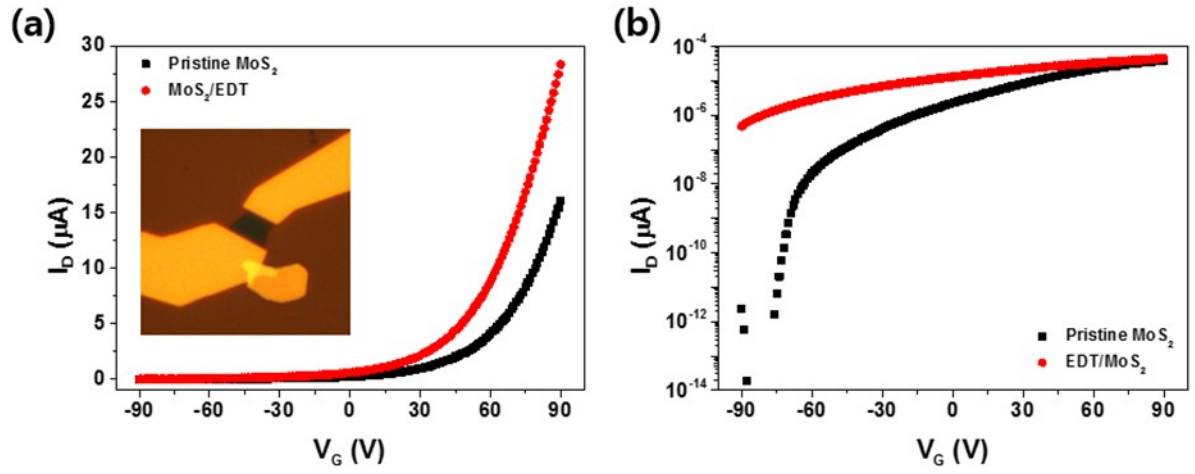

Fig. S2 Transfer curves of pristine MoS_2 and EDT treated MoS_2 FET obtained at V_{DS}=1V, (Channel Length =3 um, Width= 4 um).

3. Work-function calculation.

We performed DFT calculations to determine the VBMs of MoS_2 and CdSe NCs. In the study, the VBM was assumed to be the Fermi level calculated by the DFT calculations because the DFT was originally developed as T = 0 K ground state. The Fermi level was approximately extracted by calculating the work function, which is a surface property of the material. To determine the work functions, we investigated the stable surface of few-layer MoS_2 and CdSe. For the few-layer MoS_2, the (001) plane along the c-axis was considered stable.[1] The stable surface of CdSe is well known to be the (110) plane.[2] In agreement with the experiments, the CdSe (110) surface was the most stable for the (100), (110), and (111) surfaces, as indicated by the lowest surface energy in Table S1. The work functions of the CdSe and few-layer MoS_2 were determined using the local potential of each material, similar to Fig. S2.

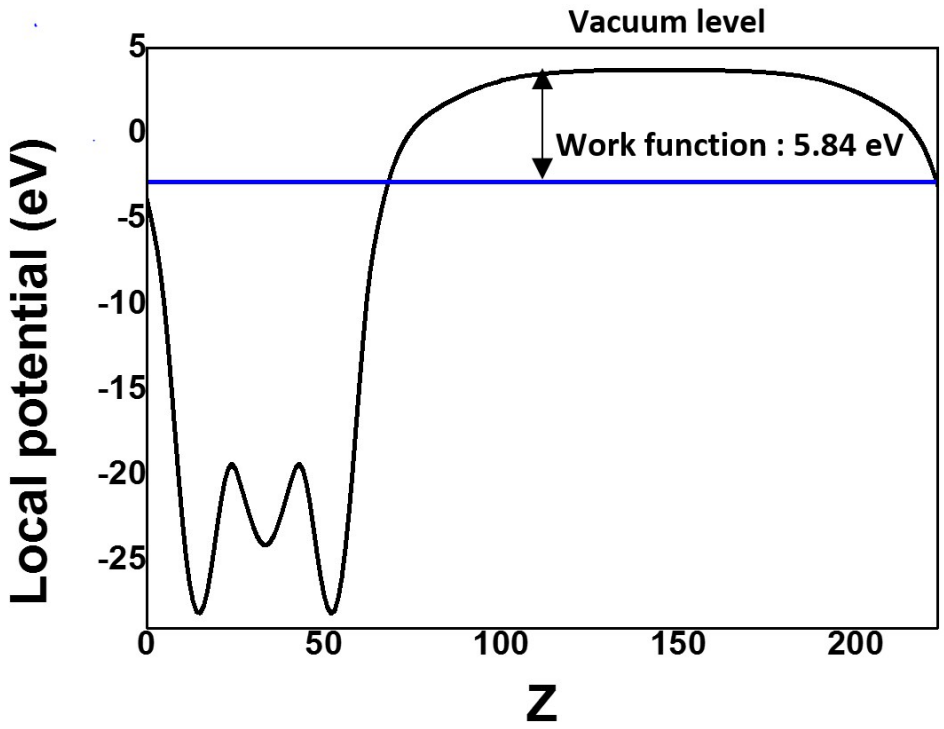

Fig. S3 Work function of monolayer MoS_2 obtained by local potential as a function of the distance along the c-axis of MoS_2.

Table S1. VBM of CdSe with various surfaces and few-layer MoS_2. The surface energy of CdSe is obtained using the following Eq. $\sigma = \frac{E_{surf} - nE_{bulk}}{2A}$, where E_{surf} and E_{bulk} are the energies of the surface and bulk, respectively. A is the area, and n is a unit for compensating for the number of atoms between the surface and bulk.

Materials	CdSe(111)	CdSe(100)	CdSe(110)	MoS_2(1L)	MoS_2(2L)	MoS_2(3L)
VBM (eV)	6.57	6.62	5.57	5.84	5.64	5.54
σ_{cal} (J/m^2)	1.26	0.76	0.38	-	-	-

The electron affinity and bandgap in monolayer MoS_2 have been reported to be 4.0 and 1.82 eV, respectively.[3-5] The VBM of monolayer MoS_2 can be obtained from summation between the electron affinity and bandgap. Based on our assumption that the Fermi level is replaced with the VBM in DFT calculations, the DFT-calculated VBM of monolayer MoS_2 is 5.84 eV, which is in reasonable agreement with the VBM of 5.82 eV derived from electron affinity and bandgap. Thus, the DFT-calculated VBMs of the bilayer MoS_2 and CdSe (110) surfaces are 5.64 and 5.57 eV, respectively.

4. Energy band diagram and calculation methods.

Fig. S3 presents the band diagram for hybridization between the bilayer MoS_2 and CdSe (110) surface. The VBMs (5.64 and 5.57 eV) of both materials (bilayer MoS_2 and CdSe (110), respectively) were obtained using the DFT calculated work function, and the optical bandgaps (1.42 eV and 2.04 eV, respectively) were extracted from PL data of both materials. The conduction levels of each material were determined based on the optical bandgap and VBM in Fig. S3(a). At equilibrium, the difference between each valence level of both materials causes the bending band as an n-n type heterojunction. In Fig. S3(b), we denote the band scheme at the interface between the CdSe NCs and bilayer MoS_2.

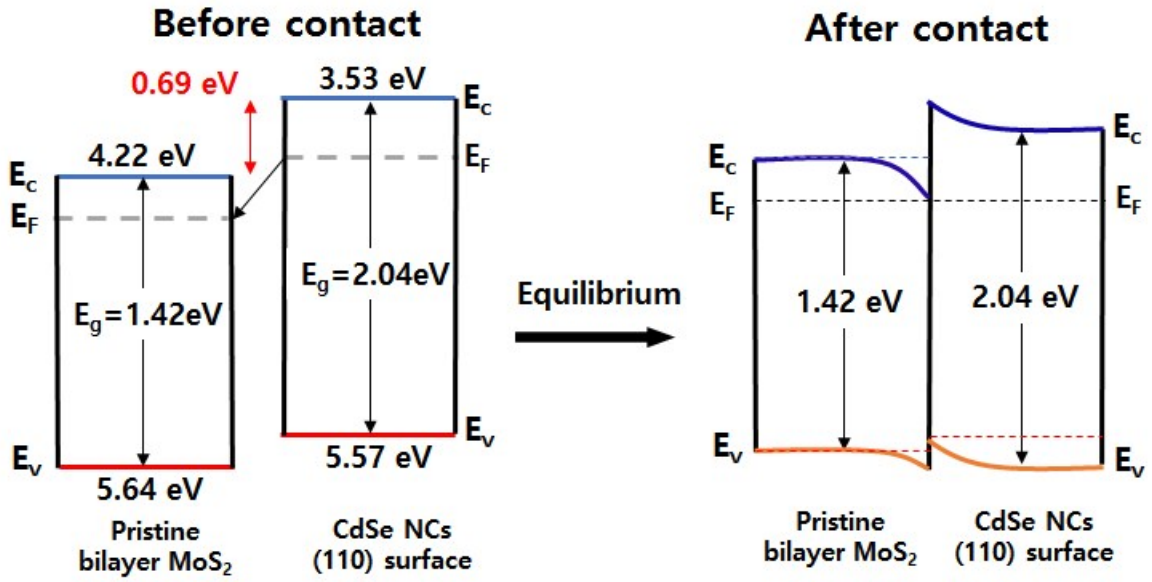

Fig. S4 Band alignment of pristine bilayer MoS_2 and CdSe NCs. Band banding at the interface of both materials through Fermi-level equilibrium mechanism.

DFT calculations were performed using the Vienna Ab-initio Simulation Package (VASP).[6,7] Projector augmented wave (PAW) pseudopotentials as implemented in VASP were used for describing the interactions between ions and electrons, and the exchange-correlation energy of electrons was described using the generalized gradient approximation (GGA) with the PBE functionals. The Kohn–Sham orbitals were expanded on a plane-wave basis set with a cutoff energy of 600 eV. All the atoms were fully relaxed until the Hellmann–Feynman force was less than 0.01 eV/Å. To integrate the Brillouin zone, 9×9×1 and 15×15×1 gamma-point meshes were used for the CdSe surface and MoS_2 with a large vacuum space of 15 Å in the direction perpendicular to the surface.

Notes and references

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