Supporting Information

Metal-Insulator-Semiconductor Diode consisting of Two-dimensional Nanomaterials

Hyun Jeong†,‡, Hye Min Oh†,§, Seungho Bang†,§, Hyeon Jun Jeong†,§, Sung-Jin An†,§, Gang Hee Han†, Hyun Kim†,§, Seok Joon Yun†,§, Ki Kang Kim∥, Jin Cheol Park†,§, Young Hee Lee†,§, Gilles Lerondel‡,§, and Mun Seok Jeong*,†,§

†Center for Integrated Nanostructure Physics (CINAP), Institute for Basic Science (IBS), Sungkyunkwan University, Suwon 440-746, Republic of Korea

‡Laboratoire de Nanotechnologie et d’Instrumentation Optique, Institut Charles Delaunay, CNRS-UMR 6281, Université de Technologie de Troyes, BP 2060, 10010 Troyes, France

§ Department of Energy Science, Sungkyunkwan University, Suwon 440-746, Republic of Korea

∥ Department of Energy and Materials Engineering, Dongguk University, Seoul 100-715, Republic of Korea

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S1. Optical microscopy image of MIS diode with low magnification

Optical microscopy was used for confirming the dimension of the 1-L MoS$_2$ flakes. Figure S2 is the optical microscopy image with low magnification for observing the many of 1-L MoS$_2$ flakes. The scale bar indicates 200 µm. In the optical microscopy image, slightly bright region is 1-L MoS$_2$ flakes. Average size of the flake is around 200 µm. The shape of the flake is triangular. However, fluctuation of the size and shape of the 1-L MoS$_2$ flake is significantly high. With a MoS$_2$ continuous layer, improved device performance of the MIS diode is in terms of stability is expected.

![Optical microscopy image of 1-L MoS$_2$ top layer of the MIS diode with low magnification.](image)

**Figure S1.** Optical microscopy image of 1-L MoS$_2$ top layer of the MIS diode with low magnification.
S2. AFM image of 1-L MoS$_2$

To confirm the crystalline structure of 1-L MoS$_2$ used in this study, atomic force microscopy (AFM) was used to measure the topography of 1-L MoS$_2$. 1-L MoS$_2$ was transferred onto SiO$_2$ flat substrate to verify the surface defect of 1-L MoS$_2$. Figure S2 shows AFM image of 1-L MoS$_2$ and the inset is the height profile for white line marked in the image. The height of 1-L MoS$_2$ is approximately 1 nm and the surface of 1-L MoS$_2$ shows significantly smooth. The AFM image supports that the 1-L MoS$_2$ used in this study has good crystalline quality for application of optoelectronic device.

![AFM image of 1-L MoS$_2$ on the SiO$_2$ substrate.](image)

**Figure S2.** AFM image of 1-L MoS$_2$ on the SiO$_2$ substrate.
S3. AFM images of graphene

It is known that the transferred CVD graphene have lots of polymer residues. In this study, we have employed thermal treatment process to eliminate the polymer residues on the graphene. After rinsing the graphene by using acetone, the graphene was thermally treated at 350 °C for 4 hours in H₂/Ar ambient gas. To verify the surface morphology of graphene, we measured topography of graphene before and after thermal treatment. Figure S3a and b show AFM topographies of graphene before and after thermal treatment, respectively. Scale bar indicates 5 µm for both AFM images. White dots presented in the Figure S3a are polymer residues. Before thermal treatment, a lot of polymer residues were observed on the graphene. However, almost polymer residues on the graphene disappeared after thermal treatment as shown in Figure S3b. Consequently, we believe that influence of polymer residues on device performance of MIS diode is almost negligible.

Figure S3. AFM images of graphene (a) before and (b) after thermal treatment.
S4. Optical characterization of MIS heterojunction structure

Spectroscopic verifications of the vertically stacked layered materials were performed using a confocal scanning Raman spectroscopy. A 532 nm laser was used as the excitation source. Figure S4a presents the Raman spectrum of the 1-L MoS$_2$ top layer of the MIS structure with the main peaks of the layered materials indicated by black arrows. The peaks at 394 and 411 cm$^{-1}$ correspond to the Raman active E$_{2g}$ and A$_{1g}$ modes of MoS$_2$, respectively. The interval between those two peaks (17 cm$^{-1}$) indicates that the MoS$_2$ was a monolayer.$^{39}$ A strong optical first-order Raman peak and a broad multi-phonon scattering peak from Si are observed at 520 and ~970 cm$^{-1}$, respectively. This implies that the total thickness of the MIS structure (< 30 nm) was much smaller than the penetration depth of the laser. The peaks at 1345, 1597, and 2694 cm$^{-1}$ correspond to the Raman active D, G, and G$'$ modes of graphene. The high background signal revealed at larger wavenumber is attributed to PL emission from the B exciton of 1-L MoS$_2$. Note that the h-BN Raman peak (1366 cm$^{-1}$) considerably overlaps with that of graphene (1345 cm$^{-1}$, D mode). Figure S4b and c present a confocal Raman image of a 30 × 30 µm area and a point spectrum for 1-L MoS$_2$, respectively. The region scanned by the Raman microscope was near the edge of the 1-L MoS$_2$ top layer and was the same for all materials. In the 2-dimensional Raman scattering image, extracted from Raman peaks at 394 and 411 cm$^{-1}$, the 1-L MoS$_2$ region is distinguishable, as shown in Figure S4b. A confocal Raman image and spectrum are shown for h-BN in Figure S4d and e, respectively. Although the h-BN Raman peak partially overlaps with the D mode peak of graphene, we were able to extract a confocal Raman image for just the h-BN layer using spectroscopic image filtering, as shown in Figure S4d. Figure S4f and g present a confocal Raman map and spectrum for graphene, respectively. As shown in Figure S4d and f, h-
BN and graphene were distributed over the entire scanning area. From the 2-dimensional Raman scattering image of the MIS structure, we found that the layered materials were well vertically stacked.

**Figure S4.** (a) Local Raman spectrum measured from the 1-L MoS$_2$ top layer of the MIS diode. Raman modes for the 1-L MoS$_2$, h-BN, graphene, and Si layers are simultaneously present. Confocal Raman maps, with 5 µm scale bars, for (b) 1-L MoS$_2$, (d) h-BN, and (f) graphene. A
fixed scanning area, corresponding to the border of the 1-L MoS$_2$ region, was used for all Raman maps. Local Raman spectra measured from (c) 1-L MoS$_2$, (e) h-BN, and (g) graphene regions.
S5. I-V curve of MS contact

Figure S5 shows the I-V curve of a metal-semiconductor (MS) diode structure consisting of graphene and MoS$_2$. The inset presents a 3D schematic of the MS diode I-V measurement setup and I-V characteristic in log scale. We observed almost linear I-V curve from MS diode. It is attributed to direct current flow passed through 1-L MoS$_2$ and considerably low Schottky barrier between graphene and 1-L MoS$_2$. Consequently, we can conclude that the MIS diode consisting of graphene, 1-L MoS$_2$ and h-BN had better current rectifying characteristic than the MS diode consisting of graphene and 1-L MoS$_2$.

![I-V curve of MS contact](image)

**Figure S5.** (b) I-V curve for the MS diode consisting of graphene and 1-L MoS$_2$. The top-left inset is schematic of the device structure. The bottom-right inset is I-V characteristic in log scale. There is no current rectifying behaviour in the MS diode.

To examine the device performance of the MIS diode in detail, leakage currents between the graphene and h-BN layers, and the h-BN and MoS_2 layers were measured. Figure S6a shows the I-V curve measured between the graphene and h-BN layers. The inset presents a schematic of the probing configuration. An almost constant, and remarkably low, current of \( \sim10^{-10} \) A was observed in both bias directions. This current value is \( 10^3 \) times smaller than the current value of the MIS diode. Figure S6b presents the I-V curve measured between the h-BN and MoS_2. The inset presents a schematic of the probing configuration. The measured current was also extremely low, \( \sim10^{-10} \) A, in both bias directions. The I-V curve results indicate that charge transport across the MIS diode was mainly due to carrier tunneling and that there was negligible leakage current between the graphene and h-BN layers, nor between the h-BN and MoS_2 layers.

Figure S6. I-V curves measured between (c) graphene and h-BN and (d) h-BN and MoS_2. Almost no leakage current was observed between layers, except between graphene and MoS_2.
S7. Light emission spectrum in the visible light range for the photocurrent

For analysis of the photocurrent, we have measured light emission spectrum of the lamp as a light source. Figure S7 is the light emission spectrum of the lamp employed for photocurrent. The wavelength range of the light is from 500 nm to 700 nm with central peaks of around 600 nm and 640 nm as revealed in the Figure S7. Since energy band gap of 1-L MoS$_2$ is ~1.8 eV (~688 nm), this light source is proper for photocurrent of the MIS diode.

Figure S7. Light emission spectrum of the lamp used for photocurrent.