**Supporting Information**

# CVD Growth of Large-Area InS Atomic Layers and Device Applications

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Keywords: Indium Sulfide, InS, CVD, 2D, FET

## Methods

### *Synthesis of InS Layers*

2D InS atomic layers were synthesized onto mica substrates ([$KMg_3(AlSi_3O_{10})F_2$], 1 $cm^2$) by the CVD process. Precursors of In and S are $In_2O_3$ powder (Alfa Aesar, 99.99% purity) and S powder (Sigma-Aldrich, 99.5% purity), respectively. The 0.3 g of $In_2O_3$ powder was put in a ceramic bowl set in the heating zone at the center of the main furnace. On the upstream side, the S powder was heated by an additional furnace. The mica substrates were put on the downstream side where the S and $In_2O_3$ vapors were brought to the mica substrates through a mixture of $H_2$ (20 sccm) in an Ar (60 sccm) flowing gas. Meanwhile, the chamber pressure was set to 100–300 Torr throughout the process. The center of main furnace was heated to 650–690 °C at a ramp rate of 25°C/min. The temperature of the upstream furnace was increased to 140°C to melt S. Meanwhile, the main furnace temperature reached 650–690°C. Both the heating furnaces were kept running for 10 min, followed by natural cooling to room temperature.

### *InS Transfer Process*

The 2D InS atomic layers were transferred from mica to arbitrary substrates or TEM grids using a PMMA-mediated transfer technique. Firstly, we spin-coated the PMMA (A4, 950 PMMA, MicroChem Corp.) onto InS samples to form a thin film. Secondly, the samples were immersed in distilled water to separate the InS/Mica interface. The PMMA-supported InS was lifted from the distilled water and then the sample was transferred onto the target substrates. Finally, the

PMMA was taken away by acetone (CMOS electronic grade, J.T. Baker) at room temperature for 1 h, and the sample was followed up by rinsing with isopropyl alcohol (CMOS electronic grade, J.T. Baker) and distilled water to finish the transfer process.

*EDLT Device Fabrication*:

The single-crystal InS flakes were transferred from mica to the $SiO_2$/Si substrate using the PMMA-mediated transfer method. The source and drain electrodes were thermally deposited at 80 nm Au onto the InS surface. And then, the ion gels were formed using a mixture of an ABA triblock copolymers of poly(styrene-block-methyl methacrylate-block-styrene) (PS-PMMA-PS; $M_{PS}$ = 5.0, $M_{PMMA}$ = 13.0, $M_w$ = 23.0 kg/mol), and ionic liquids of N, N-diethyl-N-methyl-N-(2-methoxyethyl) ammonium bis(trifluoromethanesulfonyl)imide ([DEME][TFSI]) in an ethyl propionate solvent, which served as a gate dielectric material. The weight ratio of the ion gel solutions was kept at 0.7:9.3:20 for the polymer, ionic liquid, and solvent, respectively. Finally, a 50-μm Pt foil was placed on the drop-casted ion-gel films as a top gate electrode, follwed by anealing the device by 70 ˚C to remove the solvents.

*Characterizations*: Optical images were taken with an Olympus BX53 microscope. Surface morphologies were obtained by atomic force microscope (Bruker, Dimension Icon). Scanning electron microscope (SEM) images were taken with a scanning electron microscope (Hitachi S4800). Micro-Raman spectra were acquired on samples using a confocal Raman microspectroscopy system (Tokyo Instruments, Inc. Nanofinder 30) with a standard 488 nm laser excitation (laser power: 0.1 mW; spot size: ~1 μm). The spectra were calibrated by Si peak position (520 cm$^{-1}$) from bulk Si substrate. The absorbance spectra of an 1-cm$^2$ sized InS films were collected in an UV-Vis-NIR spectrophotometer (Hitachi U4100) in the wavelength range of 300–1000 nm. Chemical configurations were determined by X-ray photoelectron spectroscope (ULVAC-PHI Phi V5000), performed with an Al Kα X-ray source on the samples. The energy calibrations were made against the C 1s peak to eliminate the charging of the sample during analysis**.** The crystal structure was characterized by grazing incident X-ray diffraction (GIXRD, Bruker D8) of Cu-Kα radiation operating at 40 kV and 40 mA. The XRD patterns were obtained with a glazing incident angle of 1 degree. Quantitative elemental analysis was performed using an EDX detector (Oxford Instruments, X-Max 80 TLE) equipped at the TEM. The InS flakes were transferred onto TEM copper grids with alacey carbon support film. HRTEM and HAADF STEM images were obtained by an aberration-corrected TEM (JEOL JEM-ARM200FTH; FEI Titan Chemi-STEM, 80 and 200 kV). The sample for TEM cross-section study was prepared by a focused ion beam (FIB, FEI Helios NanoLab G3 CX). Before cutting, the sample was capped by an ink and a platinum layer as a passivation layer to prevent the damage from Ga ions during cutting. All electrical characterizations were performed with a semiconductor device analyzer (Agilent Technologies, Inc. E5270), and the device was set on a shield probe station inside a nitrogen-filled glove box at room temperature (27 ℃). Impedance measurements were performed to evaluate the capacitance and the mobility by a frequency response analyzer (a Solartron 1252A frequency response analyzer with a Solartron 1296 dielectric interface controlled by ZPlot and ZView software). The frequency range was set from $10^0$ to $10^5$ Hz, while an AC voltage source with amplitude of 5 mV and a DC voltage of 3.2 V was provided.

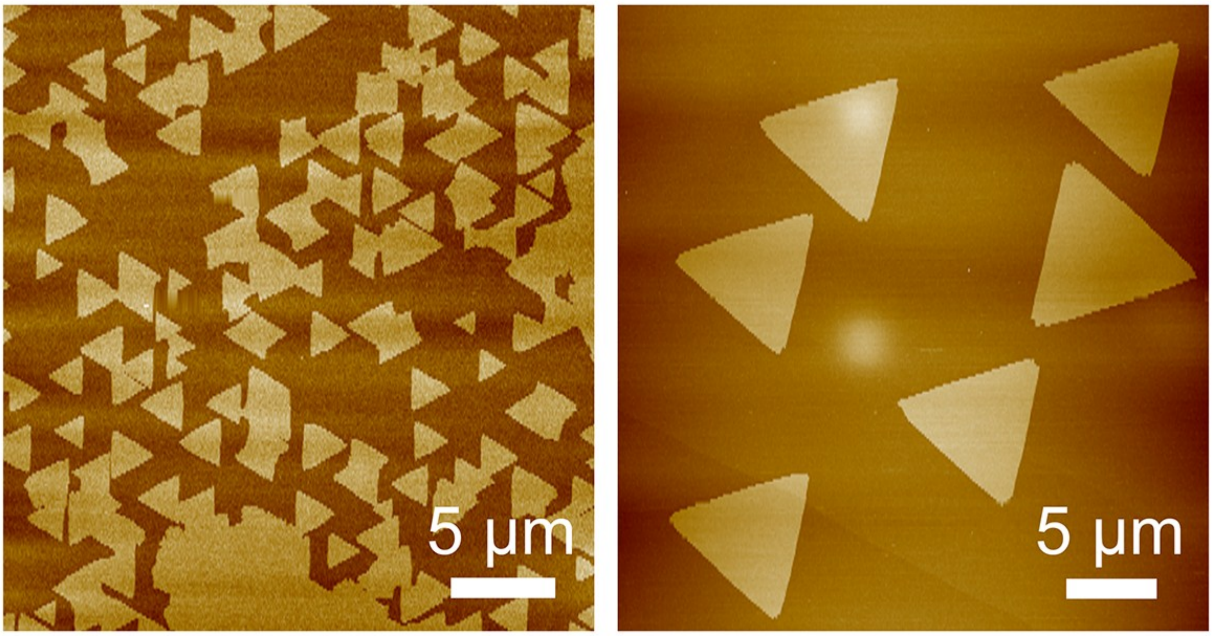


**Figure S1.** AFM image of the orientation of small InS flakes aligned to a certain direction (0° and 60°). Scale bar is 5 μm in length.

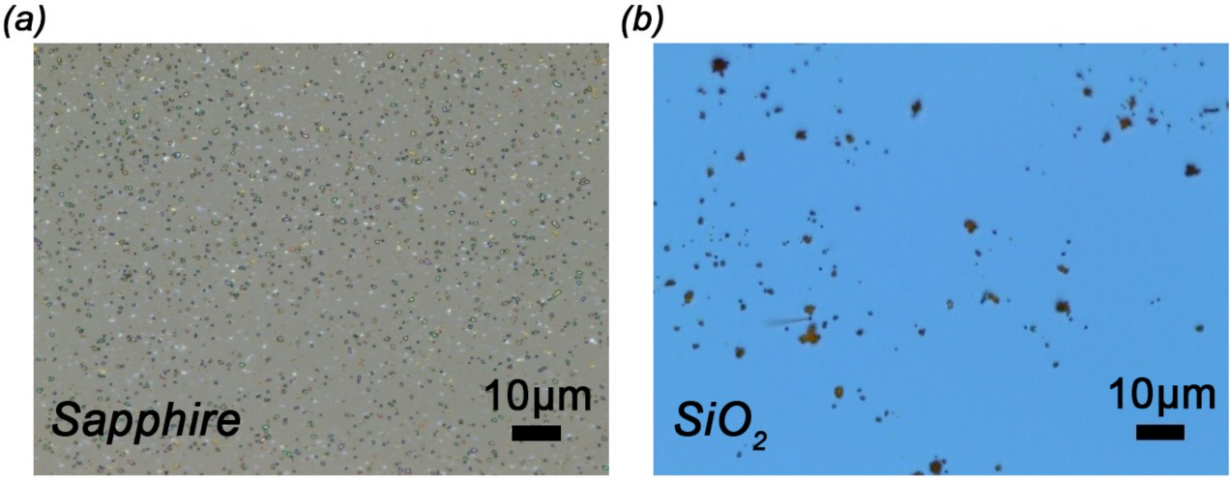


**Figure S2.** The OM images of the directly synthesized InS on (a) sapphire substrate; (b) $SiO_2$/Si substrate.

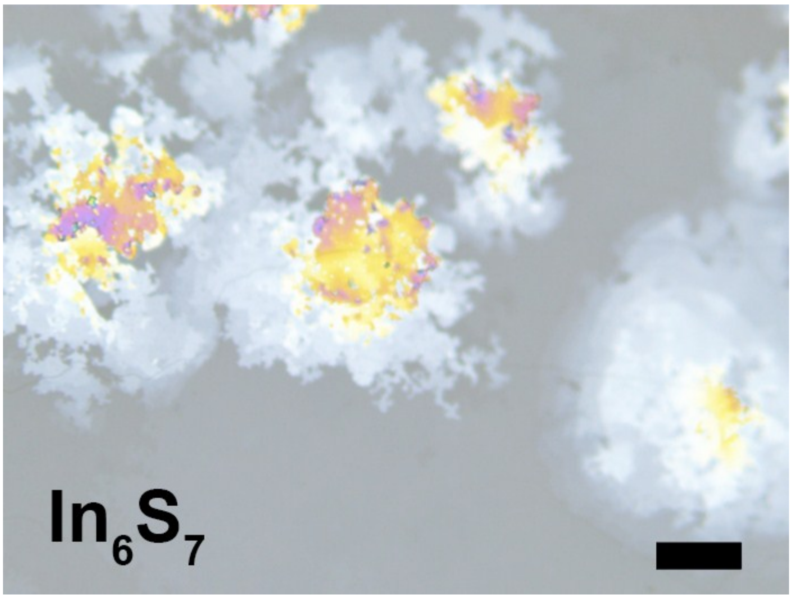


**Figure S3.** OM image of the few-layer $In_6S_7$. Scale bar, 10 μm.

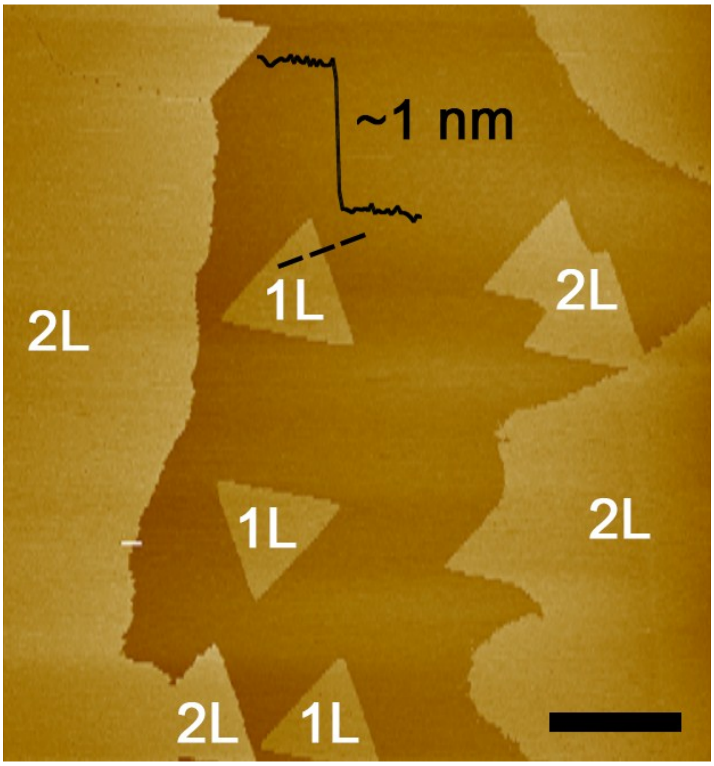


**Figure S4.** AFM image of monolayer InS flakes grown on mica substrate. Scale bar, 2 μm.

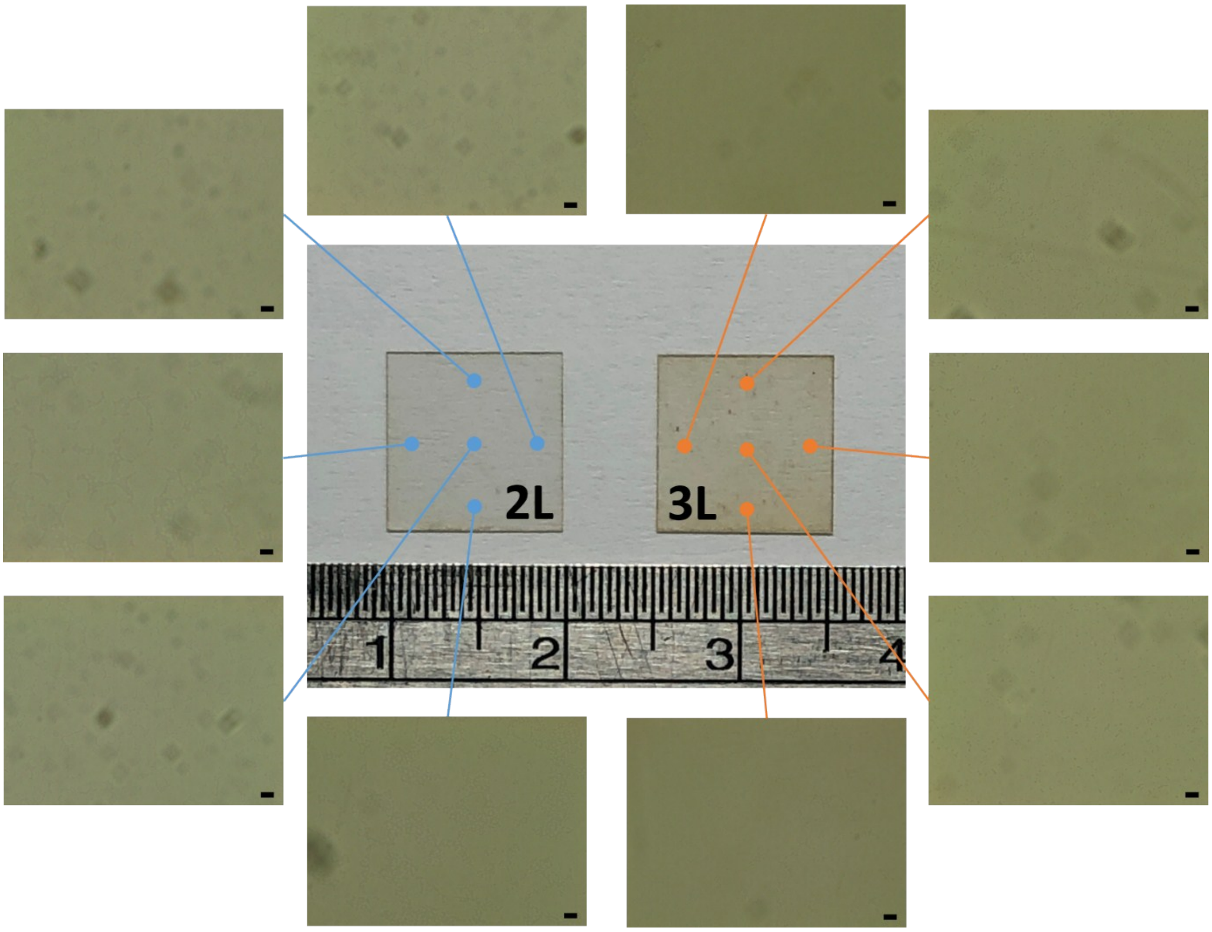
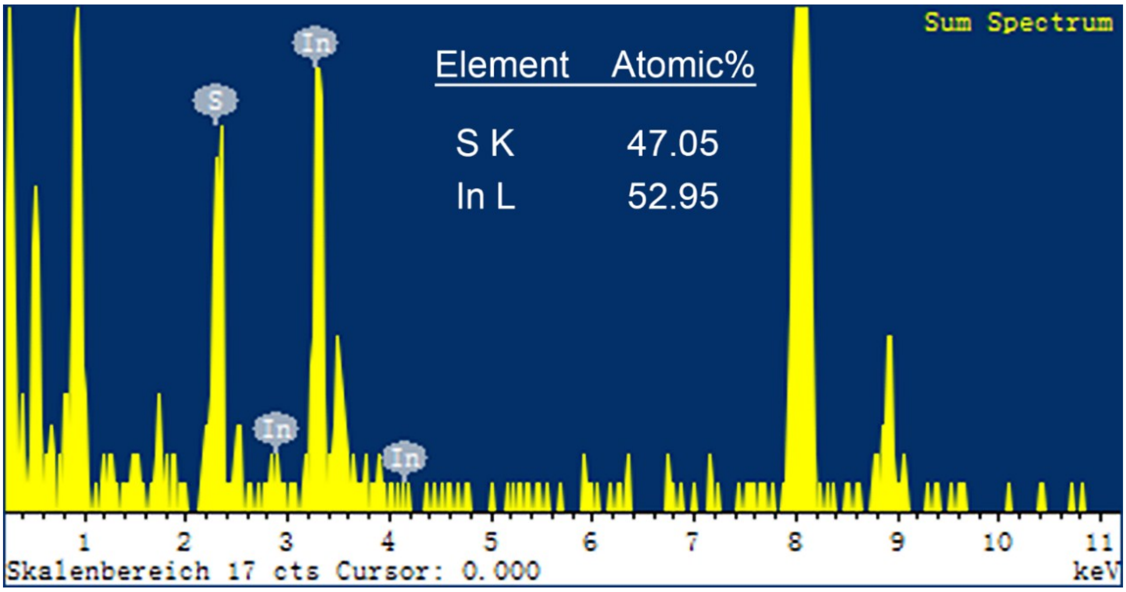


**Figure S5.** OM images of the uniformity of bi- and trilayer InS thin film. Scale bar, 10 μm.



| Element | Atomic% |
|---|---|
| S K | 47.05 |
| In L | 52.95 |

**Figure S6.** EDX spectra of InS synthesized by CVD method. EDX shows a stoichiometric ratio of 1 :1 between In and S.

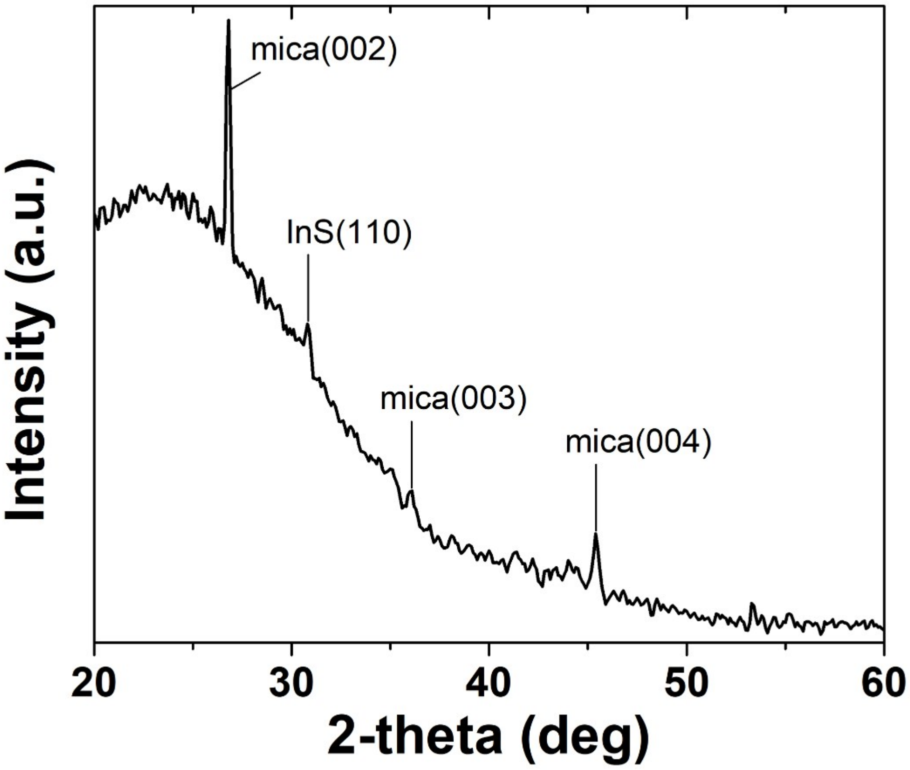


**Figure S7.** GIXRD pattern of the InS thin-film.

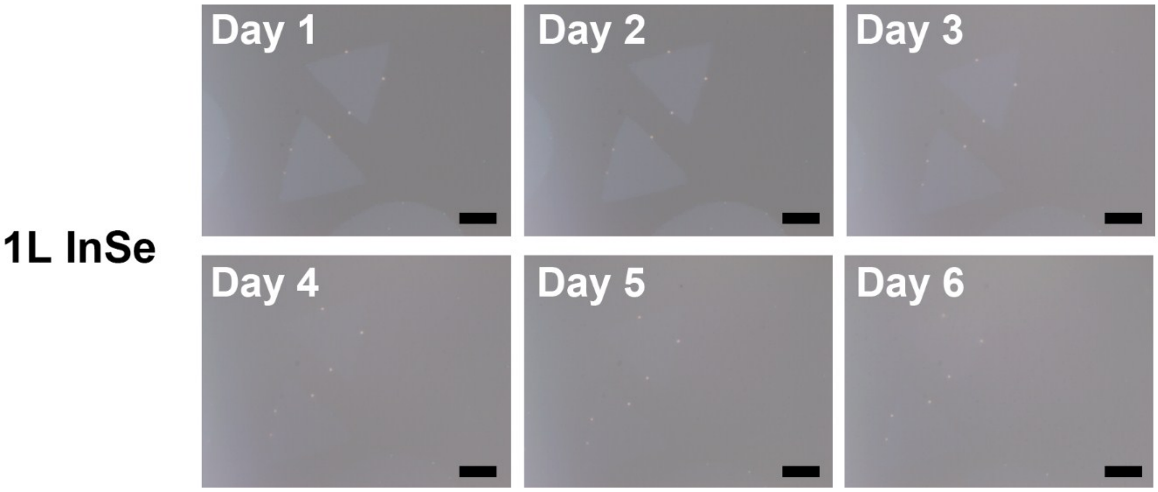


**Figure S8.** Sequence of OM images acquired at different times for monolayer InSe under ambient conditions. (Day n: after exposure to ambient conditions n days). Scale bar is 10 µm in length.

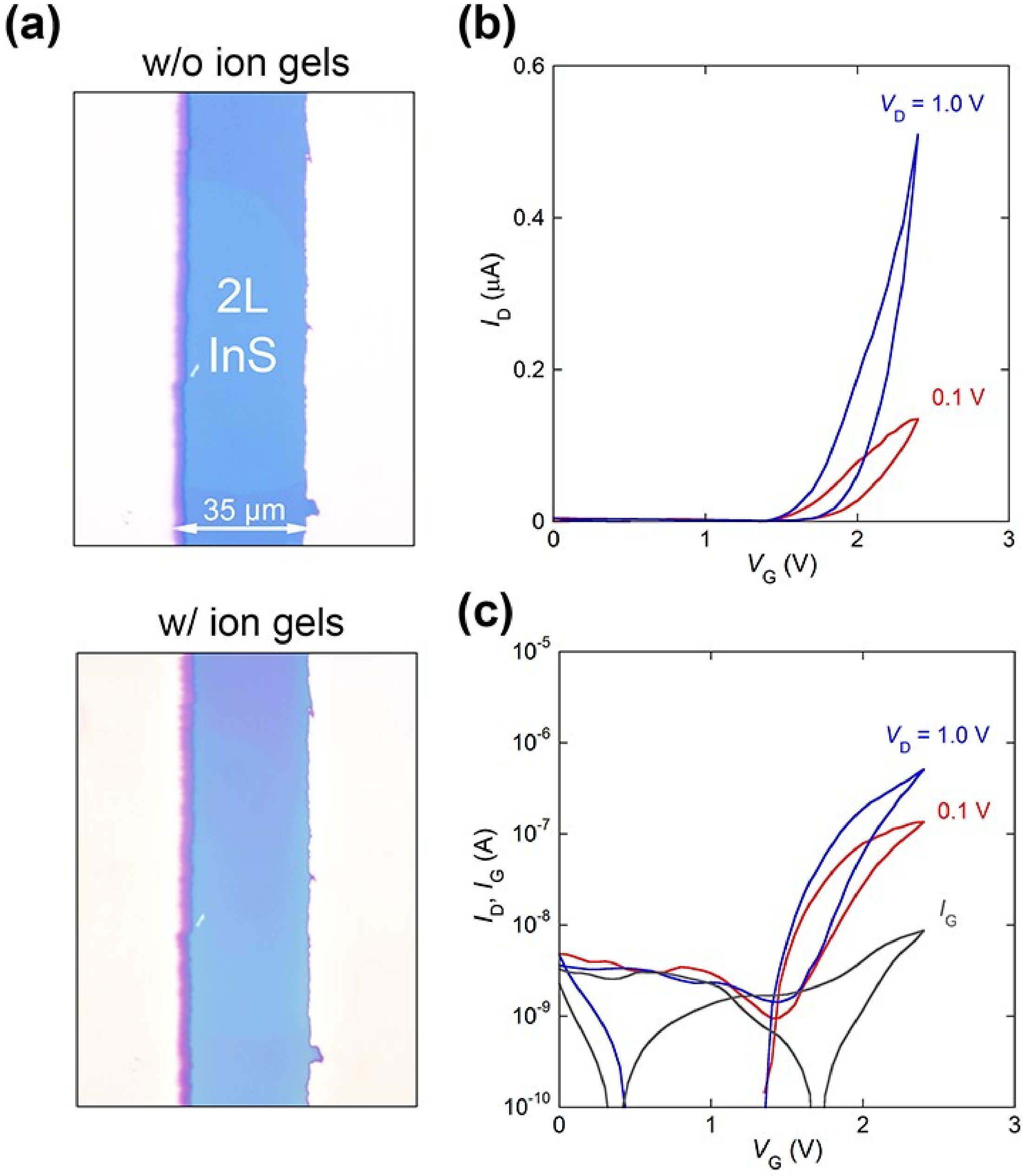


**Figure S9.** (a) Top view OM image of the bilayer InS EDLT device, which were taken before and after the ion gel. (b) Linear scale and (c) Logarithmic-scale transfer curves of the bilayer InS EDLT.