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Locally Gated SnS₂/hBN Thin Film Transistors with a Broadband Photoresponse

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Next-generation flexible and transparent electronics demand newer materials with superior characteristics. Tin dichalcogenides, Sn(S,Se)_{2z}, are layered crystal materials that show promise for implementation in flexible electronics and optoelectronics. They have band gap energies that are dependent on their atomic layer number and selenium content. A variety of studies has focused in particular on tin disulfide (SnS₂) channel transistors with conventional silicon substrates. However, the effort of interchanging the gate dielectric by utilizing high-quality hexagonal boron nitride (hBN) still remains. In this work, the hBN coupled SnS₂ thin film transistors are demonstrated with bottom-gated device configuration. The electrical transport characteristics of the SnS₂ channel transistor present a high current on/off ratio, reaching as high as 10⁵ and a ten-fold enhancement in subthreshold swing compared to a high-κ dielectric covered device. We also demonstrate the spectral photoresponsivity from ultraviolet to infrared in a multi-layered SnS₂ phototransistor. The device architecture is suitable to promote diverse studied on flexible and transparent thin film transistors for further applications.

An emerging new two-dimensional (2D) material is metal tin dichalcogenides, which have a layered structure composed of an earth-abundant compound solid. It is currently being considered as a promising candidate for flexible and heterostructured electronics¹⁻³. Remarkably, the tin-based chalcogenide alloy SnS_{2-x}Se_x shows a broad modification of the band gap with a selenium composition (for example, 2.07 eV and 0.97 eV for SnS₂ and SnSe₂, respectively)⁴⁻⁶, which provides new possibilities for optoelectronics⁷. Only a few studies have investigated the mechanical characteristics of tin dichalcogenides so far. However, the covalently bonded SnS_{2-x}Se_x alloy has shown that its lattice structure has a hexagonal CdI₂-type, analogous to the widely investigated molybdenum disulfide (MoS₂); this led us to expect that the alloy would have a higher strain limit than that of ionic-bonded bulk semiconductors⁸. Moreover, Mitzi *et al.* demonstrated that the soluble semiconducting SnS_{2-x}Se_x can offer solution-processed thin films, making the integration of polymer substrate accessible¹. All of these properties give this type of material great potential to meet the criteria for wearable and flexible devices⁸.

Pan *et al.* investigated an SnS_{2-x}Se_x crystal-based thin film transistor (TFT) under different x-contains, finding that the current on/off ratio was heavily decreased in the selenium-rich channel because of its large electron concentration⁹. This has motivated significant efforts to investigate the SnS₂ crystal for field-effect transistors (FETs)^{10,11} and photodetector applications¹²⁻¹⁴. For instance, an SnS₂ nano-membrane FET with universal back-gated device geometry reported by De *et al.* exhibited a high switching ratio of up to 10⁶ and a poor subthreshold swing (SS)¹¹. Other works have confirmed this finding and further shown that the SS parameter was typically observed in dozens of volts per decade range. The device architecture in the form of a top-gated FET capped by a high-κ Al₂O₃ layer demonstrated similar subthreshold swing values approximately 10 V/decade¹⁰. It is believed that the trapped charges located between 2D and conventional oxide significantly influence the quality of the interface. Because layered solid crystals lack dangling bonds, the materials provide primary advantages in building heterostructures that combine diverse 2D layers into a three-dimension¹⁵. However, no study has yet reported research of transistors encapsulated by a wide band-gap 2D dielectric (5–7 eV)^{16,17} that is a hexagonal boron nitride (hBN) with an integrated high-quality SnS₂ nanosheet.

In this study, we construct a multi-layered SnS₂ channel device incorporating hBN as a gate dielectric. Taking a different approach from other published works, the proposed transistors have a locally gated geometry instead of using universal silicon back-gating. We report a substantial improvement of the SS parameter of the device and characterize the effect of the Schottky-limited metal/semiconductor contact to describe the thermally activated

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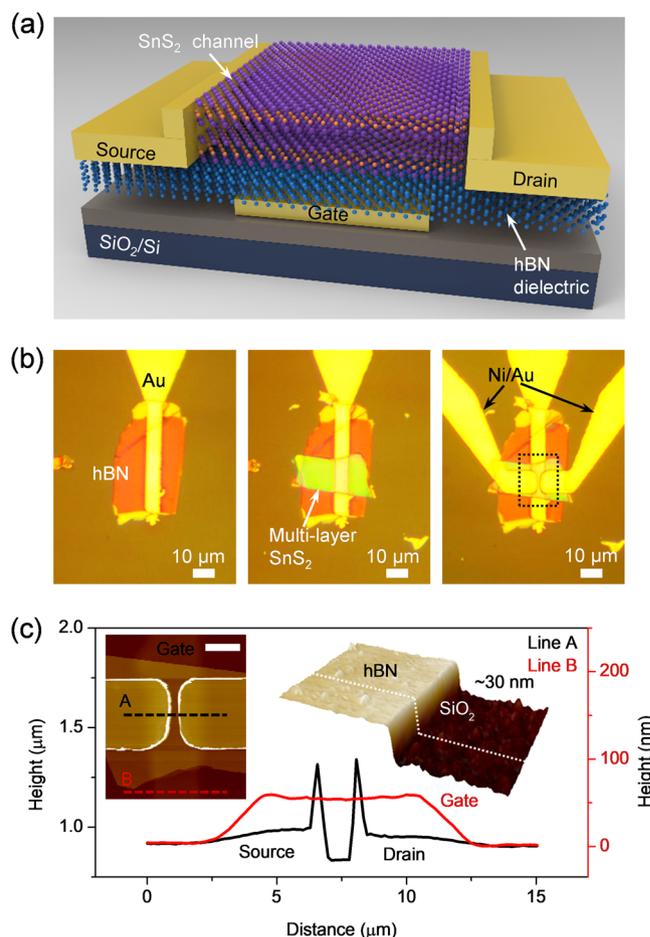


Figure 1. (a) 3D schematic representation of a bottom-gated SnS₂/hBN heterostructure transistor. The high-quality hBN insulator proves to be an ultra-flat surface acting as a substrate for the precisely aligned SnS₂ layer. (b) Optical images of the deposition of the hBN flake on the gold gate (left), the transfer of SnS₂ (thickness typically in 1–30 nm range) onto the top of the hBN (middle), and the defined metal leads for source/drain contact (right). (c) The height profiles for lines A and B acquired from the AFM image (inset, left) of the device. The scale bar is 5 μm. Inset (right): 3D topography for the hBN on the SiO₂, showing about a 30 nm thickness.

transport. Furthermore, for the phototransistor, we also include the photoelectric behavior of the light-exposed SnS₂. It appears that the SnS₂ crystal responds to a wide range of photon spectra.

Results

Figure 1a schematically illustrates the device geometry of the proposed SnS₂/hBN transistors, where atomically flat hBN acted as the gate dielectric¹⁸ and a multi-layered SnS₂ nanosheet was used as the carrier transport layer. Constructing the bottom encapsulation of hBN is beneficial because the SnS₂ layer is far from the underlying potential fluctuation (SiO₂ substrate). Our recent investigation showed that the interface trap sites located at the 2D/SiO₂ interface could represent more than 10¹² states/cm²eV¹⁹. Because contamination should be avoided during the fabrication process, we used a polymer-incorporated Scotch-tape residual-free technique to minimize the use of chemical solvents. In contrast to the wet transfer method described in other reports, this technique has the ability to control the interface trap state D_{IT} down to the 10¹⁰ states/cm²eV range for a suspended 2D channel structure¹⁹. In this work, we fabricated more than five SnS₂ devices with typical S/D dimensions: a channel length/width (L/W) ratio of 0.3 and a gate lead with a width of 5 μm. Optical images of the step-by-step preparation of the hetero-structured device are depicted in Fig. 1b. The as-made devices were subsequently characterized via atomic force microscope (AFM) analysis to quantify the thickness of the hBN dielectric and the SnS₂, as illustrated in the left inset of Fig. 1c. The AFM cross-sectional profiles labeled *line A* (black) and *line B* (red) indicate a clear overlap between the channel area and the local gate, as illustrated in Fig. 1c. For the material characterization, the Raman spectra of the as-exfoliated SnS₂ exhibit two non-degenerated scattering modes, with the out-of-plane A_{1g} mode located at 320.6 cm⁻¹ and the weak in-plane E_g peak located at 213.5 cm⁻¹ (see the Supplementary, Fig. S1) under room temperature (see Fig. 2a, left). The data are in agreement with those of previous works^{20,21}. On the other hand, in-plane mode, E_{2g} of hBN is displayed in Fig. 2a right in consistent with other literature (The full Raman spectra of the heterojunction area can be seen in Fig. 2b)²².

Next, we proceed to examine the SnS₂ channel TFTs and the influence of the hBN on the SS performance. Figure 3a shows the drain current I_{DS} behavior of the devices as a function of the gate voltage V_G under a constant

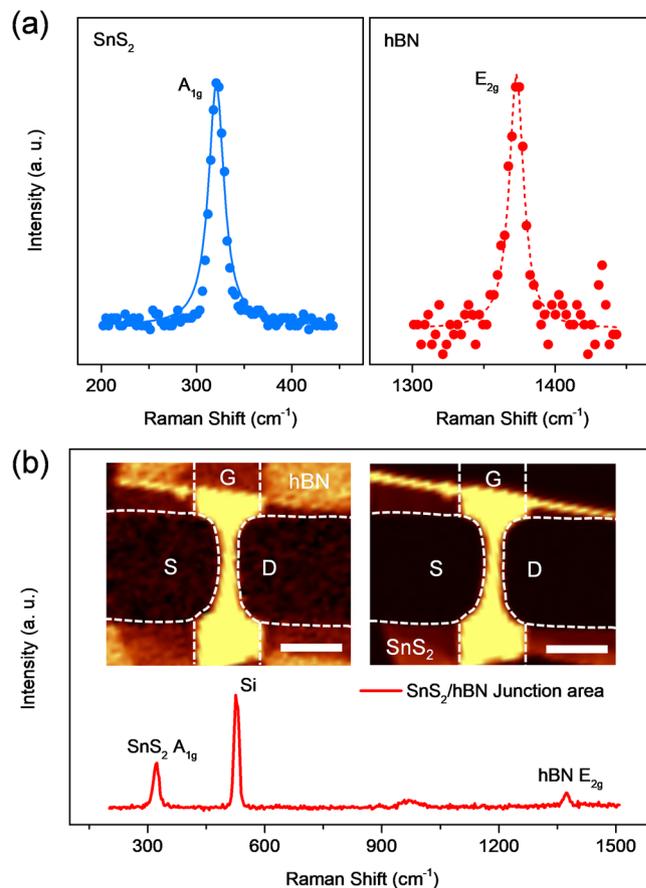


Figure 2. (a) Raman spectra of the multi-layered SnS₂ (left panel) and the hBN (right panel). From the data, the Raman peaks for SnS₂ and hBN occur respectively at 320.6 and 1372.4 cm⁻¹. (b) Raman signal taken in the SnS₂/hBN heterostructure region, showing peaks for the A_{1g} mode of SnS₂ and E_{2g} mode of hBN. Insets: Raman frequency mapping image displaying the E_{2g} peak intensity (left) and the A_{1g} peak intensity (right), respectively, for hBN and SnS₂. The scale bar is 5 μm.

drain voltage V_{DS} of 0.7 V, exhibiting an SS of 585 mV/decade in a 30-nm-thick hBN inserted device. The different drain voltages also revealed similar subthreshold swing characteristics (see Supplementary, Fig. S2a). This value is one order of magnitude smaller than that of a high- κ (Al₂O₃) covered SnS₂ transistor (in a top-gated configuration)¹⁰. It is well known that $SS = \ln(10)(k_B T/e)(1 + \eta)$ and $\eta = (C_D + C_{IT})/C_{BN}$, where k_B is the Boltzmann constant, T is the absolute temperature, e is the elemental charge, C_D is the depletion capacitance, $C_{IT} = e^2 D_{IT}$ is the interface trap capacitance, and C_{BN} is the bottom-gate capacitance¹⁹. Thus, our devices demonstrated a factor $\eta \approx 8$. Despite implications that high-quality SnS₂/hBN contact is indicated, the research has not yet fully explained how the interfacial quality is correlated with the electronic characteristics, especially for a gate stack. Nonetheless, we attribute such SS enhancement to the highly coupled interface with negligible chemical residues. Hysteretic effect in $I_{DS} - V_G$ characteristics often reflects the quality of channel/dielectric junction. The interfacial quality was further confirmed by the forward and backward direction sweeping of $I_{DS} - V_G$ transfer curves which results a negligible hysteresis by amount of <200 mV as displayed in Fig. S2b. Consistent results were observed in all of the samples with current switching ratios in the 10⁴ to 10⁵ range and n-type conduction, as described in the literature^{9,11,13}. The transconductance g_M defined by dI_{DS}/dV_G displays a maximum value of ~0.12 μS at $V_{DS} = 0.7$ V, as displayed in the inset of Fig. 3b. An important figure of merit of the transistor field-effect mobility μ_{FE} is determined by the relationship $\mu_{FE} = g_M/C_{BN}V_{DS} \times (L/W)$, where $C_{BN} = \epsilon_{BN}\epsilon_0/d_{BN}$ ($d_{BN} = 30$ nm is the thickness of the hBN layer and $\epsilon_{BN} = 3-4$ is the dielectric constant of the hBN)²³. As a result, μ_{FE} is calculated to be 0.1–0.5 cm²/Vs, comparable to those of single-layered MoS₂ (0.1–10 cm²/Vs range)²⁴. It should be noted that different growth method commonly influences the electrical properties of SnS₂ crystal. Song *et al.*¹⁰, De *et al.*¹¹ and Ahn *et al.*³³ reported the mobility of approximately 1–2 cm²/Vs from SnS₂ grown by vapor transport technique. However, the SnS₂ solid crystal prepared by vertical Bridgman technique have showed poor mobility around 0.1 cm²/Vs¹³. Beside contact engineering and dielectric interface improvement, the material's mobility seems influenced by growth method of SnS₂.

To establish efficient carrier injection from outside (e.g., S/D metal), which is needed to enhance the device performance, the contact issues have been preliminarily investigated for the MoS₂ system, and several novel approaches have been suggested²⁵. So far, the ohmic contact formation for SnS₂ materials is still unclear. Nevertheless, a linear increment in I_{DS} can be observed for different V_G bias conditions, suggesting an ohmic-like contact at the nickel/SnS₂ junction at a small V_{DS} bias range, as depicted in the inset of Fig. 3c. However, an

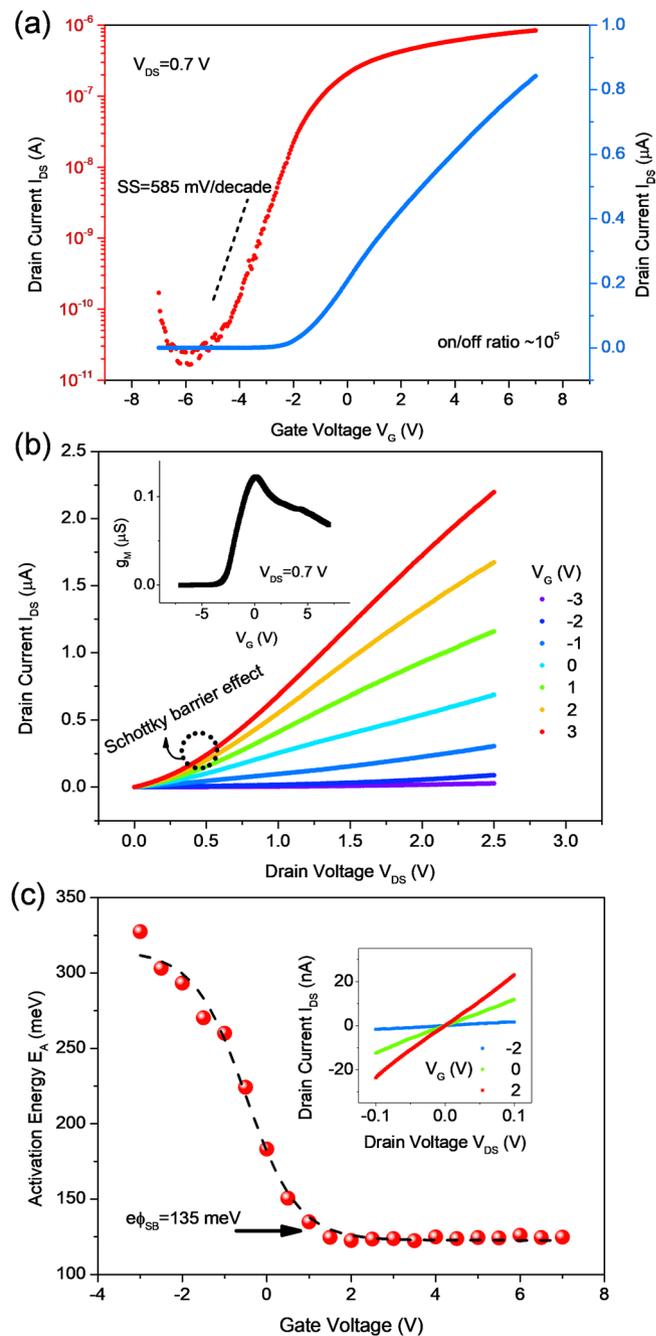


Figure 3. (a) Semi-log (left axis, red) and linear (right axis, blue) scale I_{DS} - V_G transfer characteristics of the multi-layered SnS_2 transistor biased at $V_{DS} = 0.7$ V. The device exhibited an SS as low as 585 mV/decade and an on/off ratio of approximately 10^5 at room temperature. (b) I_{DS} - V_{DS} output curves for various applied gate bias values from -3 to 3 V. The region in the black circle shows the nonlinear property. Inset: transconductance versus bottom-gate voltage at $V_{DS} = 0.7$ V, resulting in a maximum g_M peak of $0.12 \mu\text{S}$. (c) The extracted activation energy as a function of applied gate voltage. The Schottky barrier height is evaluated to be 135 meV for the Ni/ SnS_2 interface. Inset: I-V characteristics under small V_{DS} bias.

ambiguous result emerges when the V_{DS} is extended to a few voltages (quasi-linear region, before current saturation): a slightly nonlinear dependence of I_{DS} is found (indicated by the black circle) in I_{DS} - V_{DS} output characteristics of the SnS_2 device, as shown in Fig. 3b. We attribute this nonlinear behavior to the rise of the Schottky barrier height, $e\phi_{SB}$ because a contact mismatch occurs between the high work function, W_F of nickel metal ($W_F = 5.2$ eV)²⁶ and the electron affinity, $e\chi_s$ of SnS_2 ($e\chi_s = 5.0$ eV)²⁷. Owing to the similarity in crystal structure and chalcogenide compound compared with MoS_2 layered material, similar consequences could be expected for other 2D systems. To better address this point, we measured the temperature-dependent I-V characteristics as temperature varied from 300 to 410 K (Supplementary Information, Fig. S3). Carrier transport across a metal-semiconductor barrier involves a quantum mechanical tunneling and a thermionic-emission process, so that the

devices measured at high temperature regime allowed suppression of the tunneling current contribution²⁵. At a high temperature regime, an expression similar to the Arrhenius equation and also known as thermally activated transport model can be derived as $g_{DS} = g_0 \exp(-E_A/k_B T)$, where $g_{DS} = dI_{DS}/dV_{DS}$ is the conductance, and g_0 is the fitting parameter^{19,25}. The conductance g_{DS} fitted with this equation is depicted in Fig. S3b (see Supplementary Information). The activation energy E_A as a function of V_G acquired from Fig. S3b is illustrated in Fig. 3c. In this plot, we can determine a 135 meV of $e\phi_{SB}$ for Ni/SnS₂ contact by evaluation of the starting point of deviation from the linear response by following Radisavljevic and Kis²⁸. Such Schottky barrier determination is based on activation energy measurement. The details of the evaluation method of ϕ_{SB} can be found in other literatures^{28,29} as well as our previous publications^{19,25}. A measured $E_A = 0.18$ eV at zero V_G generally indicates the position of the impurity donor level with respect to the conduction band of SnS₂, and the activation energy is close to the value of 0.14 eV reported by Pan *et al.* and the value of 0.13 eV reported by De *et al.*^{9,11}.

Figure 4a shows the I–V transfer curves with (photon energy of 2.48 eV, green line) and without (dark state, black line) monochromatic light illumination at $V_{DS} = 0.1$ V. The current under illumination I_{ILL} , defined as $I_{ILL} = I_{PH} + I_{DA}$ (I_{PH} and I_{DA} are the photocurrent and dark current, respectively), exhibits a dramatic I_{DS} increment of the SnS₂ phototransistor in both the on and off states of the device, whereas the incident light with an intensity of 23.5 μ W has about a 30-fold influence in the off-state and 2-fold in the on-state. With light illumination on different states of the device (on- and off-states), the devices exhibit different photocurrent response. Lowering the Schottky barrier (on-state), an additional photocurrent excited by photo-induced band-to-band transition contributes to the drain current. Raising the Schottky barrier (off-state) restricts the dark current, resulting in a more pronounced photocurrent extraction. Therefore, we carefully conclude that photo-excited carrier transport primarily dominates over the thermionic and tunneling current, which is in agreement with other publications^{30,31}. We found that the effective transconductance g_M of the SnS₂ channel under light illumination showed clear increase compared to the dark state, as depicted in the inset of Fig. 4a. The SnS₂ phototransistor was further exposed to different monochromatic lights ranging in λ from 500 to 1000 nm, representing the series photo-induced I–V transfer properties at the on-state of the device with $V_{DS} = 0.1$ V, as displayed in Fig. 4b. Electron-hole pair generation by optical means usually requires an incident photon energy close to the band gap of the multi-layered SnS₂. Interestingly, the device weakly responds to light with a long λ (such as 600 nm, corresponding to 2.07 eV), implying an extrinsic type of the phototransistor with a defect-assisted energy level introduced. This effect can be explained by the defect-level involvement of the band gap of SnS₂; the transition between the defect-level and the conduction/valence band edge can contribute to I_{PH} ²⁹. Photoresponsivity, R_{PH} is an important metric of the phototransistor and is estimated by I_{PH}/P_L , where P_L is the optical power (see Fig. S4) and the broad spectral response is shown in Fig. 4c. In this photoresponsivity calculations, calibration of device's active area is excluded. The device performance exhibits an R_{PH} of 0.47–0.65 mA/W at the visible light range and is reduced to 0.33 mA/W at infrared due to the weak light absorption with an applied gating of 7 V. The measured R_{PH} as function of V_G is given in Fig. S4a. The responsivity of SnS₂/hBN devices is lower than that of MoS₂ based phototransistor (over 343 A/W)³⁰, but it is higher than that in a SnS₂ nanosheet photodetector reported by Tao *et al.* (around 1.13×10^{-3} mA/W under 532 nm photon wavelength)³² and vapor transport synthesized SnS₂ crystal reported by Ahn *et al.* (within 0.1–1 mA/W range)³³. Alternatively, optical characteristics of SnS₂ could be highly improved by synthesis technique, such as chemical vapor deposition (CVD) for minimizing sulfur vacancy. Yang *et al.* reported that the SnS₂ flake photodetectors prepared by CVD method archive significant improvement in photoresponsivity exceeding 1.19 A/W at 400 nm light³⁴. Analogues to MoS₂ crystal fabrication, different growth process may create different amount of sulfur vacancy in SnS₂ which have great impact on photodetector application as discussed by Xie *et al.* (details see their publication)³⁵. Therefore, we believe that extrinsic type of device with wide spectral response is probably due to sulfur vacancy induced deep states near bottom of conduction band.

Another key parameter is the detectivity, D^* which is the reciprocal of the noise equivalent power, given by $D^* = R_{PH} A^{1/2} / (2eI_{DA})^{1/2}$. Here, A is the device effective area. The calculated D^* value showed a typical range of 1.4×10^6 to 5.1×10^6 Jones at $V_{DS} = 0.1$ V and $V_G = 7$ V. Furthermore, the external quantum efficiency, EQE is measure of the ratio of the number of carriers produced by the number of photons. The EQE can be converted from R_{PH} by employing $EQE = R_{PH} hc / \lambda e$, here h and c are Planck constant and speed of light, respectively. We observed approximately 0.1% of EQE at visible light range.

Discussion

We fabricated SnS₂/hBN heterostructured devices and characterized the devices by electrical and optical measurements techniques. The interfacial behavior between the SnS₂ and hBN layered crystal is discussed. The locally placed gate separated by an hBN insulating layer presented an efficient modulation of the channel conductance with a current on/off ratio of up to $\sim 10^5$. The insertion of an ultra-flat dielectric layer allowed the device to exhibit SS values as low as 585 mV/decade. The detailed temperature-dependent electrical transport measurements led to the determination of $e\phi_{SB} = 135$ meV at the nickel/SnS₂ interface. Moreover, we demonstrated the extrinsic type of the SnS₂-based phototransistor with a wide range of light response and a high photoresponsivity of approximately 0.7 mA/W.

Methods

The bottom-gated SnS₂ devices were fabricated on a thermally oxidized n⁺-type silicon substrate in which a 90-nm-thick SiO₂ insulating layer offered electrical isolation from the back-gate, as well as optical detectability for the ultrathin nanosheet via optical contrast. The bottom electrode that served as both the optical indicator and the gate terminal of the transistor was pre-defined onto a silicon substrate by utilizing a standard photolithography process. We used SnS₂ and hBN bulk solids obtained from the 2D semiconductor Inc. to generate nano-flakes using a Scotch-tape mechanical exfoliation method. We employed a technique developed in our previous work

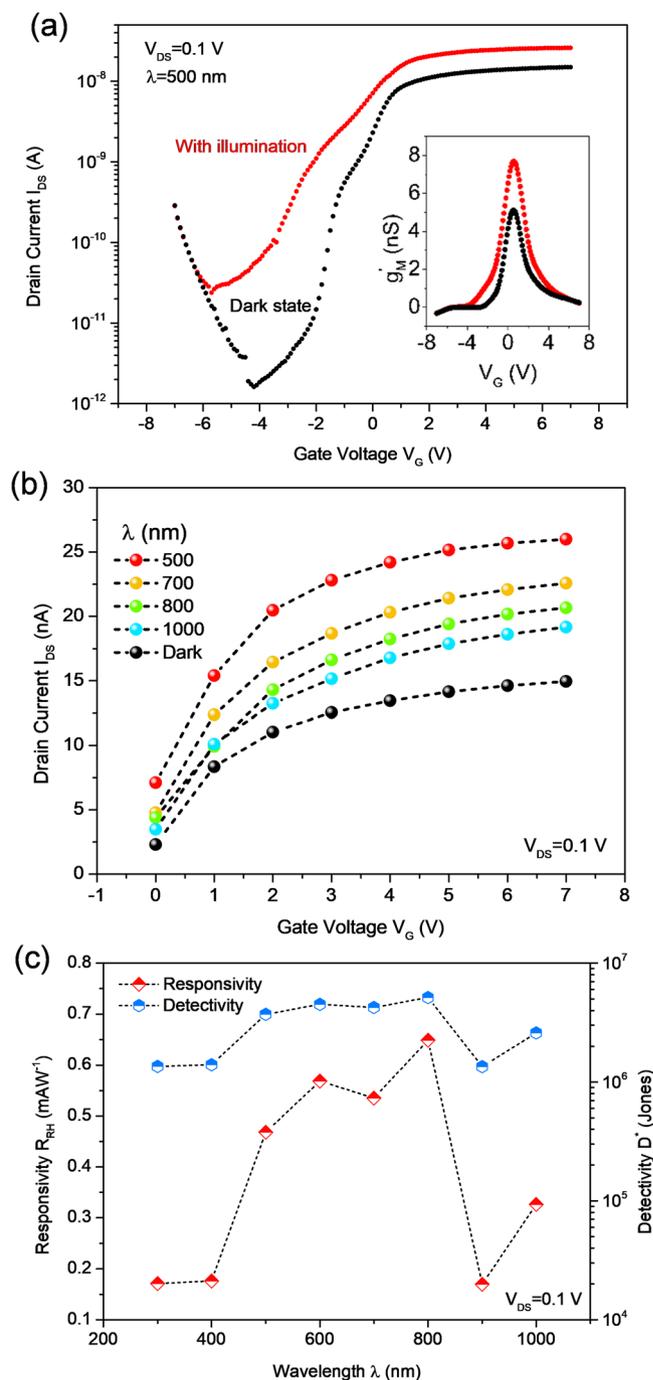


Figure 4. (a) Semi-log I_{DS} - V_G transfer characteristics of the SnS₂-based phototransistor for the dark state and for 500 nm wavelength illuminated curves at $V_{DS} = 0.1$ V. Inset: the effective transconductance of the device as a function of gate voltage. Black and red curves are the device under dark condition and 500-nm-wavelength light exposure, respectively. (b) Linear scale of transfer curves for different wavelengths (ranging from 500 to 1000 nm) in the accumulation region. (c) Photoresponsivity and detectivity of the device as functions of wavelength at $V_{DS} = 0.1$ V, showing a maximum R_{PH} of 0.65 mA/W.

called “dry transfer,” based on a polydimethylsiloxane framework, to transfer the desired hBN flake onto the pre-patterned gold bottom-gate^{2,19,25}. Subsequently, we deposited a piece of SnS₂ on top of the hBN layer using the same technique. The SnS₂ channel conductivity was monitored via metallization of the source/drain electrodes using a thermal evaporator system under a deposition rate of 5 Å/s to form a nickel/gold metal stack. An AFM (Park Systems, XE-100) operated under noncontact mode with a Nanosensor AR5-NCH tip was employed to characterize the topographic images of the devices. Raman signals were collected via commercially available confocal Raman spectroscopy (WiTec, alpha 300) with the excitation laser line of $\lambda = 488$ nm in ambient conditions.

The electrical transport properties of the SnS₂/hBN devices were obtained with a semiconductor parameter analyzer (Hewlett Packard, 4156 A) in a vacuum cryostat (ASK, 700 K) under a pressure of 10⁻³ Torr. The photo-induced I–V measurements were conducted similarly under ambient conditions. To probe the photocurrent measurements, light wavelength λ spectra ranging from 300 to 1000 nm were generated by a system that consisted of a 300 W Xenon Arc lamp, a power supply (Newport, 69911), and an automated 1/8 m monochromator (Newport, 74004) with double grating. The excitation light intensity was recorded through a silicon photodiode detector (Newport, 918D-UV-OD3) mounted optical power meter (Newport, 1918-C). The collected power and irradiance data as function of photon wavelength is given in Fig. S4(b). During the photoresponse characterization, the device (active area, $\sim 10^{-7}$ cm²) was illuminated by a monochromatic light guided by fused silica fiber optic bundle (Newport, 77577) with typical 3 mm in diameter uniform beam.

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Author Contributions

E.K.K. proposed the research and supervised the overall study. D.C. designed the experiment and performed the device fabrication, characterization, and data analysis. P.S.W. performed Raman measurements. D.C. and E.K.K. analyzed the results. D.C. prepared the data representation and wrote the manuscript. Dongil Chu was previously known as Dongri Qiu.

Additional Information

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