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## Hybrid quantum dot-tin disulfide field-effect transistors with improved photocurrent and spectral responsivity

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We report an improved photosensitivity in few-layer tin disulfide ( $\text{SnS}_2$ ) field-effect transistors (FETs) following doping with CdSe/ZnS core/shell quantum dots (QDs). The hybrid QD- $\text{SnS}_2$  FET devices achieve more than 500% increase in the photocurrent response compared with the starting  $\text{SnS}_2$ -only FET device and a spectral responsivity reaching over 650 A/W at 400 nm wavelength. The negligible electrical conductance in a control QD-only FET device suggests that the energy transfer between QDs and  $\text{SnS}_2$  is the main mechanism responsible for the sensitization effect, which is consistent with the strong spectral overlap between QD photoluminescence and  $\text{SnS}_2$  optical absorption as well as the large nominal donor-acceptor interspacing between QD core and  $\text{SnS}_2$ . We also find enhanced charge carrier mobility in hybrid QD- $\text{SnS}_2$  FETs which we attribute to a reduced contact Schottky barrier width due to an elevated background charge carrier density.

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Two-dimensional (2D) layered nanomaterials have sparked significant research interest for their potential applications in new types of electronic and optoelectronic devices, including field-effect transistors (FETs), sensors, solar cells, photodetectors, and light emitting diodes.<sup>1–8</sup> Graphene is probably the most popular example of a 2D nanomaterial that has been extensively explored over the years. However, its application in high-performance optoelectronic devices, such as photodetectors<sup>3,9</sup> and photovoltaics,<sup>5,10</sup> has been limited due to its zero energy bandgap. Layered metal dichalcogenides (LMDs), such as  $\text{MoS}_2$ ,  $\text{WS}_2$ , and  $\text{WSe}_2$ , have emerged as alternatives, primarily because of their tunable bandgap energy controlled by thickness (spanning from 1 eV to  $\sim 3$  eV) and the transition from an indirect bandgap in the bulk to a direct gap in the monolayer,<sup>2,7,11–15</sup> which in turn enhances the 2D material's photoluminescence (PL) quantum yield.<sup>15</sup> Combined with a large surface-to-volume ratio, these material characteristics make LMDs appealing for light harvesting, energy conversion, and chemical sensing device applications. Tin disulfide ( $\text{SnS}_2$ ) is a less studied type of LMD, in which the group IV element Sn substitutes the transition metals in other, more familiar LMD compounds such as  $\text{MoS}_2$  and  $\text{WSe}_2$ . We recently demonstrated FET devices based on mechanically exfoliated  $\text{SnS}_2$  flakes and achieved high performance on par with those based on  $\text{MoS}_2$ .<sup>14,16</sup> While bulk  $\text{SnS}_2$  is known to be an n-type semiconductor with an indirect bandgap of 2.2 eV, we found that the bandgap of  $\text{SnS}_2$  remained indirect even when the layer

thickness was decreased down to a single layer, indicating a potentially inferior light absorption cross-section compared with the direct bandgap LMD materials. Recently, hybrid devices combining 2D LMDs with other semiconductors have been explored for the purpose of improving the device performance and functionality.<sup>17–20</sup> Especially for enhancing light absorption and device photosensitivity, the use of colloidal quantum dots (QDs) in a light sensitizing layer in contact with the LMD has been recently proposed.<sup>17,21</sup> QDs have large optical absorption cross-section and wide spectral coverage spanning from ultraviolet to near infrared, depending on their core sizes. Combined with the size-dependent tunable bandgap, such characteristics prove highly effective in the utilization of QDs toward enhancing the optical responsivity of 2D material-based photo-FET devices.<sup>17</sup> We propose that a similar QD-based sensitization scheme should be able to improve the photosensitivity of 2D  $\text{SnS}_2$  FETs.

Here we report improved light detection sensitivity in a few-layer  $\text{SnS}_2$  FET by employing a CdSe/ZnS core-shell QD light sensitization layer. Benefiting from the strong optical absorption of QD as well as the spectral overlap between the PL emission of QDs and  $\text{SnS}_2$  absorption spectra, which enables an efficient energy transfer from photo-excited QDs to  $\text{SnS}_2$ , QD- $\text{SnS}_2$  hybrid FET devices exhibit more than 500% increase in the measured photocurrent response with the corresponding spectral responsivity greater than 650 A/W at 400 nm wavelength. We find that QD sensitization and light illumination induce an increase in the field-effect electron mobility in the layered  $\text{SnS}_2$ , which we explain based on a reduced contact Schottky barrier width via the increased photo-excited charge carrier density.

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Few-layer  $\text{SnS}_2$  flakes were prepared by mechanical exfoliation from a bulk crystal grown by the vertical Bridgman method and dry-transferred onto a silicon wafer with a 300 nm thick  $\text{SiO}_2$  dielectric layer, which was also used as a back-gated substrate for FET fabrication and characterization.<sup>14</sup> After locating target  $\text{SnS}_2$  flakes by bright-field optical microscopy, source-drain contacts (Ti/Au, 10/50 nm) of FETs with 15  $\mu\text{m}$  channel length were fabricated by optical lithography and electron-beam deposition. For the active FET channels, we utilized few-layer  $\text{SnS}_2$  flakes ( $\sim 10$  monolayers, determined by bright-field optical contrast),<sup>14</sup> instead of monolayer  $\text{SnS}_2$ , because the thicker flakes provide increased light absorption. This enhanced absorption is beneficial not only for the exciton generation within the 2D material but also for the enhanced energy transfer from QDs to  $\text{SnS}_2$ , as observed in our recent single-particle spectroscopy study, where the energy transfer rate was found to increase with the increasing number of  $\text{SnS}_2$  layers.<sup>22</sup>

Figure 1(a) shows a schematic of back-gated QD-few-layer  $\text{SnS}_2$  hybrid FET device with CdSe/ZnS QDs deposited on top of the  $\text{SnS}_2$  channel via drop-casting from mixed solvents (hexane:octane of 9:1 volume ratio with QD solid concentration of 2.5 mg/l). The  $\text{SnS}_2$  flake itself exhibits an optical absorption spectrum increasing sharply below  $\sim 550$  nm (Figure 1(b)), which suggests a bandgap energy of  $\sim 2.3$  eV. We utilized octadecylamine-coated core/shell CdSe/ZnS QDs with PL spectrum centered at  $\sim 535$  nm (Ocean NanoTech) to impose a strong overlap with the absorption spectrum of  $\text{SnS}_2$  (Figure 1(b)). The PL emission peak wavelength of 535 nm is associated with a CdSe core with  $\sim 3.4$  nm diameter and one monolayer (0.7 nm) thick ZnS shell.<sup>22</sup> Assuming a thickness of  $\sim 2.3$  nm for the octadecylamine ligand coating on the QD surface, the interspacing between the CdSe core (edge) and the first layer of the  $\text{SnS}_2$  flake is thus estimated to be  $\sim 3$  nm, a

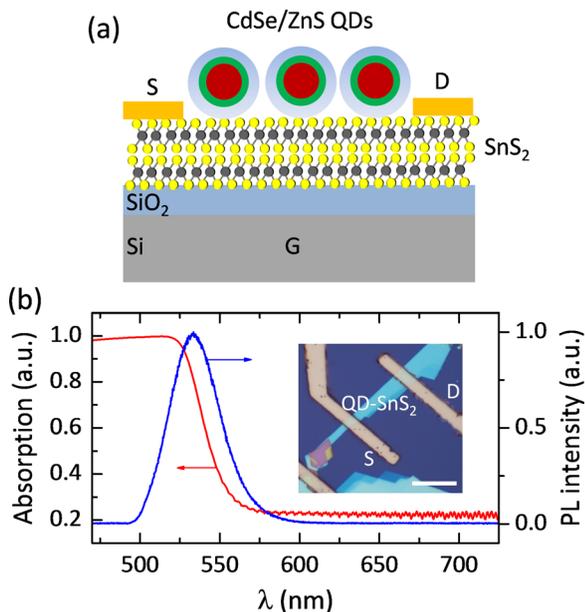


FIG. 1. (a) Schematic depicting a hybrid CdSe/ZnS QD- $\text{SnS}_2$  FET device, with S, D, and G denoting source, drain, and gate, respectively. (b) Optical absorption spectrum of a few-layer  $\text{SnS}_2$  flake (red, left) and PL spectrum of suspended CdSe/ZnS QDs in toluene (blue, right). The inset is an optical image of an actual QD-few-layer  $\text{SnS}_2$  hybrid FET device. Scale bar: 10  $\mu\text{m}$ .

distance at which we expect energy transfer to dominate over charge transfer. This large separation between QDs and  $\text{SnS}_2$ , in tandem with the strong spectral overlap between QD PL and  $\text{SnS}_2$  optical absorption, enables energy transfer from photo-excited QDs to  $\text{SnS}_2$ , as we have recently confirmed by time-resolved single-particle PL studies of QD- $\text{SnS}_2$  hybrids.<sup>22</sup>

We next measured FET device characteristics in ambient air, under dark and white-light-illuminated (tungsten lamp  $\sim 3.5$  mW) conditions. An optical image of the QD- $\text{SnS}_2$  hybrid FET device is shown in the inset of Figure 1(b). The  $\text{SnS}_2$  FET device without QDs and in the dark displays typical n-type drain-source current-voltage ( $I_{DS} - V_{DS}$ ) characteristics with increasing  $I_{DS}$  toward positive gate voltage,  $V_G$ . The dark  $I_{DS}$  reaches  $\sim 8$  nA at  $V_G = 20$  V ( $V_{DS} = 0.5$  V), being increased  $\sim 10\times$  compared to  $I_{DS}$  at  $V_G = -20$  V (Figure 2(a), black curves). Under white light illumination, the photoconductive effect in  $\text{SnS}_2$  enhances  $I_{DS}$  to  $\sim 57$  nA (at  $V_G = 20$  V,  $V_{DS} = 0.5$  V), which is  $\sim 7\times$  larger than the dark  $I_{DS}$  obtained under the same conditions (Figure 2(a), red curves). The absolute photocurrent,  $\Delta I_{DS,photo} = I_{DS,illuminated} - I_{DS,dark}$ , increases from  $\sim 10$  nA at  $V_G = -20$  V up to  $\sim 50$  nA at  $V_G = 20$  V (for  $V_{DS} = 0.5$  V, Figure 2(a) inset). Meanwhile, the FET does not turn off fully under illumination even at  $V_G = -20$  V ( $I_{DS} = \sim 14$  nA), indicating an elevated background charge carrier density induced by photo-excitation.

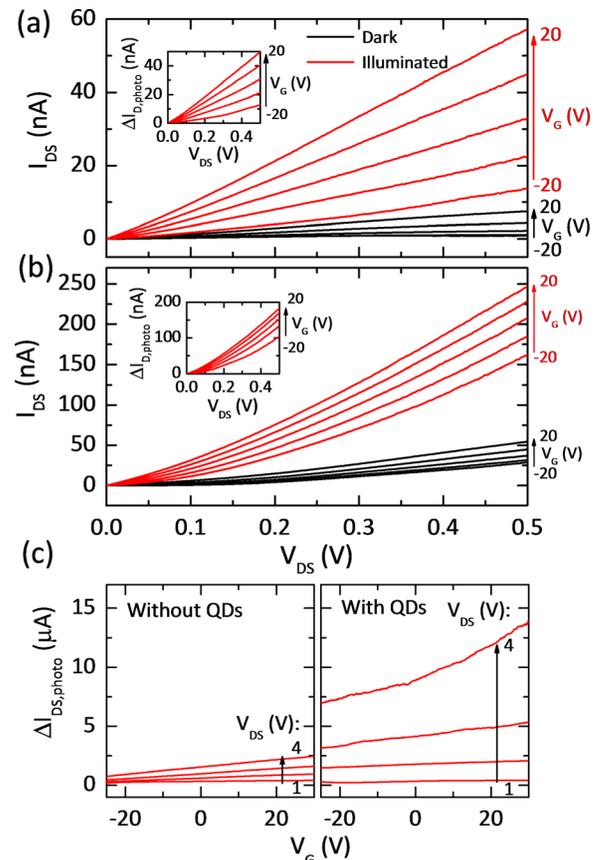


FIG. 2.  $I_{DS} - V_{DS} - V_G$  characteristics of (a)  $\text{SnS}_2$  FET device and (b) QD- $\text{SnS}_2$  hybrid FET device in the dark (black curves) and under white light illumination (red curves).  $V_G$  is varied from  $-20$  to  $20$  V, in steps of  $10$  V. Insets show absolute photocurrent outputs ( $\Delta I_{DS,photo}$ ) with respect to  $V_{DS}$  and  $V_G$ . (c)  $\Delta I_{DS,photo} - V_G$  characteristics for extended ranges of  $V_G$  and  $V_{DS}$ , for  $\text{SnS}_2$  FET device (left) and QD- $\text{SnS}_2$  hybrid FET device (right).  $V_{DS}$  varies from  $1$  V to  $4$  V in  $1$  V increments (indicated by black arrows).

For the hybrid FET device with QDs deposited on top of the SnS<sub>2</sub> channel, we observed a significant increase in  $I_{DS}$  in the dark as well as under white-light illumination. The dark  $I_{DS}$  reached 54 nA at  $V_{DS} = 0.5$  V and  $V_G = 20$  V (Figure 2(b), black curves), comparable to the  $I_{DS}$  obtained from the bare SnS<sub>2</sub> FET (without added QDs) under illumination. At  $V_G = -20$  V,  $I_{DS}$  still remains as high as 29 nA, indicating only  $1.8\times$  modulation of  $I_{DS}$  (i.e., on-off ratio) by a  $V_G$  change of  $-40$  V, much reduced compared with  $\sim 10$  on-off ratio observed in the bare SnS<sub>2</sub> FET device. This suggests an increased background carrier density in QD-SnS<sub>2</sub> hybrid FETs even under dark conditions, induced by the application of QDs. We speculate that QDs may introduce a pseudogating effect, particularly via their positively charged octadecylamine ligands, which can exert a positive gate electric field onto the QD-SnS<sub>2</sub> hybrid FET device. Similar phenomena have been observed in other hybrid systems, such as graphene/polyvinylidene fluoride (PVDF) and 2D MoS<sub>2</sub>/PbS QDs.<sup>6,17</sup>

When the hybrid QD-SnS<sub>2</sub> FET was exposed to white light,  $I_{DS}$  increased to  $\sim 250$  nA (at  $V_{DS} = 0.5$  V and  $V_G = 20$  V; Figure 2(b), red curves), compared to the dark  $I_{DS}$  of 54 nA measured at the same bias condition. Similar to the SnS<sub>2</sub>-only FET device, the photocurrent output of hybrid QD-SnS<sub>2</sub> FET increases for larger  $V_{DS}$  and  $V_G$  values, with  $\Delta I_{DS,photo}$  now reaching up to  $\sim 185$  nA ( $V_{DS} = 0.5$  V,  $V_G = 20$  V; Figure 2(b) inset), representing a 370% enhancement in the photocurrent output compared to the SnS<sub>2</sub>-only FET. Even at  $V_G = -20$  V,  $\Delta I_{DS,photo}$  for the hybrid FET is as large as  $\sim 100$  nA, again indicating a significantly elevated background carrier density. Overall, these enhanced photocurrent outputs demonstrate the positive contribution of QDs to the photosensitivity of SnS<sub>2</sub> FET devices. A survey of extended ranges of  $V_G$  (from  $-30$  V to  $30$  V) and  $V_{DS}$  (from  $1$  V to  $4$  V) shows that  $\Delta I_{DS,photo}$  of the hybrid QD-SnS<sub>2</sub> FET can be further increased to over  $10$   $\mu$ A by applying larger  $V_G$  and  $V_{DS}$ . At  $V_{DS} = 4$  V (Figure 2(c)),  $\Delta I_{DS,photo}$  of the hybrid QD-SnS<sub>2</sub> FET increases from  $\sim 7$   $\mu$ A to  $\sim 13$   $\mu$ A as  $V_G$  changes from  $-30$  V to  $30$  V, in contrast to a smaller increase of  $\Delta I_{DS,photo}$  observed in the SnS<sub>2</sub>-only FET, from  $\sim 0.2$   $\mu$ A to  $2.5$   $\mu$ A for the same bias variation. This means that the hybrid QD-SnS<sub>2</sub> FET device has  $>500\%$  enhanced  $\Delta I_{DS,photo}$  at  $V_G = 30$  V compared with the SnS<sub>2</sub>-only FET. For the entire range of explored  $V_{DS}$  values,  $\Delta I_{DS,photo}$  in the hybrid FET is always larger than that in the SnS<sub>2</sub>-only FET.

The hybrid FET meanwhile cannot be completely turned off even at  $V_G = -30$  V. A similar behavior was reported by Kufer *et al.* for hybrid PbS QD-MoS<sub>2</sub> FETs,<sup>17</sup> where the incorporation of QDs was found to significantly decrease the FET on-off ratio. It is noteworthy that in their demonstration, the authors explained that the enhanced photodetection of MoS<sub>2</sub> by the incorporation of PbS QDs was resulting from the *charge transfer* from photo-excited PbS QDs to MoS<sub>2</sub>. In our study, the insulating shell and ligand coating (giving an estimated spacing between CdSe core and SnS<sub>2</sub> of  $\sim 3$  nm) are believed to prevent such a charge transfer and favor *energy transfer* (via photons, resulting from PL of the QDs and in turn absorbed in the SnS<sub>2</sub>). The absence of charge transfer can be further confirmed by measuring the electrical conductance of a CdSe/ZnS QD-only FET device (i.e., QDs

drop-cast directly onto SiO<sub>2</sub>, with contact geometry similar to the device shown in Figure 1(b), but without a SnS<sub>2</sub> flake). In such a device, we observed a negligible current under both dark and illuminated conditions,<sup>23</sup> proving a negligible charge transfer due to the insulating ZnS shell and ligand coating, and thus confirming that the main interaction mechanism between CdSe/ZnS QDs and SnS<sub>2</sub> is the energy transfer.

We also measured the spectral responsivity,  $R = \Delta I_{DS,photo}/P_{light}$  with  $P_{light}$  being the incident light power on the device active area, for both the SnS<sub>2</sub>-only and hybrid QD-SnS<sub>2</sub> FET devices in the wavelength ( $\lambda$ ) range from  $400$  nm to  $600$  nm, at  $V_{DS} = 1$  V and  $V_G = -10$  V (Figure 3). Overall,  $R$  is enhanced for the hybrid QD-SnS<sub>2</sub> FET, reaching  $\sim 650$  A/W at  $\lambda = 400$  nm (cf. dark  $I_{DS} \sim 0.1$  nA), compared with  $400$  A/W for the non-sensitized SnS<sub>2</sub>-only FET. The onset of  $R$  starts at  $\sim 550$  nm for both FET devices (Figure 3 inset), which is consistent with the absorption band edge of SnS<sub>2</sub> (Figure 1(b)). The optical absorption spectrum of CdSe/ZnS QDs in fact also starts at  $\sim 550$  nm,<sup>24</sup> and thus the measured  $R$  after adding QDs mainly features an overall enhanced magnitude with an insignificant change in the spectral shape. It is noted that the onset of  $R$  is slow unlike the absorption spectrum (Figure 1(b)), which we suspect to be related to the presence of charge trap states near the band edges of SnS<sub>2</sub>. Such traps would induce a significant carrier recombination and decreased photocurrent output, consequently lowering the  $R$  value near the absorption band edge.

Apart from the optical sensitization, we also found that the hybrid QD-SnS<sub>2</sub> FET under illumination exhibits enhanced field-effect carrier mobility ( $\mu$ ) compared with the few-layer SnS<sub>2</sub>-only FET. Figure 4(a) shows  $I_{DS} - V_G$  plot at  $V_{DS} = 0.5$  V, where the FET is not saturated, and thus  $\mu$  is proportional to the transconductance ( $dI_{DS}/dV_G$ ) via the following relation:<sup>25</sup>  $\mu = (dI_{DS}/dV_G) \times L/(W \cdot C \cdot V_{DS})$  with  $L$  and  $W$  representing the length ( $15$   $\mu$ m) and width ( $\sim 5$   $\mu$ m) of the FET device channel, and  $C = 11.6$  nF/cm<sup>2</sup> being the capacitance of the  $300$  nm thick SiO<sub>2</sub> gate dielectric. Figure 4(b) summarizes the obtained  $\mu$  values. In the dark, the SnS<sub>2</sub>-only FET (no QDs) displays  $\mu = 0.14$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>, which is comparable to (but at the low end of) field-effect mobilities reported previously for back-gated few-layer SnS<sub>2</sub> FETs at the room temperature.<sup>14</sup> Interestingly,  $\mu$  increases to over  $0.6$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>

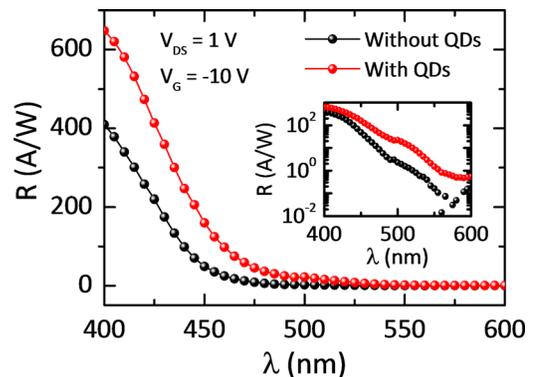


FIG. 3. Spectral responsivity  $R$  of a few-layer SnS<sub>2</sub> FET device at  $V_G = -10$  V and  $V_{DS} = 1$  V, before (black symbols) and after (red symbols) the application of the CdSe/ZnS QD sensitization. Inset shows a semi-logarithmic plot of the same data.

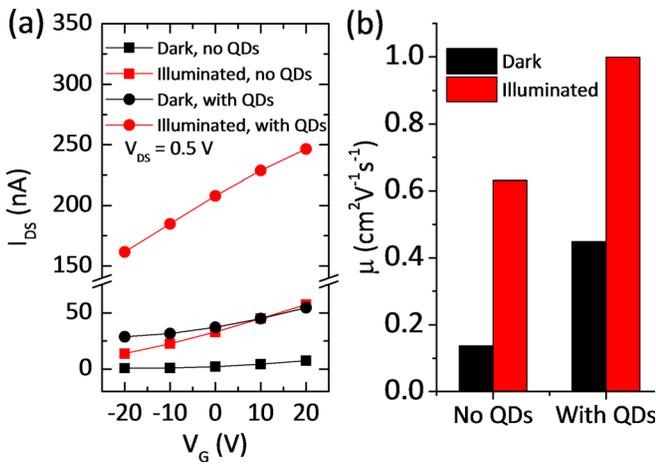


FIG. 4. (a)  $I_{DS} - V_G$  characteristics of few-layer SnS<sub>2</sub> FET at  $V_{DS} = 0.5$  V, before (square) and after (circle) CdSe/ZnS QD sensitization, in the dark (black) and under white-light illumination (red). (b) Histogram summarizing the field-effect mobility,  $\mu$ , calculated from transconductances obtained in (a).

upon white-light illumination. For the hybrid QD-SnS<sub>2</sub> FET,  $\mu$  is already higher than  $0.4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  even in the dark, and increases to  $\sim 1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  under white light illumination, which represents a  $\sim 7\times$  increase compared with the SnS<sub>2</sub>-only FET in the dark.

In general, the experimentally measured  $\mu$  can be affected by several extrinsic factors, such as contact barrier height, impurity scattering, and temperature. For example,  $\mu$  in monolayer MoS<sub>2</sub> reported in early studies was lower than  $10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ,<sup>26</sup> but it was later shown that the use of graphene contact and h-BN encapsulation could enhance the measured  $\mu$  in monolayer MoS<sub>2</sub> above  $1000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ,<sup>27</sup> and even as high as  $34\,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for six-layer MoS<sub>2</sub>.<sup>28</sup> This suggests a much higher intrinsic  $\mu$  in the 2D material. Given our observed FET device characteristics that show an increased background carrier density during illumination and/or after application of QDs on the SnS<sub>2</sub> FET, a potential cause for the observed increase in the measured  $\mu$  might be a reduced contact Schottky barrier width  $W_D$ , a property which is inversely proportional to the carrier density  $N$  (i.e.,  $W_D \propto 1/\sqrt{N}$ ).<sup>29</sup> Although the precise  $W_D$  in the ultrathin SnS<sub>2</sub> FET device should be obtained by solving the 2D Poisson equation considering the exact device geometry, the basic qualitative relation between the in-plane Schottky junction depletion width from the electrodes and the background carrier density in SnS<sub>2</sub> channel should hold true. Since  $N$  affects the FET threshold voltage ( $V_T$ ) according to  $V_T \propto \sqrt{N}$ ,<sup>29</sup> one can then expect that  $W_D$  decreases with increasing  $V_T$  (i.e.,  $W_D \propto 1/V_T$ ). From Figure 4(a), we can estimate  $V_T$  by extrapolating the linear  $I_{DS} - V_G$  plots to  $I_{DS} = 0$ , and find  $V_T$  of  $\sim 10$  V for the SnS<sub>2</sub>-only FET in the dark, which increases to  $\sim 100$  V in the hybrid QD-SnS<sub>2</sub> FET under illumination. This  $10\times$  increase in  $V_T$  implies a tenfold decrease in  $W_D$  and thus an easier carrier tunneling through the Schottky barriers at the source and drain electrodes.

We note that the observed  $V_T$  shift by the pseudo-gating effect of QDs may in part contribute to the increase in photocurrent response gain. We can estimate the extent of contribution from such a  $V_T$  shift (approximately 45 V, estimated from the dark  $I_{DS} - V_G$  curves with/without QDs in Figure 4(a)) by

examining the  $V_G$ -dependent photocurrent output ( $\Delta I_{DS,photo}$ ) data (Figure 2(c)). For instance,  $\Delta I_{DS,photo}$  of SnS<sub>2</sub> only device at  $V_G = 0$  V would become  $\sim 2.5 \mu\text{A}$  by 45 V shift of  $V_T$  (Figure 2(c) left panel, at  $V_{DS} = 4$  V). On the contrary,  $\Delta I_{DS,photo}$  of QDs-applied SnS<sub>2</sub> device at  $V_G = 0$  V is  $\sim 4 \mu\text{A}$  (Figure 2(c) right panel, at  $V_{DS} = 4$  V); therefore, showing at least  $\sim 38\%$  additional contribution that is not accounted by the simple  $V_T$  shift. Given the enhanced spectral responsivity starting near the absorption edge of QDs (Figure 3), it is logical to ascribe the origin of this additional photocurrent gain to the contribution from the energy transfer by QDs. We note that the extent of  $V_T$  shift contribution estimated above is an upper bound value and should be less considering that  $I_{DS}$  tends to saturate at high  $V_G$ .

In summary, we combined core/shell CdSe/ZnS QDs with the few-layer SnS<sub>2</sub> to fabricate hybrid QD-SnS<sub>2</sub> FETs with improved photo-detection sensitivity via energy transfer from photo-excited QDs to SnS<sub>2</sub>. Photo-sensitization of SnS<sub>2</sub> by the added QDs resulted in an over  $5\times$  enhanced photocurrent response in the hybrid FET with the corresponding device spectral responsivity reaching  $650 \text{ A/W}$  at  $400 \text{ nm}$ . We also found that the QD sensitization as well as light illumination enhanced the measured carrier mobility in SnS<sub>2</sub>, which we correlate with an elevated background charge carrier density and consequently decreased contact Schottky barrier width. Our results demonstrate that the energy-transfer-based QD sensitization can be utilized as a new route for enhancing the light harvesting performance of 2D LMD-based optoelectronic devices.

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