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High-performance photocurrent generation from two-dimensional WS$_2$ field-effect transistors

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The generation of a photocurrent from two-dimensional tungsten disulfide (WS$_2$) field-effect transistors is examined here, and its dependence on the photon energy is characterized. We found from the WS$_2$ devices that a significant enhancement in the ratio of illuminated current against dark current ($I_{\text{ill}}/I_{\text{dark}}$) of ~$10^3$–$10^4$ is attained, even with the application of electric fields of $E_D = 0.02$ and $E_G = -22$ mV/nm, which are much smaller than that of the bulk MoS$_2$ phototransistor. Most importantly, we demonstrate that our multilayer WS$_2$ shows an extremely high external quantum efficiency of ~7000%, even with the smallest electrical field applied. We also found that photons with an energy near the direct band gap of the bulk WS$_2$, in the range of 1.9–2.34 eV, give rise to a photoresponsivity of ~0.27 A/W, which exceeds the photoresponsivity of the bulk MoS$_2$ phototransistor. The superior photosensing properties of WS$_2$ demonstrated in this work are expected to be utilized in the development of future high performance two-dimensional optoelectronic devices. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4878335]

Two-dimensional (2D) materials are attractive for use in a variety of electronic devices that can benefit from their atomically thin flexible and transparent layer structures and their low-dimensionality, which provides quantum mechanical properties that are not present in conventional three-dimensional materials. 2D materials can potentially enable the devices of the post-silicon era by overcoming the major obstacles presented by current silicon semiconductor devices, including short-channel effects and poor power dissipation. Such materials open avenues for photonic applications that take advantage of variations in a material’s band gap properties as a function of the 2D layer thickness.

Transition metal dichalcogenides (TMDC) are the most widely studied 2D materials because they can provide a range of material properties: superconducting, semiconducting, or metallic, depending on the atomic and electronic structures arising from combinations of the transition metal and chalcogen atoms. Particularly, TMDCs formed by Mo or W transition metal atoms can form semiconductors with band gaps that correspond to the visible to infrared absorption spectra. These materials are potentially useful in digital electronics or photonic devices. Recent studies have demonstrated that the band gap of 1.1–2.1 eV (Ref. 3) in a TMDC material is useful for a variety of devices, including photodetectors, photovoltaics, light-emitting diodes (LEDs), field-effect transistors (FETs), logic, memory, and sensors.

The TMDC tungsten disulfide (WS$_2$) has an indirect band gap of 1.4 eV in bulk and a direct band gap of 2.1 eV in a monolayer, which is affected by quantum confinement effects. Although the chemical and atomic structures of WS$_2$ are similar to those of molybdenum disulfide (MoS$_2$), WS$_2$ has been studied to a lesser degree than MoS$_2$, possibly due to the difficulty in obtaining high quality single crystal WS$_2$. However, the inert, non-toxic, and environmentally friendly properties of WS$_2$ make it attractive as a potential electronic material. Field-effect transistors prepared from WS$_2$ have recently been demonstrated in the previous reports and the ambipolar charge carrier characteristic of WS$_2$, which is more frequently reported than other TMDC materials, makes it more attractive for use in device applications that involve homogenous or heterogeneous p-n junction. Most importantly, according to the simulation results, WS$_2$ is reported to have much smaller effective electron mass and to provide better transistor performance than Si as well as MoS$_2$. The structural, electronic, and optical properties of WS$_2$ have been studied theoretically and experimentally. The differential reflectance and photoluminescence (PL) spectra revealed indirect-to-direct gap transition features of WS$_2$ that resemble those observed in other TMDCs. A Raman spectroscopy study verified that the number of layers in a WS$_2$ sample could be determined by analyzing the Raman peak shift. Previous studies have described the photosensing properties of TMDC materials, e.g., MoS$_2$, WS$_2$, MoS$_2$-graphene, and MoS$_2$-silicon heterostructures. Interestingly, investigations into the photoresponse of WS$_2$, including the spectral photoresponse, the $I_{\text{ill}}/I_{\text{dark}}$, and the switching behavior, are rare compared to those that have investigated MoS$_2$.

In this Letter, we investigate the spectral photoresponse of WS$_2$, by using an electrical measurement system capable...
of monochromatic light illumination onto a WS$_2$ field-effect transistor. The optoelectronic properties as the key figures of merit for optical sensors and switching devices, the $I_{\text{illum}}/I_{\text{dark}}$, photoresponsivity, and external quantum efficiency (EQE), were measured from WS$_2$ devices. The spectral response of the external quantum efficiency reveals that an enhanced photocurrent is generated upon illumination at photon energies very close to the direct band gap of WS$_2$.

A $\sim 20$ nm thick multi-layer WS$_2$ film is mechanically exfoliated from a bulk WS$_2$ crystal and transferred to a silicon wafer covered by a 90 nm thick silicon dioxide (SiO$_2$) layer formed on a highly doped $n$-type Si wafer with a resistivity $<5 \times 10^{-3}$ $\Omega$ cm using the conventional micro-mechanical cleavage method. The substrate is baked on a hot plate at 100 $^\circ$C for 10 min prior to transferring the WS$_2$ layer and evaporating water molecules onto the surface of the substrate.

FIG. 1. (a) Schematic diagram showing the WS$_2$ device and the measurement setup. Right inset: AFM image of a WS$_2$ device. The yellow line indicates the profile along the green line. Left inset: optical microscope image of the device. Scale bar is 10 nm. (b) Transfer curves with and without light illumination. The $I_{\text{illum}}/I_{\text{dark}}$ over almost two orders of magnitude is observed near $V_g = -2$ V. The gate leakage level is indicated as a black arrow. Measurements were performed at $V_g = 20$ mV and $P_{\text{opt}} = 250$ mW/cm$^2$. Inset: output curves of the WS$_2$ device. It shows Ohmic contact behavior.
\[ \eta = \frac{J_{\text{ph}}}{q \phi} = \frac{J_{\text{ph}}}{q} \times \left( \frac{h}{P_{\text{opt}}} \right), \]

where \( J_{\text{ph}} \) is the photocurrent density, \( \phi \) is the photon flux \( \left( = \frac{P_{\text{opt}}}{h} \right) \), \( h \) is Planck’s constant, \( \nu \) is the wavelength of light, \( q \) is the electron charge, and \( P_{\text{opt}} \) is the illumination power density. A high EQE was achieved by modulating the electrical bias applied to the WS2 device (see Fig. 3(a)). Under a positive gate bias, the EQE is two orders of magnitude greater than the EQE measured under a negative gate bias. The EQE continues to increase as \( V_D \) increases. A maximum EQE of \( \sim 7000\% \) at a relatively high bias, e.g., \( V_D = 1 \text{ V} \) and \( V_G = 3 \text{ V} \), was attained from our measurement, whereas EQE values of \( \sim 8\% \) and \( \sim 70\% \) are attained at a low \( V_D \) of 20 \text{ mV} for the transistor off-state and on-state, respectively. Our bulk WS2 phototransistor shows comparable EQE with that of bulk MoS2 device even \( \sim 15\text{-folds} \) smaller application of electric field and three order higher EQE than that of previously reported the phototransistor with multi-layer WS2 synthesized by chemical vapor deposition (CVD) even with 30-folds smaller application of the electric field (see the inset of Fig. 3(a)). The spectral EQE was measured over the incident wavelength range of 400–700 nm, revealing three peaks (see Fig. 3(b)). The peaks A (\( \sim 630 \text{ nm} \)) and B (\( \sim 530 \text{ nm} \)) originate from the energies of the direct band gaps that formed between the split valance and conduction band at the
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K point of WS$_2$.  

Peak C ($\sim$460 nm) arises from the optical transition between the density of states peaks in the valence band and the conduction bands\textsuperscript{25} (see the inset of Fig. 3). This observation agrees well with a previously reported PL study of WS$_2$\textsuperscript{25}.

Figs. 4(a) and 4(b) show the flat band and equilibrium states of the band diagrams of WS$_2$ devices, in which the work function of Ti, the band gap of WS$_2$, and the electron affinity of WS$_2$ are 4.3, 1.3, and 4.5 eV, respectively\textsuperscript{15}. The positive drain bias bends the band structure, as shown in Fig. 4(c). The application of a negative gate bias blocks the charge carrier flow (as indicated by the red dashed-dotted arrow) due to upshifted conduction and valence bands (as indicated by the red line); however, incident photons generate excitons of which separation by the drain bias gives rise to a photocurrent. The application of a positive gate bias increases the charge carrier density by downshifting conduction and valence bands (as indicated by the blue line) and induces a relatively high transconductance in WS$_2$, which results in higher mobility (as indicated by the blue dotted arrow). The high mobility in the WS$_2$ channel promotes the collection of excited electrons and yields a higher photocurrent than is obtained under a negative gate bias (as indicated by the red dotted arrow). The photocurrent generated upon illumination increases as the wavelength is changed from 700 nm, 450 nm to 630 nm (see Fig. 2(b)). The 630 nm photons had an energy of 2.07 eV, and this value is close to the direct band gaps of bulk WS$_2$ (1.98 and 2.25 eV)\textsuperscript{37,38}. An incident photon with an energy comparable to the direct band gap of WS$_2$ (as indicated by the green arrow in Fig. 4(d)) is efficiently absorbed with a low energy loss relative to photons having a larger energy (as indicated by the purple arrow in Fig. 4(d)). Such photons generate electron–hole (e-h) pairs that give rise to a large photocurrent. The incident photons with an energy less than the direct band gap of WS$_2$ are transmitted without generating excitons (see the red arrow in Fig. 4(d)). In this work, we assumed that the photocurrent from a photothermolectric effect\textsuperscript{39} is little altered. The photocurrent generated near source-channel and drain-channel interfaces is canceled each other because the temperature differences between two interfaces are expected to be similar due to global light illumination.\textsuperscript{40}

We found a linear photoresponsivity from the WS$_2$ devices, for illumination powers in the range of 10$^{-5}$–10$^{-1}$ W/cm$^2$ and for wavelengths in the range of 450 (2.76)–700 nm (1.77 eV). We demonstrated very high optoelectronic performances from the WS$_2$ devices: a photoresponsivity of 0.27 A/W, an $\eta_{dark}$/I$_{phot}$ of 10$^{2}$–10$^{3}$, and an external quantum efficiency of $\sim$7000% which are higher than the corresponding values reported for bulk MoS$_2$ phototransistors and the other reported TMDC photosensing devices, even upon application of the much smaller electrical fields (see Table SI for the detailed comparison\textsuperscript{33}).

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FIG. 4. Band diagrams of a WS$_2$ device. (a) Flat band state. (b) Equilibrium state: an Ohmic junction is formed. (c) Biased state: the photocurrent generation mechanism depending on gate bias polarity and light illumination is depicted. (d) A schematic drawing of the photocurrent generation dependent on photon energy.
33See supplementary material at http://dx.doi.org/10.1063/1.4878335 for gate bias and wavelength dependent transfer curve and photocurrent, photoresponse measurement system setup, calculation of photocurrent density and optical power density, comparison of the figures of merit for photodetectors prepared using different TMDCs, and photoresponsivity calculation.