High photoresponse of individual WS₂ nanowire-nanoflake hybrid materials

Georgies Alene Asres,^{1,a)} Topias Järvinen,^{1,a)} Gabriela S. Lorite,¹ Melinda Mohl,¹ Olli Pitkänen,¹ Aron Dombovari,¹ Geza Tóth,² Anita Lloyd Spetz,^{1,3} Robert Vajtai,⁴ Pulickel M. Ajayan,⁴ Sidong Lei,⁵ Saikat Talapatra,⁶ and Krisztian Kordas^{1,b)}

¹Microelectronics Research Unit, Faculty of Information Technology and Electrical Engineering, University of Oulu, P.O. Box 4500, FI-90014 Oulu, Finland

²VTT Finland, P.O. Box 1100, FI-90571 Oulu, Finland

³Sensor and Actuator Systems, Department of Physics, Chemistry and Biology, Linköping University, SE-58183 Linköping, Sweden

⁴Department of Material Science and NanoEngineering, Rice University, Houston, Texas 77005, USA

⁵Department of Materials Science and Engineering, University of California, Los Angeles, California 90095, USA

⁶Department of Physics, Southern Illinois University, Carbondale, Illinois 62901, USA

(Received 22 March 2018; accepted 21 May 2018; published online 6 June 2018)

van der Waals solids have been recognized as highly photosensitive materials that compete conventional Si and compound semiconductor based devices. While 2-dimensional nanosheets of single and multiple layers and 1-dimensional nanowires of molybdenum and tungsten chalcogenides have been studied, their nanostructured derivatives with complex morphologies are not explored yet. Here, we report on the electrical and photosensitive properties of WS₂ nanowire-nanoflake hybrid materials we developed lately. We probe individual hybrid nanostructured particles along the structure using focused ion beam deposited Pt contacts. Further, we use conductive atomic force microscopy to analyze electrical behavior across the nanostructure in the transverse direction. The electrical measurements are complemented by *in situ* laser beam illumination to explore the photoresponse of the nanohybrids in the visible optical spectrum. Photodetectors with responsivity up to $\sim 0.4 \text{ A W}^{-1}$ are demonstrated outperforming graphene as well as most of the other transition metal dichalcogenide based devices. *Published by AIP Publishing.*

<https://doi.org/10.1063/1.5030490>

Transition metal dichalcogenide (TMD) semiconductors are truly multifunctional materials that have attracted much attention in recent years owing to their fascinating physical properties that can be exploited in solar cells,¹ catalysts,^{2,3} advanced electronics,⁴ electrochemical energy storage,⁵ and gas sensing⁶ applications among many others. While the 2-dimensional (2D) forms of TMD (as single or multilayer) crystals, particularly MoS₂ and MoSe₂, are in the forefront of scientific interest due to their excellent optoelectronic switching behavior as well as large specific surface area and their ability to intercalate small cations, only a few studies report on the applications of 1-dimensional (1D) nanowires or nanotubes.^{7–10}

In this work, we investigate the electrical and photosensitive properties of individual WS₂ nanowire-nanoflake hybrid materials we synthesized and reported lately.¹¹ These hybrids combine both 1D and 2D forms of WS₂ within a single structure in such a way that nanoflakes of multi-layered WS₂ protrude from the body of the WS₂ nanowire. The formation of the hybrid structure is believed to be caused by local shear stresses when sulfurizing WO₃ nanowires that result in peeling of the layered van der Waals solid on the surface.

The bandgap of WS₂ is between 1.4 eV (multilayer as in our case) and 2.1 eV (single layer) which matches the visible

spectrum. These values of the bandgap make these materials sensitive to visible light and near infrared radiation¹² and hence are extremely lucrative from the point of view of developing them for applications related to optoelectronics. Motivated by this, we explore the electrical and optoelectrical properties of the hybrid material using devices based on individual nanohybrid particles. We assess local transport behavior using conductive atomic force probe microscopy that suggests local inhomogeneities of chemical composition in the structure. Further, we study the photoresponse of the nanohybrids in two-terminal measurements through Pt contacts deposited by focused-ion beam. The latter measurements indicate fast and high photoresponse in the entire visible spectrum with nonlinear photocurrent dependence on the light intensity. The highest measured responsivity of $\sim 0.4 \text{ A W}^{-1}$ competes with bulk Si devices and outperforms 2D materials including graphene and TMDs making the WS₂ nanohybrid a promising candidate for fast photodetector devices working at visible wavelengths.¹³

WS₂ nanohybrids were synthesized via sulfurization of WO₃ nanowires as previously reported.¹¹ 12 mg of the WS₂ nanowire-nanoflake hybrid was dispersed in 6 ml acetone by ultrasonic agitation (30 min) and then used by drop-casting $\sim 5 \mu\text{l}$ volume on the test chips. The photodetectors were made as follows. First, Pt contact pads (thickness of 200 nm) were sputtered on Si/SiO₂ chips (laser cut to size of $7 \times 7 \text{ mm}^2$) through a transmission electron microscopy (TEM) grid (50 mesh size) used as a shadow mask. After drop-casting the sparse random networks of the WS₂

^{a)}G. A. Asres and T. Järvinen contributed equally to this work.

^{b)}Author to whom correspondence should be addressed: krisztian.kordas@oulu.fi

nanomaterials on the surface, we connected the tips of the particles to adjacent Pt pads using Pt contact wires of ~ 100 nm diameter by means of focused ion beam-assisted chemical vapor deposition (FIB, Helios NanoLabTM DualBeam 600, 90 pA ion current and 30 kV accelerating voltage). The connections between Pt contact wire and the individual WS₂ nanomaterials were verified by field emission scanning electron microscopy (FESEM, Zeiss Ultra Plus).

Current-voltage (I-V) analysis was performed for each device by probing the sputtered Pt pads using a Wentworth Labs probe station connected to a computer-controlled source meter (Keithley 2636A Dual-channel System SourceMeter[®], Keithley Instruments, Inc.). Photocurrent measurements were performed using laser beams (Coherent[®] High Performance OBISTM Laser System, TEM₀₀ mode, $\lambda = 661$, 552, and 401 nm with corresponding $1/e^2$ beam waist sizes of 9, 7, and 8 mm) modulated with a 10 kHz square-wave by a signal generator (TGA1244, Thurlby Thandar Instruments Ltd.) applied on the chips. A load resistor of 19.6 M Ω was added in series with the device and probed with an oscilloscope (Agilent InfiniiVision DSO-X 3024A, Agilent Technologies, Inc.). The 10 M Ω oscilloscope probe (Agilent N2863B, Agilent Technologies, Inc.) formed a parallel resistor circuit with the load resistor with a net impedance of 6.62 M Ω . The measurement was triggered by the synchronization signal from the signal generator which was used to modulate the laser (Fig. S1). The photocurrent and photoresponsivity of the devices were assessed as a function of laser power (1, 2, 5, 10, and 20 mW at all wavelengths plus 50 mW at 401 nm as well as 50 and 100 mW at 661 nm). The laser power on the device is calculated as $2P \cdot l \cdot d / (\pi \cdot r_o^2)$, where P is the total laser power, l and d are the length and diameter of the nanohybrid, and r_o is the $1/e^2$ beam radius.

Topography and current maps of WS₂ nanohybrids were acquired simultaneously by using atomic force microscopy (AFM, MultiMode 8, Nanoscope V, Bruker, USA) under Peak Force-TUNA (PF-TUNA) modes. The AFM was also operated in the spectroscopy mode to obtain I-V analysis. The measurements were performed by using Pt coated Si tips (NSC18/Pt, MicroMash, Tallin, Estonia) with a radius less than 30 nm. In this case, WS₂ nanohybrid dispersion was drop-cast on gold sputtered (50 nm) Si (111) chips preliminarily mounted with a conductive carbon tape on a metal disk and grounded with silver paste. For the current map, a constant bias of 3 V was applied on the sample. I-V analysis was performed by selecting several points on the surface of the nanohybrids and the voltage was ramped between -1 V and 1 V while the current signal was recorded. The measurements were repeated 5 times in each point and then averaged.

The WS₂ nanowire-nanoflake structures obtained by annealing WO₃ nanowires in sulfur vapor at 800 °C for 30 min are identical to those we reported in our previous works. The hybrid structure forms in such a way that the surface of elongated rod-shaped structures of 1–5 μ m length is decorated by partially peeled nanoflakes of ~ 10 nm thickness and 200–300 nm diameter (Fig. S2).

Individual nanohybrids contacted by focused ion beam deposited Pt leads show linear I-V behavior, indicating that

FIG. 1. Focused ion beam contacted WS₂ nanowire/nanoflake hybrids. (a) Low and high magnification SEM images of a nanowire contacted by Pt leads on a Si/SiO₂ chip. (b) Current-voltage characteristics of 5 devices and the corresponding band diagram (inset) displaying ohmic behavior. Reproduced with permission from Acta Univ. Oul. C 652 (2018). Copyright 2018 University of Oulu.

the metal-semiconductor interface formed perhaps has a very negligible Schottky barrier and is near ohmic (Fig. 1). Such a behavior is reasonable according to the equilibrium band diagram [inset in Fig. 1(b)] considering the work function (~ 5.6 eV) of Pt as well as the electron affinity (~ 4.0 eV),^{14,15} work function (~ 5.1 eV),^{16–19} and bandgap (~ 1.4 eV) of the WS₂ nanowire-nanoflake hybrid having p-type semiconducting behavior.

The large (more than 2 orders of magnitude) variation of conductance measured for devices with similar geometries suggests considerable chemical and structural inhomogeneity in the hybrid materials. Variation in their structure is reasonable, since the nanoflakes are not uniformly spread on the surface of nanowires, which is also evident from the SEM and TEM images as shown in Figs. 1 and S2. On the other hand, chemical inhomogeneity within the individual nanowire-nanoflake hybrids (and among different particles) may be explained by the inhomogeneous sulfurization (9.3 at. % O and 41.4 at. % S content in the form of WO₃ and WS₂, respectively) and thus the statistical distribution of O and S rich regions within the structures.²⁰ For instance, the nanoflakes are expected to be dominantly sulfides because of the formation mechanism of the structure, which is associated with the local stress which leads to the subsequent delamination of WS₂ layers after O is substituted by S and the 3D hexagonal WO₃ lattice is transformed to a layered 2D structure. Accordingly, the 1D core of the nanoparticle shall be somewhat enriched in O in the anionic sites of the lattice.

Both chemical and structural inhomogeneities seem to be in agreement with conductive AFM mapping, which reveals that the local conductance of the hybrid structure has large variation (Fig. 2). In the locations where the WS₂ nanoflakes protrude from the wire, the local current may be even three decades higher than at other parts of the structure. I-V curves collected from different locations of the nanowires show slightly nonlinear behavior indicating the presence of barrier(s) across the specimen, i.e., the Pt tip, WS₂ nanohybrid, and Au substrate. While both Pt and Au form ohmic contacts with WS₂ (as we have also observed for robust Pt contact leads) due to their high work functions with respect to the electron affinity of WS₂, the picture may be different when considering a nano-sized contact area the tip forms with the surface. Any chemical irregularity (e.g., localized oxygen-rich regions in the crystal) caused by imperfect sulfurization (oxygen to sulfur atomic ratio of ~ 0.22 measured by XPS for the anionic sites) can result in junctions that deviate from the ohmic Pt-WS₂ interface (Fig. S3). Although

FIG. 2. Conductive atomic force microscopy probed WS₂ nanowire/nanoflake hybrids. (a) Current-voltage characteristics measured in 5 locations of a WS₂ hybrid nanostructure on the Au surface (as shown in the AFM topography scan in the inset) displaying slightly asymmetric nonlinear behavior. (b) Topography and (c) conductive probe mode imaging of a hybrid nanostructure measured on the Au coated surface at a bias of +3 V. Reproduced with permission from Acta Univ. Oul. C 652 (2018). Copyright 2018 University of Oulu.

the radius of our Pt probe is ≤ 30 nm, the tip contact area through which the current flows is only ~ 1 nm², and thus, the measured current will depend on the local chemical environment.²¹ Furthermore, the tunneling barrier between the conductive probe and the sample can also contribute to the nonlinear I-V behavior and may explain the nearly symmetric shape of the curves.²²

Photoconductivity measurements on individual nanohybrids electrically connected to sputtered Pt probe pads using FIB deposited Pt wires show laser beam intensity dependent modulation of the device current [Figs. 3(a) and S4]. The photocurrent displays rapid onset and decay upon switching on and off the light at each wavelength [Fig. 3(b)]. Both excitation and relaxation curves can be fitted well using bi-exponential growth and decay functions having shorter (~ 1 μ s) and slightly longer (~ 10 μ s) time constant components associated with the RC delays of our electrical setup caused by the distributed resistances of the WS₂ nanohybrid (66.7 M Ω), load resistor (19.6 M Ω), oscilloscope (10 M Ω), and substrate leakage (~ 10 G Ω) as well as by the coplanar Pt probe pad capacitances (~ 50 fF) and parallel plate substrate and Pt pad capacitances (~ 10 pF). The actual photo-generation/relaxation is expected to be faster, having time constants typically in the sub-ns regime as demonstrated by optical pump-and-probe measurements on liquid-phase exfoliated MoS₂ films.²³ The light power dependence of photocurrent [Fig. 3(c)] can be fit using power functions at each measured wavelength having very similar exponents of 0.207 ± 0.015 . Such power law variations are extremely common in a variety of semiconductors, especially disordered semiconducting materials. Typically, it is assumed that such variations come from the presence of mid-gap states, which play a crucial role in photo-carrier relaxation and hence the photocurrent generation. It is worth mentioning here that electrode deposition using FIB processing can lead to a disorder in the crystal in the proximity of the Ga⁺ impact region as it has been shown earlier for Pt contacts on GaN nanowires.^{24,25}

Similar exponents were found by several groups for other low dimensional or nanostructured photosensitive materials such as graphene (~ 0.25),²⁶ ZnO (~ 0.28),²⁷ single layer WS₂ (0.4),²⁸ and InSb nanowires (0.2)²⁹ although also higher exponent values were reported, e.g., for sputtered but thermally annealed films of WS₂ (0.79)³⁰ as well as for WS₂ nanotubes grown at high temperatures from WO₃ particles in H₂S (~ 1)³¹ probably due to better crystallinity of the latter

FIG. 3. Photoconductivity of individual WS₂ nanohybrids. (a) Current modulation by red laser pulses ($\lambda = 661$ nm, $w_0 = 9$ mm $1/e^2$ beam waist, TEM₀₀, 10 kHz square wave with a duration of 50 μ s) at a 10 V source measured by an oscilloscope on a serially connected load resistor. The total laser power on the chip was adjusted to 1, 2, 5, 10, 20, 50, and 100 mW. (b) Photoresponse to laser pulses ($\lambda = 661$, 552, and 401 nm denoted as red, green, and blue plots) of 20 mW total power corresponding to 29, 52, and 40 nW power on the WS₂ nanohybrid considering the $1/e^2$ beam waists of 9, 7, and 8 mm, respectively (nanohybrid length and diameter of 5 μ m and 330 nm, respectively). The time constants correspond to the fitting parameters of bi-exponential growth and decay fitting parameters. (c) Log-log plot of photocurrent (i.e., $I_{\text{light}} - I_{\text{dark}}$) as a function of total laser power and wavelength, and corresponding power function fittings. (d) Responsivity calculated as the ratio of photocurrent (i.e., $I_{\text{light}} - I_{\text{dark}}$) and light power on the device [i.e., $2P \cdot l \cdot d / (\pi \cdot r_o^2)$, where P is the total laser power, l and d are the length and diameter of the nanohybrid, and r_o is the $1/e^2$ beam radius]. Reproduced with permission from Acta Univ. Oul. C 652 (2018). Copyright 2018 University of Oulu.

two. The power exponents associated with the power law variation of photocurrent with light intensity can provide some insight into the photocarrier generation/recombination process in semiconductors. For example, a power exponent value of unity is associated with monomolecular recombination processes, whereas a value of 0.5 is due to bimolecular recombination processes. Studies have also suggested that this power exponent value will also depend on the distribution of the trap states within the bandgap. For example, it is generally assumed that an exponential distribution of trap states leads to a power exponent value ranging between 0.5 and 1.0. As observed, the value of the exponent obtained in our case is even lower than that observed for bimolecular processes (0.5). Such behavior can have several explanations, for example, if during initial stages of the measurements the samples are sensitized by the low light intensity (which makes the trap distribution within the midgap state extremely complex), then there is a possibility that the photocurrent versus intensity curve will have a power dependence less than 0.5.³² Another plausible explanation could be that the low power exponent values measured on these devices are manifestation of the photogating effect, i.e., substrate generated photovoltage that can act as electrical gate on the measured device.³³ Such mechanisms are recently demonstrated in other 2D material based FETs.³⁴

From our measurements, we find that the light responsivity of our WS₂ hybrid is practically uniform within the

FIG. 4. Finite element modeling of the chip surface temperature illuminated with a laser pulse of (a) 50 μ s and (b) 1 s duration. (c) Evolution of the surface temperature in the center of the Gaussian laser beam as a function of time. (d) Reproduced with permission from Acta Univ. Oul. C 652 (2018). Copyright 2018 University of Oulu. Resistance vs. temperature of the WS_2 nanowire-nanoflake hybrid measured for percolated thin film networks.¹¹

visible spectral range [Fig. 3(d)], which suggests that the photogeneration and also the relaxation of carriers are mediated by trap states in the semiconductor. The maximum photoresponsivity value is $\sim 0.4 \text{ A} \cdot \text{W}^{-1}$ being similar to those of bulk Si based detectors³⁵ and composites of carbon nanotube and perovskite films,³⁶ clearly outperforming 2D graphene,^{37,38} MoS_2 ,^{39,40} and WS_2 ,⁴¹ however much lower than those reported for CdSe and CdSe/CdS core-shell nanocrystals⁴² and arrays of ZnO⁴³ and Si⁴⁴ nanowires in phototransistors. The calculated corresponding external quantum efficiencies for 661, 552, and 401 nm are 76%, 88%, and 124%, respectively.

Adsorbed illumination with the laser beams causes an inherent heating of the materials and consequently results in a thermal activation and increased electrical conductivity of the semiconductor. The thermal coefficient of resistance at room temperature for thin networks of the WS_2 nanowire-nanoflake hybrids is relatively high,¹¹ having a value of $\sim -0.04 \text{ deg}^{-1}$ [Fig. 4(d)], meaning that the relative change of $\sim 1\%$ in the photocurrents (or the resistance) that we measure may be induced even with a very moderate heating ($\Delta T \sim 0.25 \text{ }^\circ\text{C}$) of the nanomaterial. Accordingly, to validate our photoresponse data, we need to evaluate the rise of local temperature in the proximity of the nanowire-nanoflake hybrid. For this, we use finite-element modeling and calculate the surface temperature of the chip in the position of the photodetector nanohybrid particle. The results show that illumination with the short laser pulses we apply in our experiments (50 μ s) causes only minor heating, e.g., $\Delta T \sim 0.013 \text{ }^\circ\text{C}$ at 5 mW [Figs. 4(a) and 4(c)]; thus, the thermal effects may be neglected and the measured improvement of electrical conductivity is truly caused by the photogeneration.⁴⁵ However, upon longer laser pulses or with continuous wave beams, even such a low power unfocused ($1/e^2$ beam radius of 0.45 mm) can induce considerable surface heating ($\Delta T \sim 0.165 \text{ }^\circ\text{C}$), indicating the necessity of pulsed optical signals for reliable photocurrent measurements.

In conclusion, we have shown that the WS_2 nanowire-nanoflake hybrid materials grown by the sulfurization of

WO_3 nanowires show a high photoresponsivity of $\sim 0.4 \text{ A} \cdot \text{W}^{-1}$ with corresponding external quantum efficiencies of 76%, 88%, and 124% at 661, 552, and 401 nm, respectively. The light sensitivity of individual WS_2 nanowire-nanoflake hybrid particle is as good as that of bulk Si photodetectors or recently reported CNT-perovskite composites and clearly outperforms 2D single-layer based devices of TMDs or graphene. The results reported here suggest the exploration of other TMD nanohybrids that presumably can be synthesized in a similar fashion, with great potential to be used in photodetector, photoelectrode, and photocatalyst applications.

See [supplementary material](#) for the experimental setup of light response measurements, additional TEM images of WS_2 nanowire-nanoflake hybrids, band diagrams of Pt- WO_3 , Pt- WS_2 , and Au- WS_2 interfaces, and light response to green (551 nm) and purple (401) laser pulses.

Funding received from the Academy of Finland (Suplacat), University of Oulu (More than Moore Research Community), and University of Oulu Graduate School (Infotech Oulu) is acknowledged. We also would like to thank Sami Saukko and Esa Heinonen (Center of Microscopy and Nanotechnology, University of Oulu) for their assistance with TEM analyses and FIB processing, respectively.

- ¹M. Shanmugam, C. A. Durcan, and B. Yu, "Layered semiconductor molybdenum disulfide nanomembrane based Schottky-barrier solar cells," *Nanoscale* **4**, 7399–7405 (2012).
- ²D. Chu, C. Zhang, P. Yang, Y. Du, and C. Lu, " WS_2 as an effective noble-metal free cocatalyst modified TiSi_2 for enhanced photocatalytic hydrogen evolution under visible light irradiation," *Catalysts* **6**, 136 (2016).
- ³J.-F. Lin, O. Pitkanen, J. Maklin, R. Puskas, A. Kukovecz, A. Dombovari, G. Toth, and K. Kordas, "Synthesis of tungsten carbide and tungsten disulfide on vertically aligned multi-walled carbon nanotube forests and their application as non-Pt electrocatalysts for the hydrogen evolution reaction," *J. Mater. Chem. A* **3**, 14609–14616 (2015).
- ⁴R. Levi, O. Bitton, G. Leitun, R. Tenne, and E. Joselevich, "Field-effect transistors based on WS_2 nanotubes with high current-carrying capacity," *Nano Lett.* **13**, 3736–3741 (2013).
- ⁵C. C. Mayorga-Martinez, J. G. S. Moo, B. Khezri, P. Song, A. C. Fisher, Z. Sofer, and M. Pumera, "Self-propelled supercapacitors for on-demand circuit configuration based on WS_2 nanoparticles micromachines," *Adv. Funct. Mater.* **26**, 6662–6667 (2016).
- ⁶D. J. Late, R. V. Kanawade, P. K. Kannan, and C. S. Rout, "Atomically thin WS_2 nanosheets based gas sensor," *Sens. Lett.* **14**, 1249–1254 (2016).
- ⁷Y. Liu, N. Zhang, H. Kang, M. Shang, L. Jiao, and J. Chen, " WS_2 Nanowires as a high-performance anode for sodium-ion batteries," *Chem. - Eur. J.* **21**, 11878–11884 (2015).
- ⁸R. Yuvasravan, G. Apsana, P. P. George, I. Genish, Y. Koltypin, and A. Gedanken, "Synthesis of WS_2 and WSe_2 nanowires on stainless steel coupon by reaction under autogenic pressure at elevated temperature method," *Appl. Nanosci.* **6**, 855–862 (2016).
- ⁹C. Zhang, Z. Ning, Y. Liu, T. Xu, Y. Guo, A. Zak, Z. Zhang, S. Wang, R. Tenne, and Q. Chen, "Electrical transport properties of individual WS_2 nanotubes and their dependence on water and oxygen absorption," *Appl. Phys. Lett.* **101**, 113112 (2012).
- ¹⁰C. Y. Zhang, S. Wang, L. J. Yang, Y. Liu, T. T. Xu, Z. Y. Ning, A. Zak, Z. Y. Zhang, R. Tenne, and Q. Chen, "High-performance photodetectors for visible and near-infrared lights based on individual WS_2 nanotubes," *Appl. Phys. Lett.* **100**, 243101 (2012).
- ¹¹G. A. Asres, A. Dombovari, T. Sipola, R. Puskas, A. Kukovecz, Z. Konya, A. Popov, J.-F. Lin, G. S. Lorite, M. Mohl, G. Toth, A. Lloyd Spetz, and K. Kordas, "A novel WS_2 nanowire-nanoflake hybrid material synthesized from WO_3 nanowires in sulfur vapor," *Sci. Rep.* **6**, 25610 (2016).

- ¹²F. H. L. Koppens, T. Mueller, P. Avouris, A. C. Ferrari, M. S. Vitiello, and M. Polini, "Photodetectors based on graphene, other two-dimensional materials and hybrid systems," *Nat. Nanotechnol.* **9**, 780–793 (2014).
- ¹³G. A. Asres, Synthesis, characterization and application of WS₂ nanowire-nanoflake hybrid nanostructures. Doctoral thesis, University of Oulu, Acta Universitatis Ouluensis, C 652, 2018.
- ¹⁴T. Yamaguchi, R. Moriya, Y. Inoue, S. Morikawa, S. Masubuchi, K. Watanabe, T. Taniguchi, and T. Machida, "Tunneling transport in a few monolayer-thick WS₂/graphene heterojunction," *Appl. Phys. Lett.* **105**, 223109 (2014).
- ¹⁵M. W. Iqbal, M. Z. Iqbal, M. F. Khan, M. A. Shehzad, Y. Seo, J. H. Park, C. Hwang, and J. Eom, "High-mobility and air-stable single-layer WS₂ field-effect transistors sandwiched between chemical vapor deposition-grown hexagonal BN films," *Sci. Rep.* **5**, 10699 (2015).
- ¹⁶S. H. Lee, D. Lee, W. S. Hwang, E. Hwang, D. Jena, and W. J. Yoo, "High-performance photocurrent generation from two-dimensional WS₂ field-effect transistors," *Appl. Phys. Lett.* **104**, 193113 (2014).
- ¹⁷J. Kim, S. Byun, A. J. Smith, J. Yu, and J. Huang, "Enhanced electrocatalytic properties of transition-metal dichalcogenides sheets by spontaneous gold nanoparticle decoration," *J. Phys. Chem. Lett.* **4**, 1227–1232 (2013).
- ¹⁸S. Fiechter, "Defect formation energies and homogeneity ranges of rock salt-, pyrite-, chalcopyrite- and molybdenite-type compound semiconductors," *Sol. Energy Mater. Sol. Cells* **83**, 459–477 (2004).
- ¹⁹M. Shanmugam, T. Bansal, C. A. Durcan, and B. Yu, "Schottky-barrier solar cell based on layered semiconductor tungsten disulfide nanofilm," *Appl. Phys. Lett.* **101**, 263902 (2012).
- ²⁰G. A. Asres, J. J. Baldoví, A. Dombóvari, T. Järvinen, G. S. Lorite, M. Mohl, A. Shchukarev, A. P. Paz, L. Xian, J.-P. Mikkola, A. Lloyd Spetz, H. Jantunen, A. Rubio, and K. Kordas, "Ultra-sensitive H₂S gas sensors based on p-type WS₂ hybrid materials," *Nano Res.* (2018).
- ²¹C. Rodenbücher, G. Bihlmayer, W. Speier, J. Kubacki, M. Wojtyniak, M. Rogala, D. Wrana, F. Krok, and K. Szot, "Detection of confined current paths on oxide surfaces by local-conductivity atomic force microscopy with atomic resolution," e-print [arXiv:1611.07773](https://arxiv.org/abs/1611.07773).
- ²²X. Zhu, S. Lei, S.-H. Tsai, X. Zhang, J. Liu, G. Yin, M. Tang, C. M. Torres, Jr., A. Navabi, Z. Jin, S. P. Tsai, H. Qasem, Y. Wang, R. Vajtai, R. K. Lake, P. M. Ajayan, and K. L. Wang, "A study of vertical transport through graphene toward control of quantum tunneling," *Nano Lett.* **18**(2), 682–688 (2018).
- ²³S. Ghosh, A. Winchester, B. Muchharla, M. Wasala, S. Feng, A. L. Elias, M. B. M. Krishna, T. Harada, C. Chin, K. Dani, S. Kar, M. Terrones, and S. Talapatra, "Ultrafast intrinsic photoresponse and direct evidence of sub-gap states in liquid phase exfoliated MoS₂ thin films," *Sci. Rep.* **15**, 11272 (2015).
- ²⁴C. Y. Nam, D. Tham, and J. E. Fischer, "Disorder effects in focused-ion-beam-deposited Pt contacts on GaN nanowires," *Nano Lett.* **5**, 2029–2033 (2005).
- ²⁵D. Tham, C. Y. Nam, and J. E. Fischer, "Microstructure and composition of focused-ion-beam-deposited Pt contacts to GaN nanowires," *Adv. Mater.* **18**, 290–294 (2006).
- ²⁶X. Sun, C. Qiu, J. Wu, H. Zhou, T. Pan, J. Mao, X. Yin, R. Liu, W. Gao, Z. Fang, and Y. Su, "Broadband photodetection in a microfiber-graphene device," *Opt. Express* **23**, 25209 (2015).
- ²⁷S. I. Inamdar, V. V. Ganbavle, and K. Y. Rajpure, "ZnO based visible-blind UV photodetector by spray pyrolysis," *Superlattices Microstruct.* **76**, 253–263 (2014).
- ²⁸C. Lan, C. Li, Y. Yin, and Y. Liu, "Large-area synthesis of monolayer WS₂ and its ambient-sensitive photo-detecting performance," *Nanoscale* **7**, 5974–5980 (2015).
- ²⁹C.-H. Kuo, J.-M. Wu, S.-J. Lin, and W.-C. Chang, "High sensitivity of middle-wavelength infrared photodetectors based on an individual InSb nanowire," *Nanoscale Res. Lett.* **8**, 327 (2013).
- ³⁰L. Zeng, L. Tao, C. Tang, B. Zhou, H. Long, Y. Chai, S. P. Lau, and Y. H. Tsang, "High-responsivity UV-vis photodetector based on transferable WS₂ film deposited by magnetron sputtering," *Sci. Rep.* **6**, 20343 (2016).
- ³¹Y. J. Zhang, M. Onga, F. Qin, W. Shi, A. Zak, R. Tenna, J. Smet, and Y. Iwasa, "Optoelectronic response of a WS₂ tubular p-n junction," *2D Mater.* **5**, 035002 (2018).
- ³²R. H. Bube, *Photoelectronics Properties of Semiconductors* (Cambridge University Press, 1992).
- ³³M. S. Marcus, J. M. Simmons, and O. M. Castellini, "Photogating carbon nanotube transistors," *J. Appl. Phys.* **100**, 084306 (2006).
- ³⁴J. O. Island, S. I. Blanter, M. Buscema, H. S. J. van der Zant, and A. Castellanos-Gomez, "Gate controlled photocurrent generation mechanisms in high-gain In₂Se₃ phototransistors," *Nano Lett.* **15**, 7853–7858 (2015).
- ³⁵See https://www.hamamatsu.com/resources/pdf/ssd/e02_handbook_si_photodiode.pdf for the photoresponsivity of bulk Si based detectors.
- ³⁶T. Ka, L. F. Gerlein, R. Nechache, and S. G. Cloutier, "High-performance nanotube-enhanced perovskite photodetectors," *Sci. Rep.* **7**, 45543 (2017).
- ³⁷F. Xia, T. Mueller, Y. Lin, A. Valdes-Garcia, and P. Avouris, "Ultrafast graphene photodetector," *Nat. Nanotechnol.* **4**, 839–843 (2009).
- ³⁸F. Xia, T. Mueller, R. Golizadeh-Mojarad, M. Freitag, Y. Lin, J. Tsang, V. Perebeinos, and P. Avouris, "Photocurrent imaging and efficient photon detection in a graphene transistor," *Nano Lett.* **9**, 1039–1044 (2009).
- ³⁹Z. Yin, H. Li, L. Jiang, Y. Shi, Y. Sun, G. Lu, Q. Zhang, X. Chen, and H. Zhang, "Single-layer MoS₂ phototransistors," *ACS Nano* **6**, 74–80 (2012).
- ⁴⁰H. S. Lee, S.-W. Min, Y.-G. Chang, M. K. Park, T. Nam, H. Kim, J. H. Kim, S. Ryu, and S. Im, "MoS₂ nanosheet phototransistors with thickness-modulated optical energy gap," *Nano Lett.* **12**, 3695–3700 (2012).
- ⁴¹N. Perea-López, A. L. Elías, A. Berkdemir, A. Castro-Beltrán, H. R. Gutiérrez, S. Feng, R. Lv, T. Hayashi, F. López-Urías, S. Ghosh, B. Muchharla, S. Talapatra, H. Terrones, and M. Terrones, "Photosensor device based on few-layered WS₂ films," *Adv. Funct. Mater.* **23**, 5511–5517 (2013).
- ⁴²H. Chen, H. Liu, Z. Zhang, K. Hu, and X. Fang, "Nanostructured photodetectors: From ultraviolet to terahertz," *Adv. Mater.* **28**, 403–433 (2016).
- ⁴³W. Y. Weng, S. J. Chang, C. L. Hsu, and T. J. Hsueh, "A, ZnO-nanowire phototransistor prepared on glass substrates," *ACS Appl. Mater. Interfaces* **3**, 162–166 (2011).
- ⁴⁴A. Zhang, H. Kim, J. Cheng, and Y. H. Lo, "Ultrahigh responsivity visible and infrared detection using silicon nanowire phototransistors," *Nano Lett.* **10**, 2117–2120 (2010).
- ⁴⁵Note that our model slightly overestimates the surface temperature for two reasons. First, the surface reflection of the laser beam is neglected. Second, the temperature is calculated for the chip surface, and since nanowire-nanoflake hybrid is not in a seamless contact with the surface, the thermal interface between those is not ideal.