

# Harnessing plasmon-exciton synergy in multilayer WS<sub>2</sub> based self-powered blue photodetector for high-visibility underwater imaging

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# Harnessing plasmon-exciton synergy in multilayer WS<sub>2</sub> based self-powered blue photodetector for high-visibility underwater imaging

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**ABSTRACT:** Blue photodetectors are critical components for underwater wireless optical communication, high-resolution bioimaging, and environmental monitoring. However, there remains a need for more practical and energy-saving devices with facile synthesis and stable materials. Herein, we developed a blue photodetector featuring a unilateral Schottky junction on multilayer WS<sub>2</sub> decorated with Ag nanoparticles (Ag NPs). The synergistic cooperation of high-energy exciton from WS<sub>2</sub> and localized surface plasmon resonance (LSPR) induced by Ag NPs enhance responsivity and external quantum efficiency up to 244 mA/W and 74.7% under 405 nm with zero external bias, respectively. Ultrafast transient absorption spectroscopy directly reveals the LSPR effect significantly enhances the generation of high-energy excitons, providing a microscopic mechanism for the improvement. Critically, this device enables 128 × 128 resolution blue-light single-pixel imaging in both clear and turbid water, achieving a high peak signal-to-noise ratio (PSNR) of 25.5 dB. This work provides a new strategy for developing the self-powered underwater optical communication and high-visibility imaging technologies.

**KEYWORDS:** blue photodetector, multilayer WS<sub>2</sub>, exciton, plasmon, self-powered, underwater imaging

## 1 Introduction

Blue-light photodetectors have garnered significant research interest due to their critical applications in underwater optical communication [1–3], subsea positioning [4], and 3D scanning technologies [5]. Particularly, blue light, characterized by the smallest absorption coefficient in seawater, has emerged as a promising carrier for next-generation underwater wireless optical communication systems, offering advantages of high data rates and low propagation latency [6–8]. Concurrently, the demand for precise detection in specialized scenarios such as environmental light monitoring and high-resolution imaging continues to drive advancements in this field [9–14]. Nevertheless, the difficulty of underwater energy supply renders traditional photodetectors that rely on an external bias voltage disadvantaged in the field of underwater detection [15–17]. To enable low-power-consumption and portable applications, the development of self-powered photodetectors has become a pivotal research direction [18, 19]. Recent progress has demonstrated various strategies to achieve efficient self-powered blue-light detection. Yang *et al.* constructed a high-work-function transparent electrode by coating silver nanowires with nickel, thereby forming a high Schottky barrier with SrTiO<sub>3</sub> which enabled the realization of high-performance self-powered short-wavelength blue light photodetection functionality [20]. Zhang *et al.* selected Rubrene and C<sub>60</sub> as the donor-acceptor pair and successfully constructed a high-performance blue

narrowband organic heterojunction photodetector through MoO<sub>3</sub> doping and energy-level engineering [21]. Yun *et al.* significantly improved the quality and carrier transport performance of wide-bandgap perovskite films through A-site cation composition engineering (optimizing FA/MA ratio and introducing Cs<sup>+</sup>), thereby realizing a self-powered blue photodetector with an external quantum efficiency of 84.9% [22]. These approaches have successfully realized self-powered operation with notable responsivity (R) and external quantum efficiency (EQE). However, further exploration of richer physical mechanisms is still required to develop high-efficiency blue photodetectors and enable their application at the underwater device.

Two-dimensional (2D) transition metal sulfides (TMDs), with their atomic-scale thickness, tunable bandgaps, mechanical flexibility, and superior environmental stability, offer an attractive platform for next-generation optoelectronics [23–32]. The inherent advantages of van der Waals structures facilitate band engineering and enable the construction of self-powered devices with ultralow dark currents under zero bias [33–36]. TMDs exhibit strong light absorption within the spectral range (blue light) corresponding to the band nesting region (also called as C-exciton) of the Brillouin zone [37,38]. This high optical absorption can be attributed to the intrinsic van Hove singularities in the joint density of states, where the conduction and valence bands are nearly parallel across the C-exciton region of the Brillouin zone. Furthermore, the localized surface

plasmon resonance (LSPR) effect has been utilized to generate additional charge carriers in TMDs, achieving wavelength-selective absorption by adjusting the dimensions of metallic nanostructures such as Au, Ag, and Cu. LSPR effect enhances light absorption in the photoactive layer by enhancing the local electromagnetic field and enabling hot-electron injection, thus offering a viable route to boost photoresponse [39–45].

In this work, we fabricated a self-powered plasmonic photodetector based on WS<sub>2</sub>:Ag nanocomposites featuring Au/graphene (Gr) electrode. The LSPR absorption property of Ag NPs and Schottky barrier of the asymmetric electrode enable the formation of a competitive blue-light photodetector under zero external bias. Experimental characterization and theoretical simulations confirmed the generation capability of high-energy excitons is significantly enhanced under the synergistic effect of near-field enhancement and hot-electron injection. Transient absorption (TA) spectroscopy was employed to reveal the carrier dynamics within the WS<sub>2</sub> layer, showing a unilateral built-in electric field facilitated rapid carrier separation. As a result, the device delivers a zero-bias responsivity of 244 mA/W and an external quantum efficiency of 74.7%, representing one of the best-performing self-powered blue photodetectors to date, along with remarkable stability. Leveraging this self-powered WS<sub>2</sub>:Ag device, we further demonstrated single-pixel blue-light imaging in two underwater environments and achieved a high peak signal-to-noise ratio (PSNR) of 25.5 dB, highlighting its practical potential for low-power practical optical communication and sensing systems.

## 2 Results and discussion

Optical waves, particularly those in the blue band, exhibit substantially lower attenuation in water compared to acoustic communication, which suffers from inherent limitations such as narrow bandwidth [46], high power consumption, and severe attenuation of radio-frequency signals [47–49]. As a result, blue light has emerged as the core enabler of underwater wireless optical communication and imaging [50–52]. It is progressively becoming the key information carrier for underwater optical networks, playing an increasingly vital role in marine resource exploration, environmental monitoring and underwater communication [53–58], as depicted in Fig. 1(a). The transmission advantage of the blue band in underwater environments is attributed to the wavelength-dependent attenuation characteristics of light in water in Fig. 1(b). Within the visible spectrum, various water types exhibit a low absorption coefficient for blue light, creating a natural “optical transmission window” for underwater light propagation. Thus, blue light is an ideal information carrier for high-performance underwater optical detection, and developing high-sensitivity photodetectors for the blue spectrum is crucial to advancing practical applications in underwater imaging and optical communication.

However, the responsivity of current blue-light photodetectors still requires further improvement. The band nesting structure in two-dimensional TMDs enables transitions between Van Hove singularity peaks, thereby significantly enhancing light absorption [59,60]. Among them, WS<sub>2</sub>, as a typical 2D transition metal dichalcogenide, possesses a large intrinsic bandgap that allows for response to the blue light band [61]. As shown in Fig. 1(c), the DFT-calculated band structure

of WS<sub>2</sub> reveals a band nesting region (C-exciton) within the blue-light energy range, where the conduction and valence bands are nearly parallel across the Brillouin zone. This leads to a substantially enhanced joint density of states, endowing WS<sub>2</sub> with intrinsically strong blue-light absorption capability. Fig. 1(d) presents the density of states (DOS) of WS<sub>2</sub>, which exhibits a prominent peak at approximately 3.1 eV, further confirming the high response potential of WS<sub>2</sub> to blue light from the perspective of electronic states. The absorption spectrum of WS<sub>2</sub> in Fig. 1(e) also exhibits a peak around 3.1 eV, which is consistent with the calculated results. The LSPR effect, which enhances the local electromagnetic field and facilitates hot-electron injection, has been demonstrated as an effective strategy to overcome the intrinsic absorption limitations of materials and significantly boost photodetector performance [61–70]. Based on above discussions, WS<sub>2</sub> was selected as the light-absorption layer in this study, and Ag nanoparticles (NPs) were introduced to achieve LSPR effect. By optimizing the size of Ag NPs, the LSPR resonance peak was adjusted to ~3.09 eV (the inset of Fig. 1(e)), achieving resonance with the blue-light band. In summary, the synergistic combination of intrinsic strong blue-light absorption of WS<sub>2</sub> and the plasmonic enhancement provided by Ag NPs offers a viable material design strategy for constructing high-performance self-powered blue-light detectors tailored for underwater applications.

Fig. 2(a) shows a schematic diagram of the Au/WS<sub>2</sub>:Ag/Gr blue light photodetector, and the schematic of the device fabrication process is provided in Fig. S1. The mechanical exfoliation method minimizes defects in the intrinsic material, and the dry transfer technique ensures a clean interface and avoids possible Fermi level pinning. Since excessively thin WS<sub>2</sub> leads to a significant decrease in light absorption, while excessively thick WS<sub>2</sub> increases the carrier transport distance and the probability of carrier recombination and relaxation, it is necessary to determine the optimal thickness of the channel material. Combining atomic force microscopy (AFM) imaging with optoelectronic performance tests, it is found that using WS<sub>2</sub> with a thickness of approximately 130 nm is the optimal choice for achieving high device performance, as shown in Fig. S2(a). The thickness of the selected WS<sub>2</sub> is about 136 nm (Fig. S2(b)) and the uniform color in the scanning image indicates a flat and clean material surface. The thickness of Gr is about 70 nm (Fig. S2(c)). Fig. 2(b) presents high-resolution transmission electron microscopy (HRTEM) images of WS<sub>2</sub> flake and Ag NPs, confirming high-quality interfacial integrity and good dispersion of Ag NPs. Moreover, the X-ray Photoelectron Spectroscopy and Raman spectra of WS<sub>2</sub> with and without Ag NPs presented in Fig. S3 and Fig. S4 confirm that the introduction of Ag NPs. Furthermore, the two materials exhibit good interfacial compatibility, which is a key factor ensuring device stability. To optimize the device performance, we investigated the effect of quantum dot concentration by varying the droplet number (Fig. S5). The optimal responsivity was observed at a concentration of 0.4 mg/ml, and this condition was used for all subsequent measurements. To further understand the plasmonic enhancement mechanism in photodetection, finite-difference time-domain (FDTD) simulations were performed to calculate the electric field distribution around Ag nanoparticles under 405 nm incident light [71–74]. As shown in Fig. 2(c), a strong electromagnetic field along the Ag/WS<sub>2</sub> interface (the diameter

of Ag NP is 5 nm) is produced. The simulated electric field distribution reveals that light is effectively enhanced and confined in the vicinity of the nanoparticle array. The wavelength-dependent photocurrent enhancement, which reaches a maximum at ~400 nm and coincides with the LSPR peak of Ag NPs, is provided in Fig. S6 to directly verify the plasmonic contribution. This result, together with the FDTD simulations, unambiguously confirms that the LSPR effect of Ag NPs is responsible for the enhanced photoresponse.

Fig. 2(d) illustrates the output (I-V) characteristics of the photodetector under dark conditions and under 405 nm illumination with incident power densities ranging from 191 mW/cm<sup>2</sup> to 12739 mW/cm<sup>2</sup>. The I- V characteristics under illumination exhibit pronounced rectifying behavior. Under forward bias, the device is turned on, whereas under reverse bias, the applied voltage expands the built-in electric field, thereby keeping the device in the off state. Benefiting from the constructed unilateral Schottky junction that effectively suppresses the reverse current, the device exhibits an ultralow dark current down to 0.226 pA and yields an  $I_{on}/I_{off}$  ratio of  $5.6 \times 10^4$  at zero bias. Fig. S7 presents the photocurrent curves of the WS<sub>2</sub>/Ag blue light photodetector measured under different incident powers, confirming that the detector exhibits a favorable photoresponse to blue light across all tested power levels. The rise and fall times, extracted from the 10% to 90% levels during the rising and falling edges, are 650  $\mu$ s and 581  $\mu$ s, respectively, indicating a fast response of the detector to blue light. Subsequently, a quantitative evaluation of the self-powered capability was conducted by extracting and plotting the power-dependent short-circuit current ( $I_{sc}$ ) and open-circuit voltage ( $V_{oc}$ ) in Fig. 2(e). Both  $I_{sc}$  and  $V_{oc}$  increase with rising power density. The photocurrent ( $I_{ph}$ ) exhibits a nearly linear dependence on power density and can be fitted by the power-law relationship  $I_{ph} \propto P^\alpha$  [34,75], where  $P$  represents the incident power density and  $\alpha$  is the power-law exponent. Under 405 nm illumination,  $\alpha$  is determined to be 0.85, indicating the photogating effect in the device [76–79]. Additionally, the linear dynamic range (LDR) of the device was calculated using the formula  $LDR = 20 \lg(I_{max}/I_{min})$  [80,81], yielding a value of approximately 32 dB, where  $I_{max}$  and  $I_{min}$  represent the upper and lower limits of the photocurrent linear range, respectively. Under 405 nm illumination with a power density of 12.7 W/cm<sup>2</sup>, a  $V_{oc}$  of 0.16 V and a  $I_{sc}$  of 12.7 nA were obtained. The maximum electrical power ( $P_{max}$ ), fill factor (FF), and power conversion efficiency (PCE) of the photodetector were calculated to be 0.76 nW, 0.76, and 0.37%, respectively, confirming the photovoltaic nature of the Schottky junction. To further characterize the photodetection properties of the device, three key performance parameters were extracted in Fig. S8 to further evaluate the detection capability: responsivity (R), specific detectivity ( $D^*$ ), and external quantum efficiency (EQE). The R and  $D^*$  values reach 244 mA W<sup>-1</sup> and  $5.2 \times 10^{11}$  Jones, respectively, while EQE attains 74.6%. The high EQE indicates the excellent capability to convert incident photons into photocarriers, thereby ensuring robust electrical signal output even under low-light illumination conditions. The R,  $D^*$ , and EQE values of the blue photodetector, along with statistically compiled previously reported blue-light detectors, are summarized in Fig. 2(g) (see Table S1 in Supporting Information for more details). Compared with reported self-

powered blue-light photodetectors, our device exhibits superior EQE values. Furthermore, device performance metrics including NEP, spectral response range, noise current spectrum, long-term stability, -3 dB bandwidth, and high-frequency modulation response have been shown in Fig. S9. Statistical analysis and reproducibility are fundamental to demonstrating the reliability of device performance, which has been shown in Fig. S10. Moreover, to evaluate the operational stability of the photodetector, the photocurrent response was monitored over 800 consecutive cycles in ambient air, during which it remained consistently stable and well-maintained (Fig. S11). Following encapsulation with a PMMA layer, water droplets were applied to the device surface prior to further stability testing. Notably, the device retained a robust and stable photoresponse even after 800 cycles under these conditions. The above results demonstrate that the device holds promising prospects for developing high-performance, highly stable, and low-noise self-powered blue-light photodetectors.

To elucidate the mechanism underlying the enhanced performance of the detector, the energy band alignments of three device configurations were systematically investigated. As shown in Fig. 3(a), in intrinsic WS<sub>2</sub> (denoted as device1), due to the absence of built-in electric field, the hot carriers generated from photoexcited high-energy excitons cannot be efficiently separated under blue light. The carriers undergo recombination during thermal relaxation. In contrast, a strong built-in electric field induced by the unilateral Schottky junction exists at the interface between Au and WS<sub>2</sub>, as depicted in Fig. 3(b). To confirm the Schottky barrier and provide direct evidence for band bending, we performed KPFM measurements, the potential differences at the Gr/WS<sub>2</sub> and Au/WS<sub>2</sub> interfaces were measured using Kelvin probe force microscopy in Fig. S12, confirming the existence of a unilateral Schottky barrier. This built-in potential establishes a strong internal electric field that efficiently drives the separation of photogenerated carriers even at zero external bias, which corresponds to a built-in potential of 0.456 eV (detailed calculation in Note S1, Supporting Information).

Driven by built-in electric field, the high-energy excitons generated in WS<sub>2</sub> within the Au/WS<sub>2</sub>/Gr structure (denoted as device2) can be efficiently separated and transferred, with only a few of those relaxing to the band edge. The corresponding optical images of all devices discussed in the are available in Fig. S13. To further investigate the significant influence of the unilateral Schottky barrier on detector performance, two types of devices with different configurations—Au/WS<sub>2</sub>/Gr and Au/WS<sub>2</sub>/Au—were fabricated for comparison. Photocurrent mapping measurements were performed on both devices under zero bias using 405 nm light illumination at the same power, as shown in Fig. S14(a) and Fig. S14(b). The Au/WS<sub>2</sub>/Gr device exhibits higher photocurrent response than the Au/WS<sub>2</sub>/Au one, demonstrating the effect of the unilateral Schottky junction. The I- V characteristic curve of the Au/WS<sub>2</sub>/Gr device also shows distinct rectifying behavior, consistent with the presence of a unilateral Schottky barrier. In addition, the symmetric I-V characteristic curve of the Gr/WS<sub>2</sub>/Gr confirmed the ohmic contact between Gr and WS<sub>2</sub>.

In Au/WS<sub>2</sub>/Ag/Gr structure (denoted as device3), the incorporation of Ag NPs induces a pronounced LSPR effect, which substantially boosts the carrier concentration and consequently amplifies the photocurrent, as shown in Fig. 3(c).

Specifically, the near-field enhancement effect (Energy Transfer, ET) directly generates more electron-hole pairs through an increase in the electromagnetic field density, thereby producing more high-energy excitons. Meanwhile, hot-electron injection (Charge Transfer, CT) injects the generated hot electrons into the absorption layer to contribute to the photocurrent. At the same time, the Schottky barrier formed between the Au electrode and WS<sub>2</sub> promptly separates the carriers, preventing unfavorable recombination processes. Furthermore, Raman measurements were performed on the Au/WS<sub>2</sub>/Gr and Au/WS<sub>2</sub>:Ag/Gr devices before and after the deposition of Ag NPs in Fig. S15. The results show that the peak shifts indicate that the presence of the LSPR effect indeed induces ET in the heterojunction.

To quantitatively elucidate the efficient separation and transfer of high-energy excitons driven by the built-in electric field and LSPR, the broadband femtosecond TA spectroscopy was employed to measure and calculate the lifetimes of high-energy excitons and band-edge excitons in WS<sub>2</sub> within different structures. Based on the spectral signals, a probe wavelength of 401 nm was selected. Fig. 3(d) presents the normalized decay curves of high-energy carriers in WS<sub>2</sub>. The decay kinetics curves were fitted using a triple-exponential function for the two decay curves:

$$y = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + A_3 \exp\left(-\frac{t}{\tau_3}\right) \quad (1)$$

Among them,  $A_1$ ,  $A_2$ , and  $A_3$  represent the weighting factors, and  $\tau_1$ ,  $\tau_2$  and  $\tau_3$  are the time constants. The effective lifetime ( $\tau_{\text{eff}}$ ) can be obtained using the following formula:

$$\tau_{\text{eff}} = \frac{A_1 \tau_1^2 + A_2 \tau_2^2 + A_3 \tau_3^2}{A_1 \tau_1 + A_2 \tau_2 + A_3 \tau_3} \quad (2)$$

According to the calculated results, the lifetime of high-energy excitons in intrinsic WS<sub>2</sub> is 262.9 ps. After the introduction of the strong built-in electric field, the lifetime of high-energy excitons in WS<sub>2</sub> decreases to 98 ps, which is significantly shorter than that of intrinsic WS<sub>2</sub>. These results effectively validate the proposed hypothesis regarding the role of the unilateral Schottky barrier. Furthermore, the lifetime of high-energy excitons in the Au/WS<sub>2</sub>:Ag/Gr structure is 245 ps. With the plasmonic layer serving as an enhancement layer, and based on the near-field enhancement and hot-electron injection effects arising from the LSPR effect, the presence of Ag NPs leads to enhanced near-field effects that generate a greater number of high-energy excitons in WS<sub>2</sub>. This lifetime extension reflects the combined effects of slowed phonon-assisted intervalley scattering and enhanced Pauli blocking, both arising from the increased C-exciton population induced by hot-electron injection from the Ag NPs [82]. Subsequently, the lifetimes of band-edge excitons in the different structures were also investigated. Fig. 3(e) presents the normalized decay curves of band-edge exciton lifetimes, measured using a probe wavelength of 629 nm. According to the fitting results, the band-edge exciton lifetime in intrinsic WS<sub>2</sub> is 793 ps. After the introduction of the strong built-in electric field, the band-edge exciton lifetime in WS<sub>2</sub> decreases to 426 ps, which is significantly shorter than that of intrinsic WS<sub>2</sub>, further confirming the rapid carrier separation effect of the unilateral Schottky junction. For the Au/WS<sub>2</sub>:Ag/Gr structure, the LSPR effect induced by the Ag nanoparticles promotes the formation of a greater number of band-edge excitons. The increase in the number of band-edge excitons leads to a prolonged lifetime.

To investigate the roles of hot-electron injection and near-field enhancement, an Au/WS<sub>2</sub>/h-BN:Ag/Gr structure was fabricated and characterized by transient absorption spectroscopy. A thick h-BN layer (~20 nm) was inserted as a blocking layer to suppress charge transfer, as identified in Fig. S16. As shown in Figs. 3(d) and 3(e), after the introduction of h-BN, the lifetime of high-energy excitons shows no significant change, only becoming slightly shorter than that without h-BN. Under the LSPR effect, both near-field enhancement and hot-electron injection processes coexist. However, when h-BN is present, the electron transfer process is blocked, leaving only the near-field energy transfer process. After the loss of the electron transfer process, the reduced number of charge carriers leads to a shorter lifetime. The lifetime of A excitons also decreases slightly after the addition of the thick h-BN layer. Some A excitons are converted from C excitons; therefore, when the introduction of h-BN reduces the number of C excitons, the number of A excitons also decreases accordingly.

A high-performance Au/WS<sub>2</sub>:Ag/Gr blue-light photodetector is employed and integrated into a single-pixel imaging system to explore its potential for practical applications. The imaging setup is illustrated in Fig. 4(a). The system includes a light source that illuminates the target object. After passing through the object, the light is directed onto a digital micromirror device (DMD). The spatially modulated light patterns from the DMD are optically coupled to the photodetector. Computational reconstruction of the object image is achieved by decoding the detected intensity signals with the corresponding DMD modulation patterns. As depicted in Fig. 4(b), a Hadamard matrix, denoted by the letter  $A$ , was used for light-field modulation in single-pixel imaging, with the object reconstructed as a one-dimensional vector. The interaction between the object and the matrix formed by all rows of the Hadamard matrix produces a one-dimensional vector, which is captured by the photodetector. Subsequently, object information  $X$  is extracted using an inverse method ( $X = Y \cdot A^{-1}$ ), enabling reconstruction of the object image.

To demonstrate the potential of the WS<sub>2</sub>:Ag detector for underwater detection, the object "H" was placed in air, clear water, and turbid water, respectively, followed by imaging under each condition. Fig. 4(b) shows the imaging results of the object in these different environments. Clear differences in image clarity can be observed across the environments. When the sample pattern "H" was placed in air and illuminated by blue, green, and red-light sources, the photocurrent of the device reached saturation due to sufficient light intensity, resulting in similar imaging clarity without significant differences. When the sample pattern "H" was placed in clear water, the clarity under different light sources varied: blue light provided the highest clarity, followed by green light, while red light yielded the least clear image. This is attributed to the varying degrees of light transmission through water at different wavelengths. When the sample pattern "H" was placed in a turbid underwater environment, the imaging results matched expectations, with clarity gradually decreasing as the wavelength of the light source increased. To quantify the clarity of our imaging, we extracted and compared the edge gradients of each image, as shown in Fig. 4(c). The edge gradient is defined as the intensity of sharp changes in brightness or color within the image. Additionally, we calculated and compared the PSNR values of

each image, as shown in Fig. 4(d). PSNR is defined as the ratio between the maximum possible power of a signal and the power of corrupting noise that affects the fidelity of its representation. In air, the PSNR value of blue-light imaging is approximately 24.8. These results confirm that the blue-light photodetector is capable of underwater imaging. In the context of the unique underwater environment, our imaging demonstration highlights the advantage of using a self-driven blue-light detector without an external bias voltage, which is unachievable with traditional photoconductive photodetectors.

### 3 Conclusions

In summary, we have successfully developed a self-powered single-pixel blue photodetector based on multilayer WS<sub>2</sub> decorated with Ag NPs, featuring a unilateral Schottky junction. The synergistic effect between the high-energy excitons of WS<sub>2</sub> and the localized surface plasmon resonance of Ag NPs enables a competitive zero-bias photoresponse, including a responsivity of 244 mA/W and an external quantum efficiency of 74.7% under 405 nm illumination. Ultrafast transient absorption spectroscopy further reveals that the LSPR effect significantly promotes the generation of high-energy excitons, offering microscopic insight into the observed performance enhancement. Importantly, the device demonstrates practical applicability by achieving high-resolution (128 × 128) single-pixel blue-light imaging in both clear and turbid water. This work not only provides a promising material and device design strategy for efficient blue light detection, but also validates the feasibility of self-powered photodetectors in realistic underwater scenarios, paving the way for advancing low-power underwater optical communication and high-visibility imaging technologies.

## 4 Experimental and theoretical methods

### 4.1 Device fabrication

First, a pair of Au electrodes (~40 nm) was fabricated on the silicon substrate coated with a 300 nm layer of silicon dioxide via a combination of maskless lithography (MLA100) and thermal evaporation (Technol Science, ZHD-400) techniques. By using PDMS as an assisting layer, the exfoliated WS<sub>2</sub> and graphene samples were transferred onto the bottom Au electrode in a step-by-step manner. Finally, the vertically aligned device was subjected to a mild vacuum annealing process for 20 min at 420 K, with the objective of achieving high-quality van der Waals interfacial contacts.

### 4.2 Characterization

The thickness of the multilayer WS<sub>2</sub> and few-layer graphene flakes was determined by an atomic force microscope (AFM) system (Dimension Icon, Bruker) in the tapping mode under ambient conditions. Corresponding optical images were acquired using an optical microscope (Olympus BX51) equipped with a 50× objective lens. The steady-state absorption spectra, and all electrical and photoelectric properties of devices were measured with High-resolution & High-speed Multi-physical Scanning Integrated Test System (Metatest, ScanPro Advance) equipped with a Xenon lamp (300-1100 nm) light source, a monochromator, a source-meter (Keithley, 2614b), and multiple continuous-wave lasers (265, 320, 405, 532, and 638 nm). A regeneratively amplified Ti:sapphire laser from Coherent Corporation, nonlinear frequency mixing techniques, and a transient absorption spectrometer (TA100) from Time-tech

Spectra LLC form the basis of the femtosecond TA setup.

### 4.3 Theoretical Calculation

The electronic properties of multilayer WS<sub>2</sub> were investigated via density functional theory (DFT) using the Vienna Ab initio Simulation Package (VASP). In these calculations, the Perdew-Burke-Ernzerhof (PBE) functional within the generalized gradient approximation (GGA) was used for the exchange-correlation potential, while the projector augmented-wave (PAW) method was employed to describe electron-ion interactions. A kinetic energy cutoff of 300 eV was set for the plane-wave basis. The Brillouin zone integration was carried out using a 12×12×6 k-point mesh generated by the Monkhorst-Pack scheme. For further computational details, please refer to our previous work.

**Electronic Supplementary Material:** Supplementary material (the variation of EQE with thickness for multilayer WS<sub>2</sub> flakes, typical AFM images of a multilayer WS<sub>2</sub> and graphene flake; the detailed fabrication process of the WS<sub>2</sub>/Gr photodetector; XPS spectra of the Au/WS<sub>2</sub>/Gr detector before and after the deposition of quantum dots; Raman spectra of multilayer WS<sub>2</sub>, graphene, and the WS<sub>2</sub>/Gr heterostructure; response time and I-T curves of the Au/WS<sub>2</sub>/Gr/Ag blue photodetector, EQE, power-dependent R and D\* under 405 nm light illumination at zero bias; comparison of EQE, Rand specific D\* between the Au/WS<sub>2</sub>/Gr device and other reported self-powered blue photodetectors; the response time stability of the Au/WS<sub>2</sub>/Gr/Ag blue photodetector; photocurrent mapping image and potential difference of Au/WS<sub>2</sub>/Gr and Au/WS<sub>2</sub>/Au; I-V characteristics of the Gr/WS<sub>2</sub>/Gr and Au/WS<sub>2</sub>/Gr photodetector under dark conditions; Raman spectra and Raman peak shifts of the WS<sub>2</sub> and WS<sub>2</sub>/Ag structures) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909030>.

### Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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### Declaration of competing interest

All the contributing authors reports no conflict of interests in this work.

### Author contribution statement

Y. Z. L.: Conceptualization, data curation, formal analysis, funding acquisition, investigation, methodology, writing original draft, reviewing and editing manuscript. J. X. L. and C. M. Z.: Data curation, investigation, methodology, writing original draft, reviewing and editing manuscript. Q. B. W.: Investigation. C. X. Y.: Investigation. W. X.: Methodology. Y. S. G.: Investigation. C. Z.: Conceptualization. H. Y. X.: Conceptualization, funding acquisition, project administration, resources, software, supervision, reviewing and editing manuscript.

## Use of AI statement

During the preparation of this work, Y. Z. L. used Nano Banana in order to generate the base map of Fig. 1a to illustrate underwater detection. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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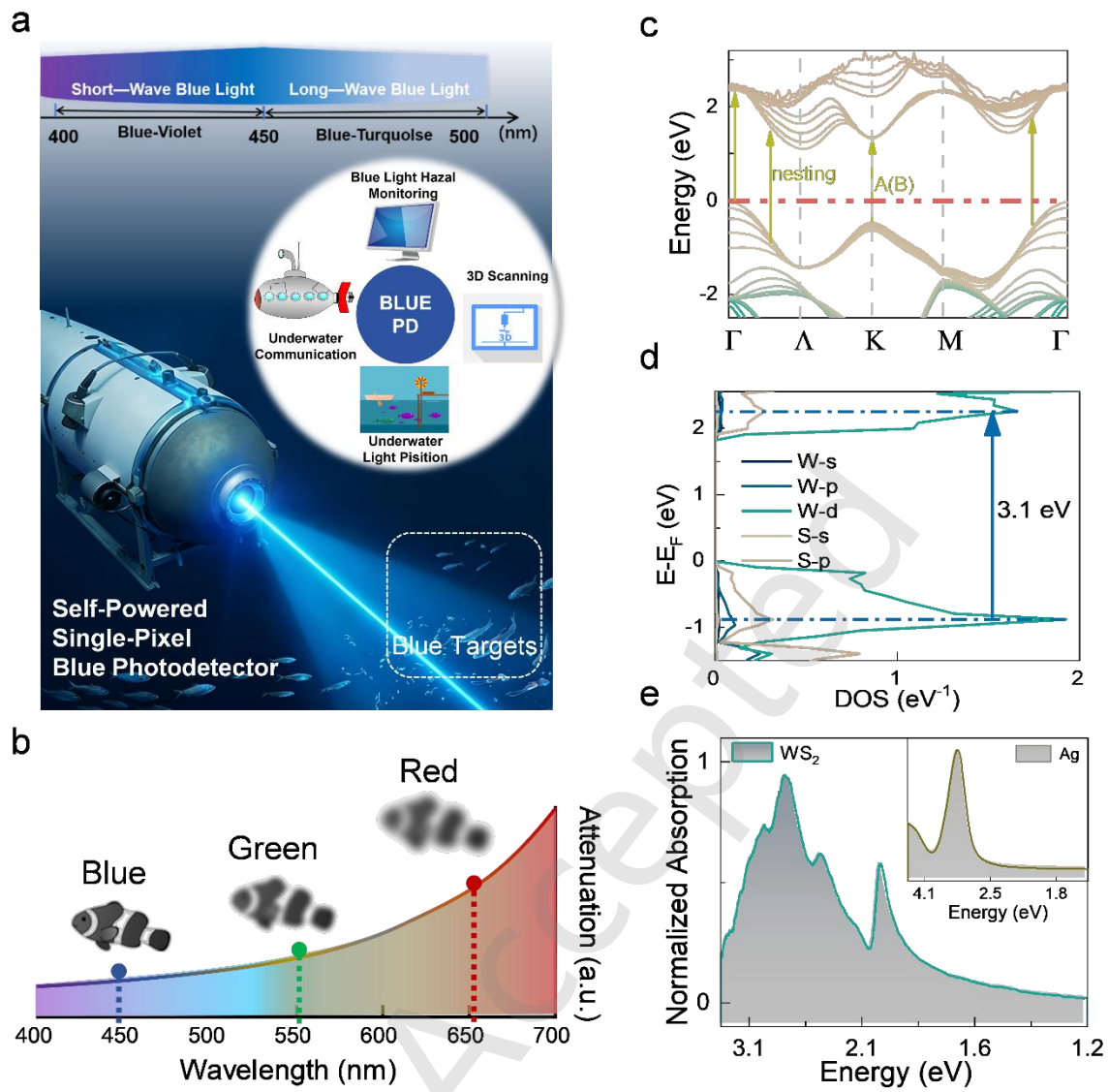
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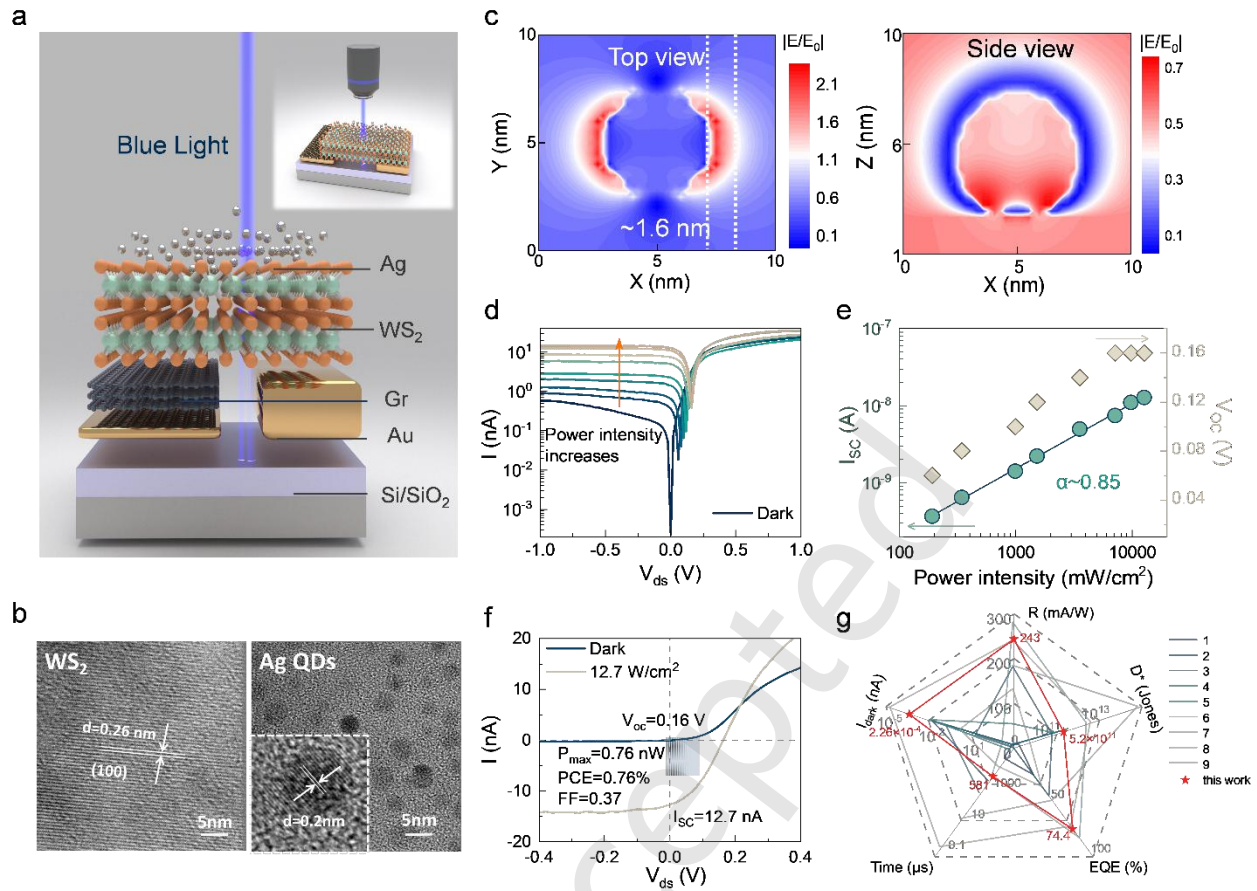
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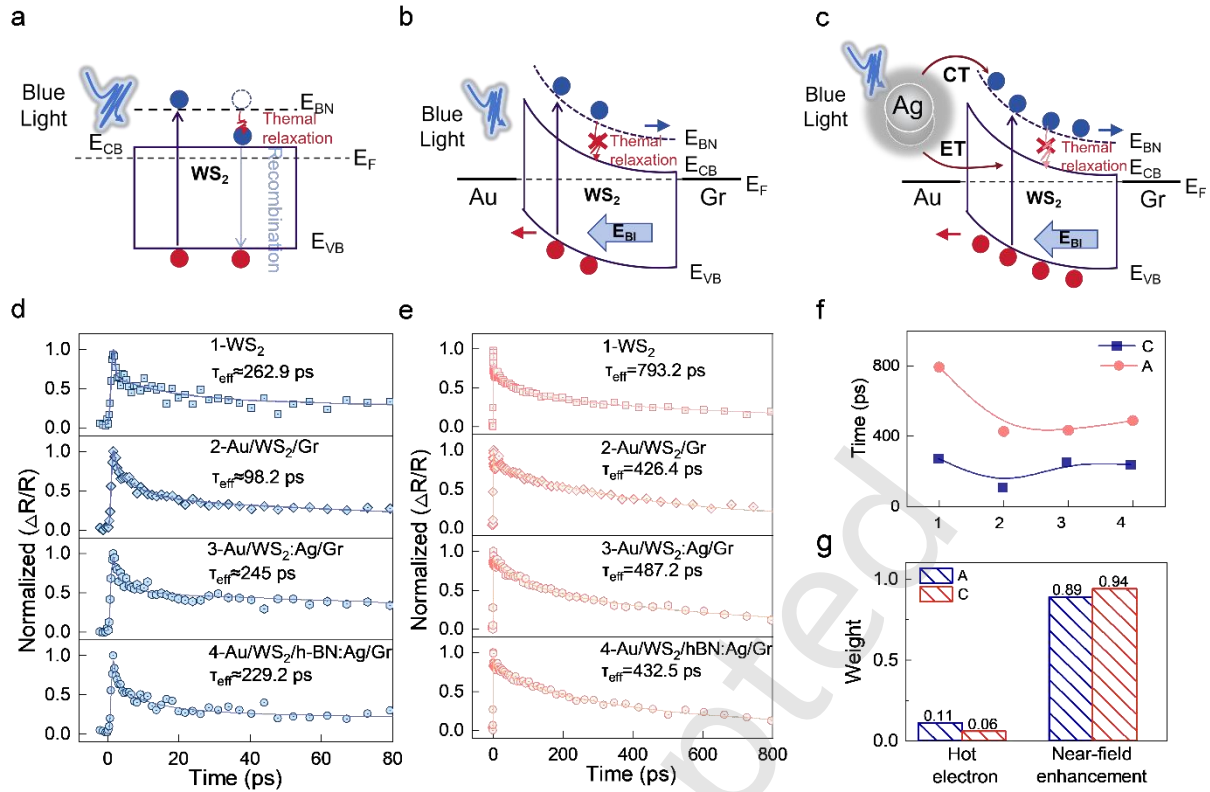
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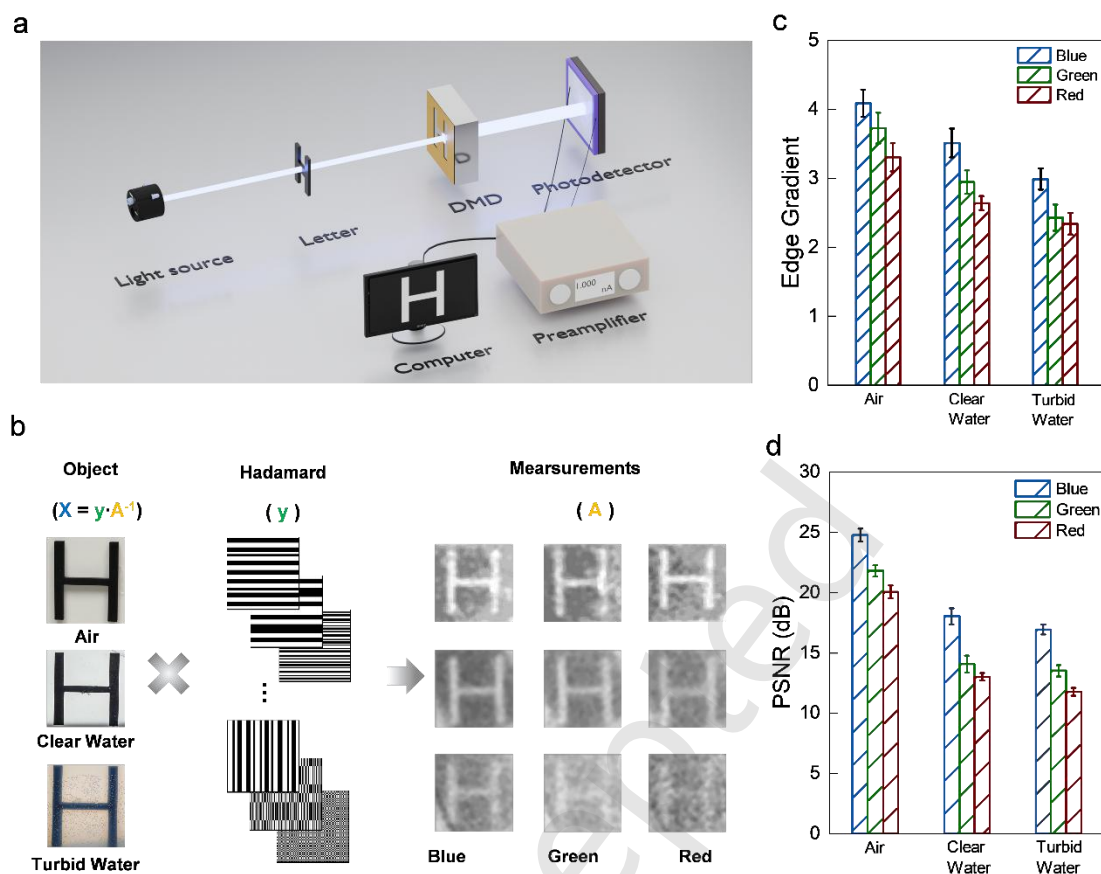
**Figure 1.** Design of high-performance blue photodetectors by efficient utilization of high-energy excitons and LSPR effect. (a) Schematic diagram of underwater detection by a blue light detector. (b) Refraction of light of different wavelengths by seawater. (c) Theoretically calculated band structure of multilayer WS<sub>2</sub> based on DFT. (d) Density of states of WS<sub>2</sub>. (e). Absorption spectrum of multilayer WS<sub>2</sub>, inset: absorption spectrum of Ag NPs.



**Figure 2. Self-powered blue photodetection performance.** (a) Schematic diagram of the Au/WS<sub>2</sub>/Ag/Gr device. (b) TEM images of WS<sub>2</sub> and Ag NPs indicates good material quality. (c) Right: Simulated electric field distribution of 5 nm Ag NPs. (d) Current-voltage characteristics of the photodetector under dark conditions and 405 nm light illumination (semi-log scale). (e) Power-dependent  $I_{sc}$  and  $V_{oc}$  under 405 nm light illumination at zero bias. (f) (g) Comparison of EQE,  $R$ ,  $D^*$ , dark current, and response time of the Au/WS<sub>2</sub>/Ag/Gr device with other reported blue light self-powered photodetectors, as listed in Table S1.

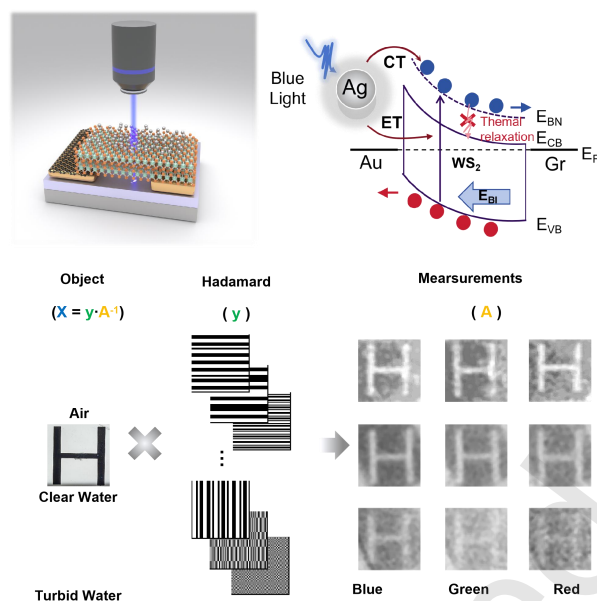


**Figure 3. Schematic diagrams of band arrangement and dynamic process of photogenerated carriers.** Schematic diagrams of photogenerated carriers in the (a)  $\text{WS}_2$ , (b)  $\text{Au}/\text{WS}_2/\text{Gr}$  and (c)  $\text{Au}/\text{WS}_2:\text{Ag}/\text{Gr}$  upon blue light irradiation.  $E_{BN}$ ,  $E_{CB}$ ,  $E_{VB}$ , and  $E_F$  represent band nesting region, the conductive band minimum, the valence band maximum, and the Fermi level, respectively. The normalized decay curves of (d) high-energy exciton and (e) band-edge exciton lifetimes. The solid lines are the fitting curves. (f) Lifetime variation of high-energy exciton and band-edge exciton. (g) The weight of hot-electron effect and near-field enhancement effect in the lifetimes of band-edge excitons and high-energy excitons.



**Figure 4. Single-pixel underwater imaging of objects.** (a) Illustration of the experimental setup for single-pixel imaging system. (b) Principle of single-pixel imaging using a Hadamard matrix for light field modulation. (c) Edge gradient comparison of imaging in air, clear water, and turbid water, and (d) peak signal-to-noise ratio (PSNR).

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Leveraging the synergistic effect of the LSPR effect and high-energy excitons, we developed a high-responsivity blue photodetector achieving an external quantum efficiency of 74.7%, capable of single-pixel imaging under various environments.

# Harnessing plasmon-exciton synergy in multilayer WS<sub>2</sub> based self-powered blue photodetector for high-visibility underwater imaging

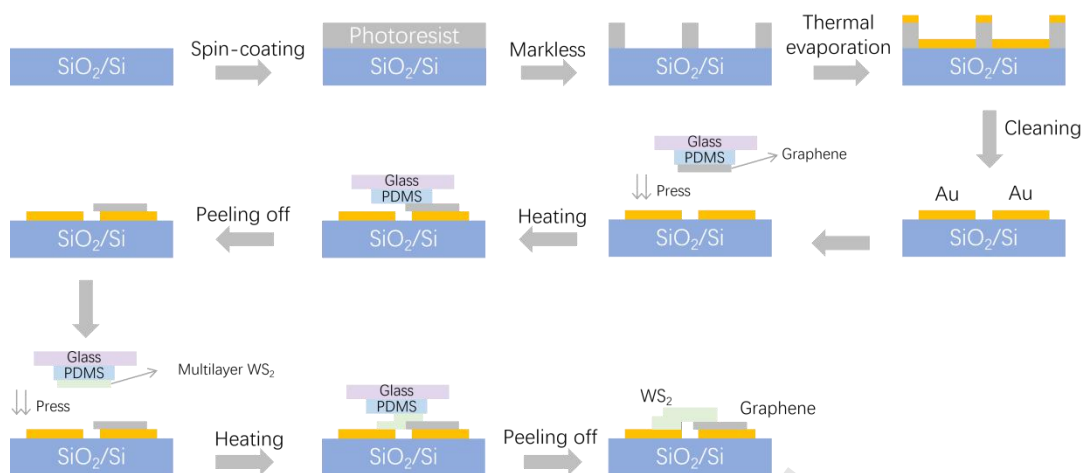
Jixiu Li<sup>§</sup>, Chenmeng Zhao<sup>§</sup>, Yuanzheng Li<sup>id</sup>✉, Chuxin Yan, Qingbin Wang, Yongsheng Gao, Wei Xin, Cheng Zhou<sup>id</sup>✉ and Haiyang Xu<sup>id</sup>✉

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<sup>§</sup> Jixiu Li and Chenmeng Zhao contributed equally to this work.

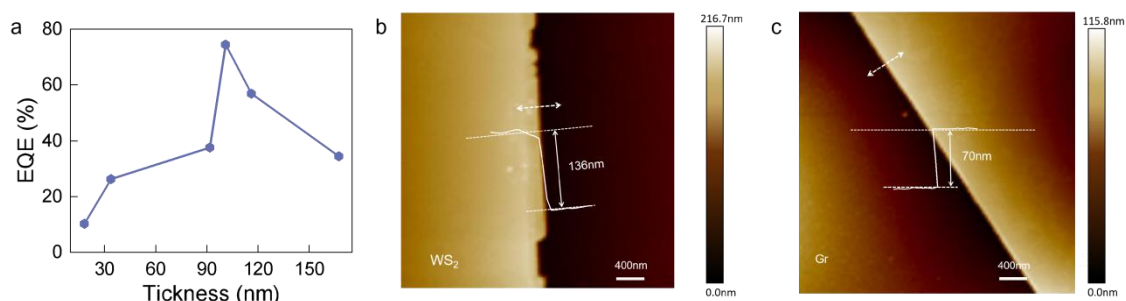
Supporting information to <https://doi.org/10.26599/NR.2026.94909030>

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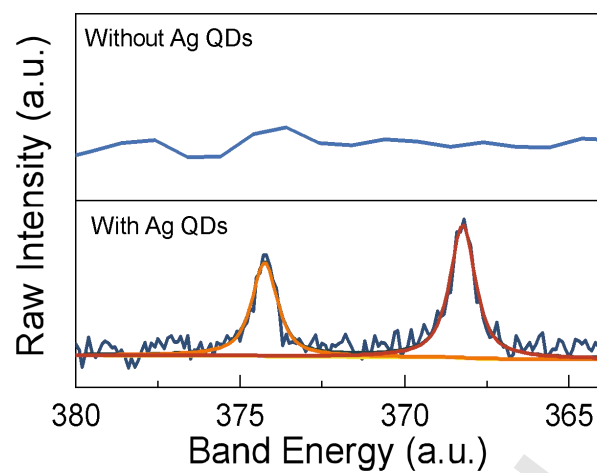
**Figure S1.** A schematic diagram illustrating the detailed fabrication process of the WS<sub>2</sub>/Gr photodetector using the dry transfer technique.

The silicon substrate with 300 nm silicon dioxide (SiO<sub>2</sub>/Si) was sequentially cleaned by ultrasonication in deionized water, trichloroethylene, acetone, anhydrous ethanol, and secondary deionized water for 10 minutes each, and finally dried with pure nitrogen gas. Subsequently, micropatterns serving as source and drain electrodes were fabricated on the SiO<sub>2</sub>/Si substrate using standard photolithography, followed by the deposition of Au electrodes with uniform thickness via thermal evaporation. Next, the substrate with patterned Au electrodes was sequentially immersed in acetone and ethanol to remove residual photoresist, and finally dried with pure nitrogen gas. Multilayer graphene and multilayer WS<sub>2</sub> were first exfoliated onto polydimethylsiloxane (PDMS) films using Scotch tape, and then transferred onto the prepatterned Au electrodes using a precise transfer stage. Here, multilayer graphene was placed on one side of the Au electrode, and then multilayer WS<sub>2</sub> flakes were deposited onto the multilayer graphene and the Au electrode on the other side, forming a horizontally structured WS<sub>2</sub>/Gr photodetector. The PDMS film can be released and detached by heating to 80 °C.

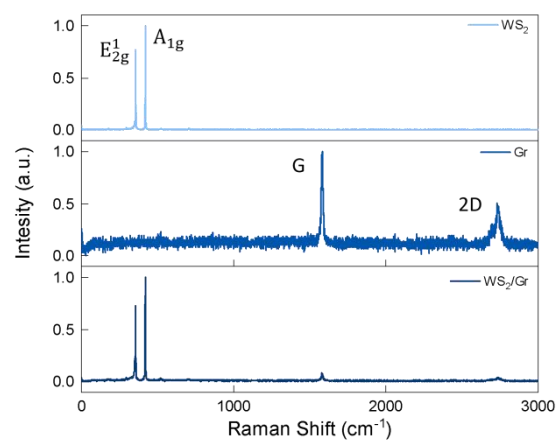


**Figure S2.** (a) The variation of EQE with thickness for multilayer WS<sub>2</sub> flakes. (b) Typical AFM images of a multilayer WS<sub>2</sub> flake and (c) graphene flake with their thicknesses determined to be approximately 136 nm and approximately 70 nm, respectively. The white lines depict the height profiles along the scan lines indicated by the dashed white double-headed arrows.

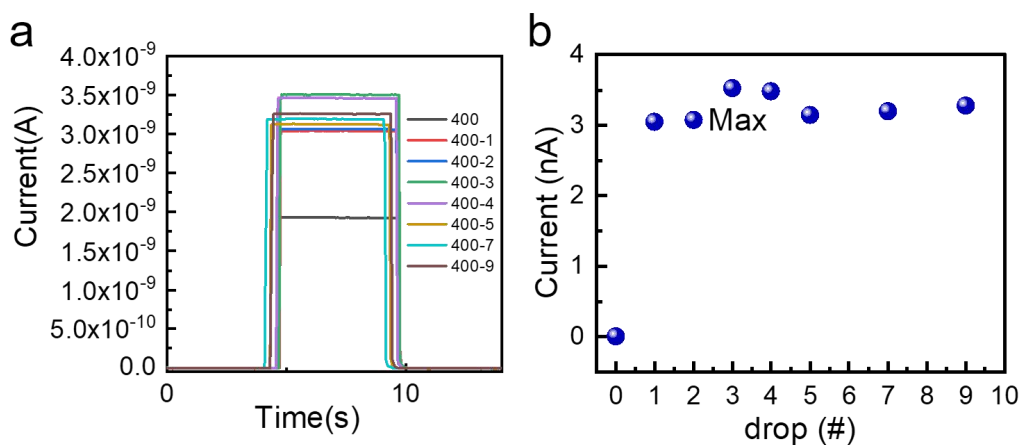
To determine the optimal thickness of WS<sub>2</sub>, we fabricated devices using WS<sub>2</sub> flakes of varying thicknesses. Photocurrent measurements were performed on each device, and the corresponding external quantum efficiency (EQE) was calculated. Based on these results, a multilayer WS<sub>2</sub> flake with a thickness of 136 nm was selected for the fabrication of the blue photodetector.



**Figure S3.** XPS spectra of the Au/WS<sub>2</sub>/Gr detector before and after the deposition of quantum dots.

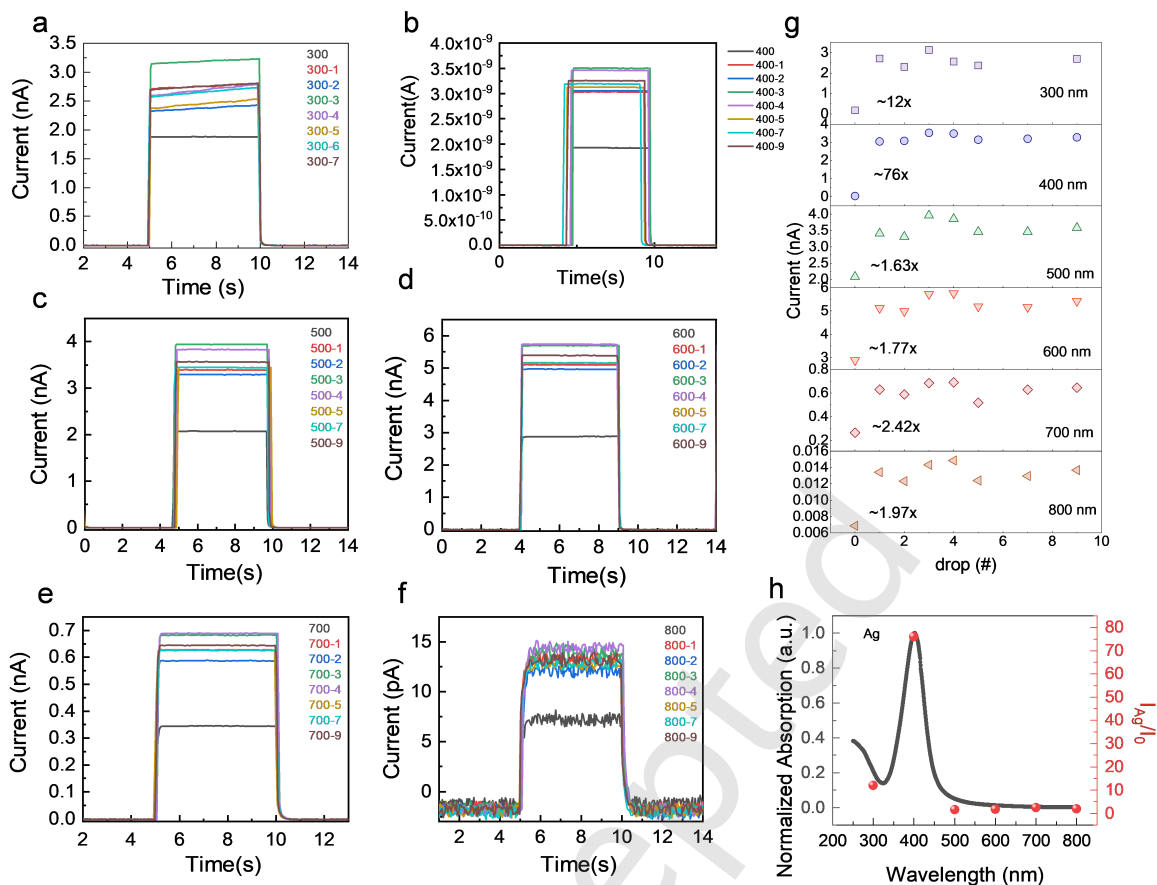


**Figure S4.** Raman spectra of multilayer  $\text{WS}_2$ , Gr, and the  $\text{WS}_2/\text{Gr}$  heterostructure from the horizontally aligned photodetector.



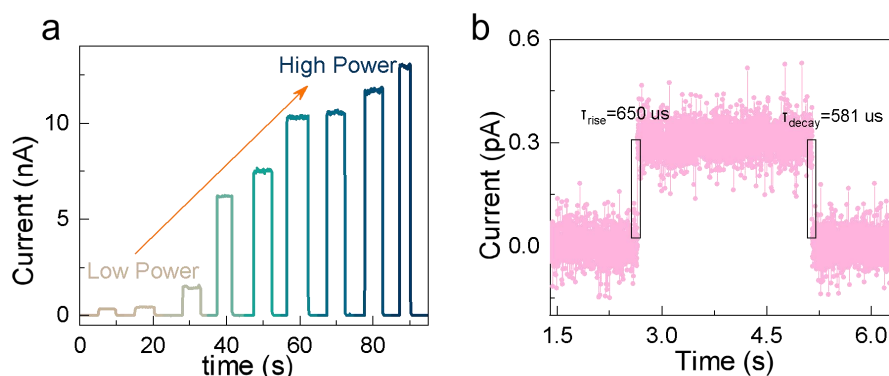
**Figure S5.** (a) Photocurrent of the device with various solution droplet under 400 nm light. (b) The photocurrent values extracted from (a) as a function of quantum dot concentration.

To determine the optimal concentration of quantum dots, we measured the photocurrent of the device by varying the concentration through controlled droplet deposition. As shown in Fig. S5, the device exhibited the highest photocurrent response when four droplets were applied. Given that the as-prepared quantum dot solution had a concentration of 0.1 mg/mL, the optimal concentration was determined to be 0.4 mg/mL, which was therefore used in all subsequent measurements.



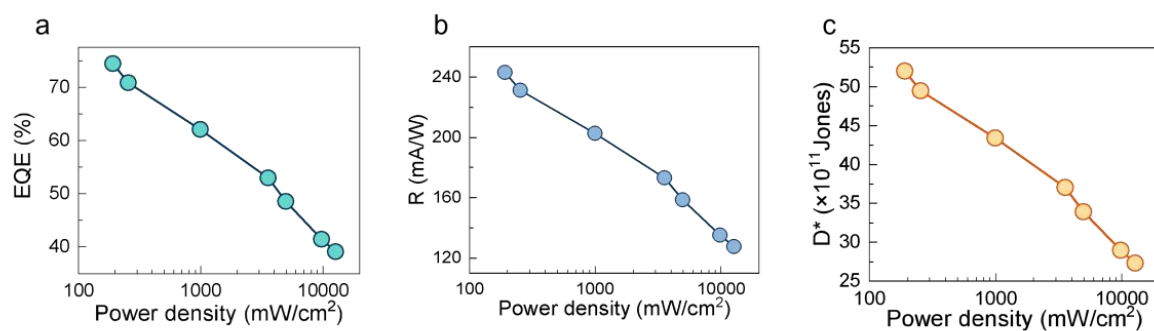
**Figure S6.** Wavelength-dependent photocurrent enhancement of the WS<sub>2</sub> photodetector induced by Ag NPs. Photocurrent responses of the device measured at (a) 300, (b) 400, (c) 500, (d) 600 nm, (e) 700 nm and (f) 800 nm before and after the deposition of Ag NPs. (g) Photocurrent of the photodetector as a function of the number of Ag NP deposition droplets. (h) Enhancement factor as a function of wavelength extracted from (g), overlaid with the normalized absorption spectrum of Ag NPs.

To verify the LSPR contribution, systematic photocurrent measurements have been performed across the spectral range from 300 nm to 800 nm, both before and after the deposition of Ag NPs on the WS<sub>2</sub> device. As shown in Fig. S6, the photocurrent enhancement factor ( $I_{Ag}/I_0$ ) exhibits a clear maximum at 400 nm, which coincides precisely with the LSPR absorption peak of the Ag NPs (Fig. S6h). At other wavelengths, the enhancement factor is significantly smaller. This wavelength-dependent behavior provides direct evidence that the performance improvement is predominantly attributed to the LSPR effect of the Ag NPs, which resonantly enhances the local electromagnetic field and promotes hot-electron injection specifically within the spectral region matching the plasmon resonance. We acknowledge that the enhancement factors at other wavelengths are relatively modest. This is partially attributed to the fact that the Ag NPs were dispersed in ethanol before deposition. Although the ethanol was removed, trace solvent effects during the deposition process may have slightly modified the WS<sub>2</sub> surface state or doping level, leading to a weak, broadband photoresponse modulation that is not related to the LSPR effect.

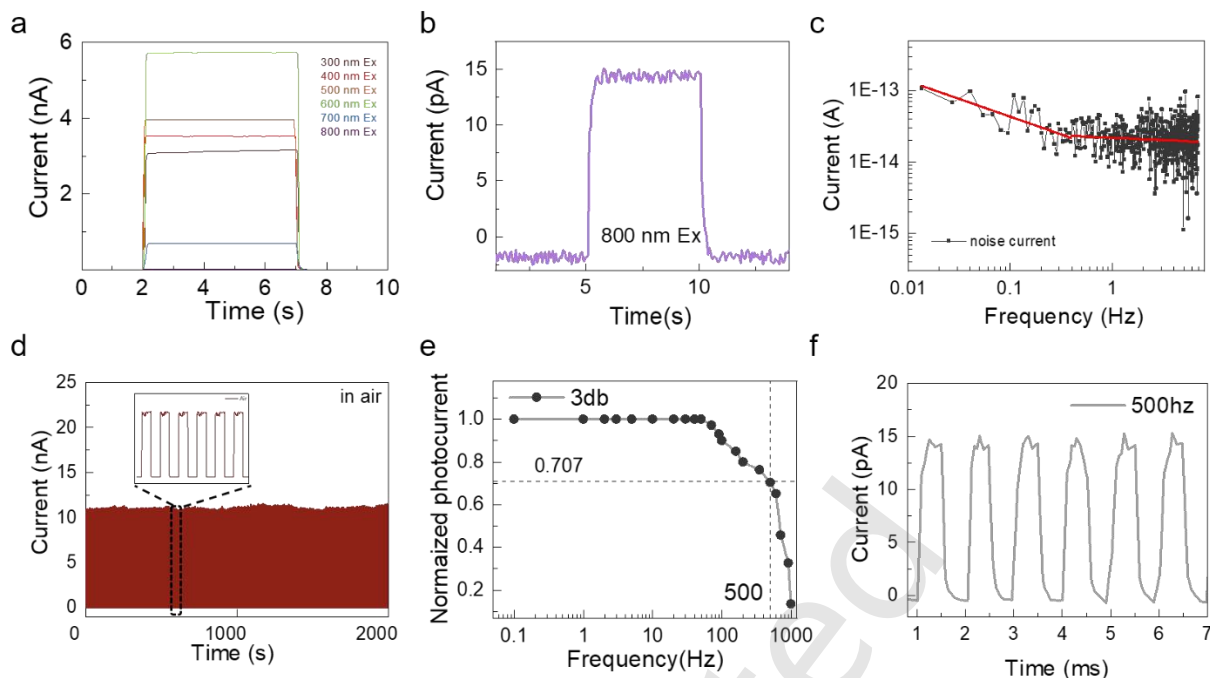


**Figure S7.** (a) I-T curves of the Au/WS<sub>2</sub>/Gr/Ag blue photodetector under different illumination powers. (b) Response time of the Au/WS<sub>2</sub>/Gr/Ag blue photodetector.

The photocurrent curves of the Au/WS<sub>2</sub>/Gr/Ag blue photodetector were measured under various illumination powers, demonstrating that the device exhibits a favorable photoresponse to blue light across all tested power levels. The response time was calculated by taking the 10% to 90% intervals of the rising and falling edges, yielding a rise time of 650  $\mu\text{s}$  and a fall time of 581  $\mu\text{s}$ . These results indicate that the detector possesses a fast response capability to blue light.

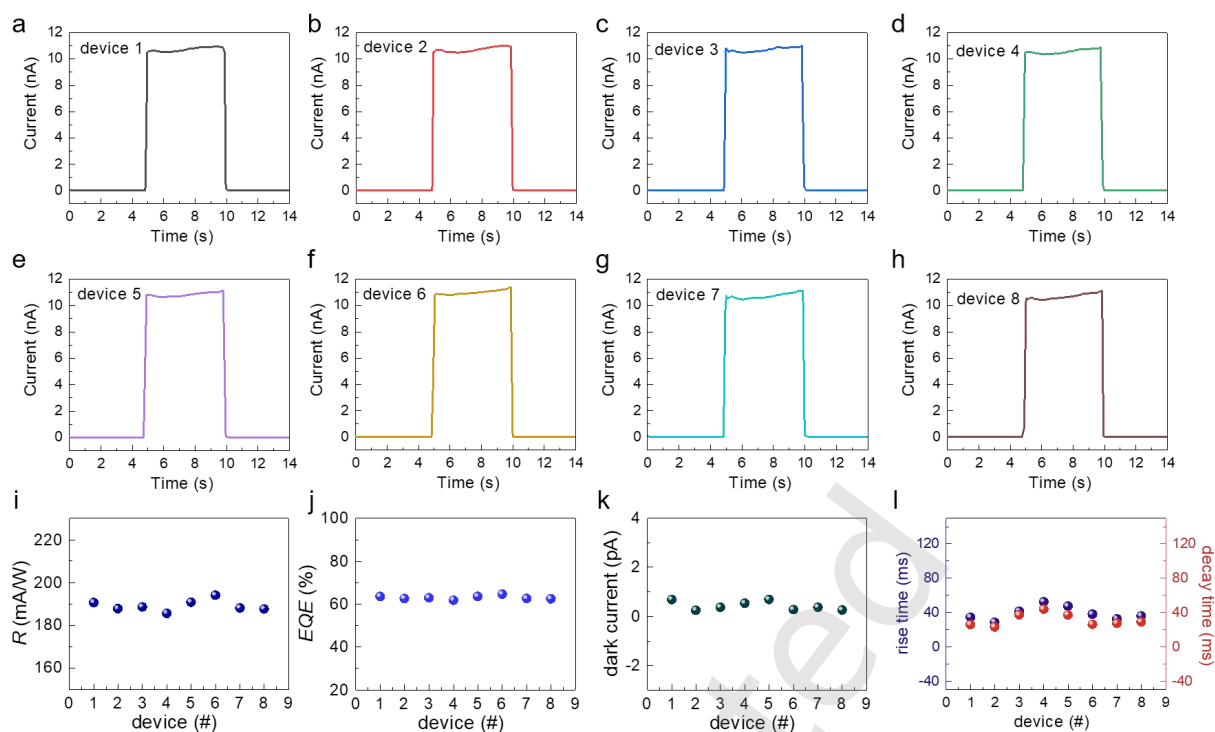


**Figure S8.** Power-dependent (a) EQE, (b) R and (c)  $D^*$  under 405 nm light illumination at zero bias.

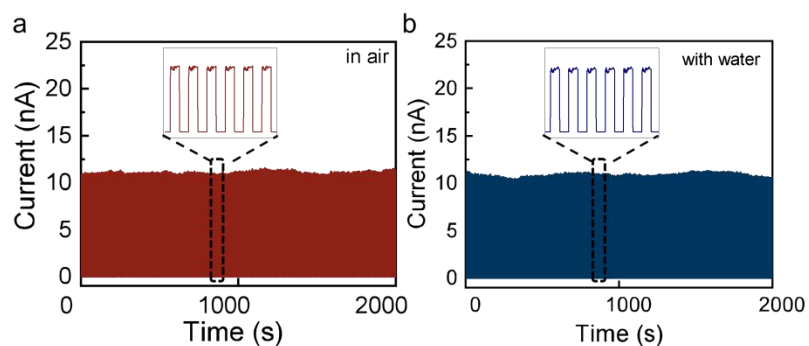


**Figure S9.** (a) Spectral response of the device under excitation from 300 nm to 800 nm. (b) Photocurrent response curve under 800 nm excitation. (c) Noise current spectrum. (d) The response time stability of the photodetector in air. (e) -3 dB bandwidth measurement of the device. (f) High-frequency modulation response measurement of the device.

Noise equivalent power (NEP) is defined as the minimum incident optical power required for a photodetector to achieve a signal- to- noise ratio of unity at a 1 Hz bandwidth. It can be calculated as  $NEP = i_N / R$ , where  $i_N$  is the noise current spectra at 1 Hz bandwidth with units of  $A \text{ Hz}^{-1/2}$ ,  $R$  is the responsivity of the device. Therefore, the NEP is calculated to be  $9.71 \text{ W Hz}^{-1/2}$  with  $i_N$  of  $2.37 \times 10^{-14} \text{ A Hz}^{-1/2}$  and  $R$  of  $244 \text{ mA/W}$ .

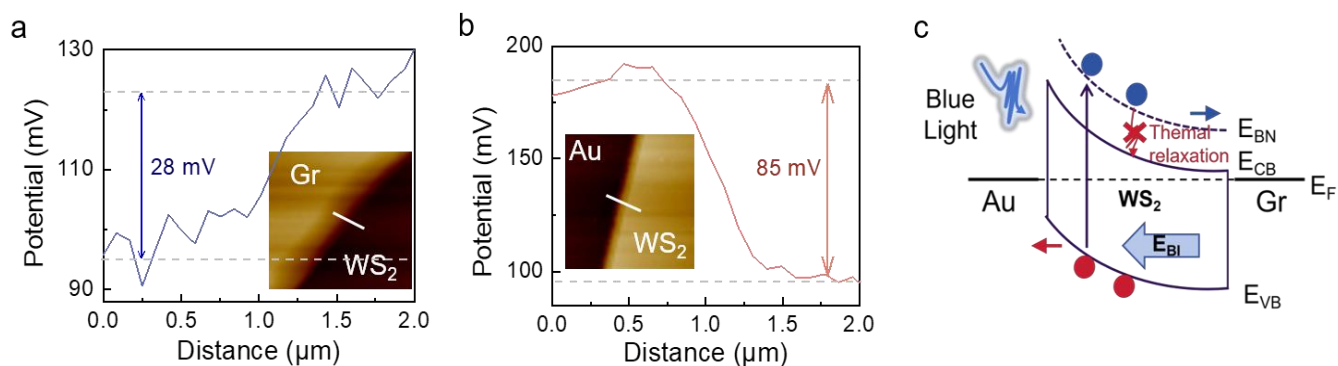


**Figure S10.** (a)-(h) Reproducibility test of eight Au/WS<sub>2</sub>:Ag/Gr photodetectors (devices 1-8), showing negligible device-to-device variation in photocurrent response under 405 nm illumination at zero bias. Statistical distribution of (i) R, (j) EQE, (k) dark current, and (l) response time.



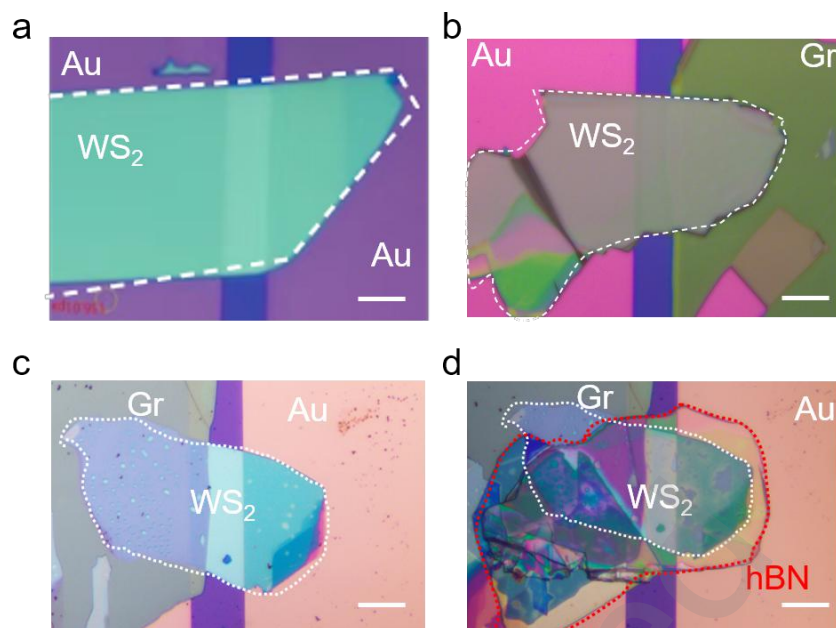
**Figure S11.** The response time stability of the Au/WS<sub>2</sub>/Gr/Ag blue photodetector in (a) air and (b) water.

To investigate the stability of the detector, the photocurrent remained at a consistently stable level after 800 cycles in air. After encapsulating the device with PMMA and applying water droplets onto it, the stability was further evaluated, and the device still exhibited a stable response after 800 cycles.

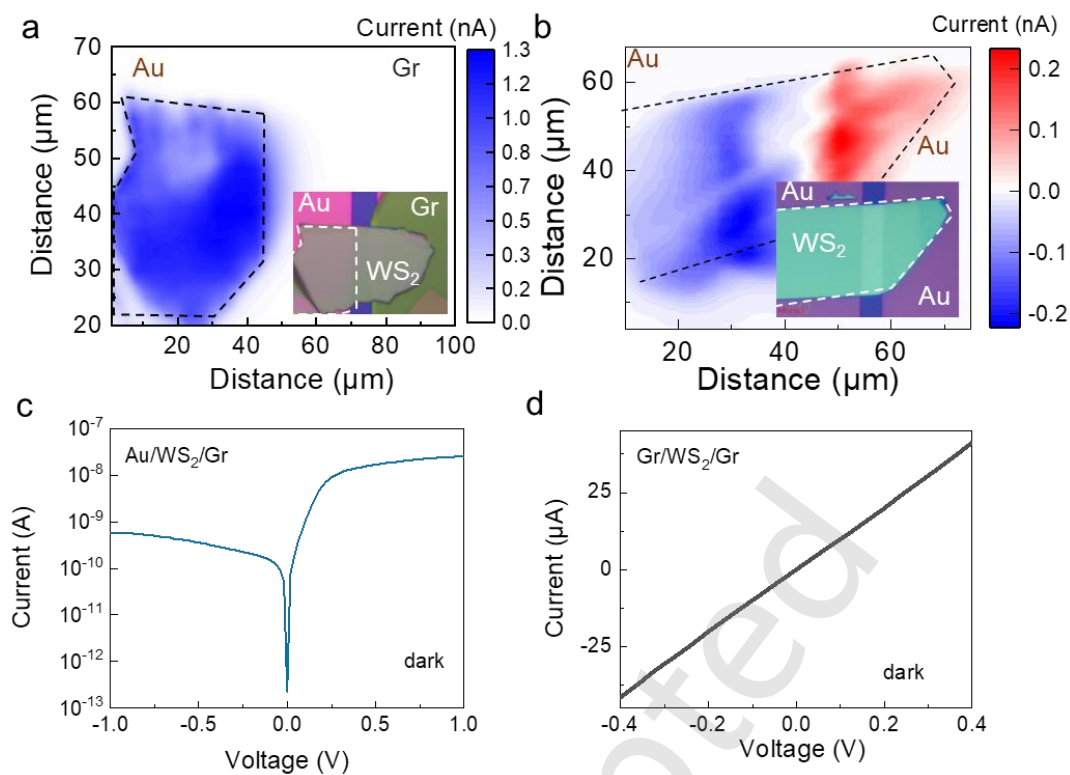


**Figure S12.** (a) Potential difference between Gr and  $\text{WS}_2$ . (b) Potential difference between Au and  $\text{WS}_2$ . (c) Band alignment in the Au/ $\text{WS}_2$ /Gr device upon blue light irradiation.

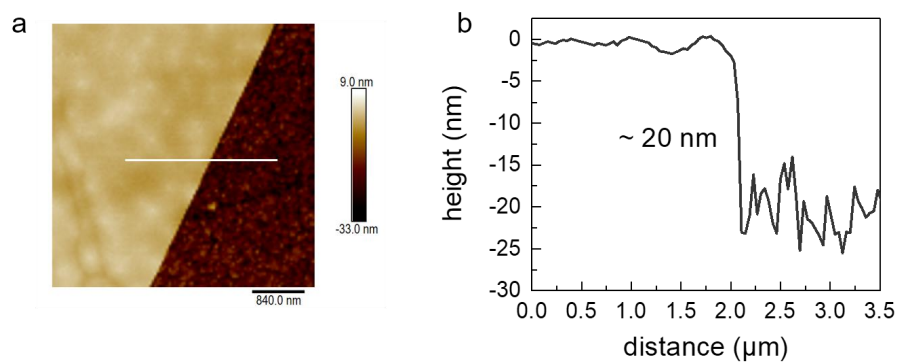
The potential differences at the Gr/ $\text{WS}_2$  and Au/ $\text{WS}_2$  interfaces were measured using Kelvin probe force microscopy. The experimental results are shown in the figure. The disparity in the measured potential differences confirms the existence of a unilateral Schottky barrier. The resulting strong built-in electric field provides the fundamental driving force for the separation of electron-hole pairs.



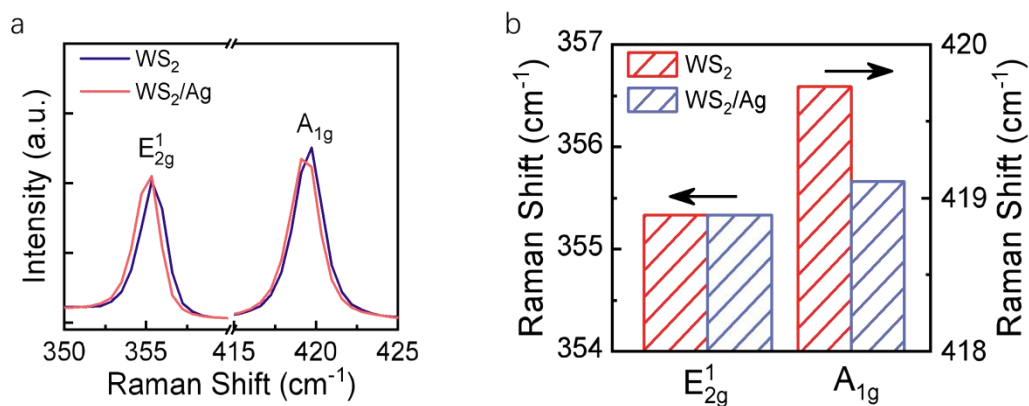
**Figure S13.** Optical images of devices (a) Au/WS<sub>2</sub>/Au, (b) Au/WS<sub>2</sub>/Gr, (c) Au/WS<sub>2</sub>:Ag/Gr and (d) Au/WS<sub>2</sub>/h-BN. Scale bar is 10  $\mu\text{m}$ .



**Figure S15.** (a) Photocurrent mapping image of Au/WS<sub>2</sub>/Gr. (b) Photocurrent mapping image of Au/WS<sub>2</sub>/Au. (c) I-V curves of the Au/WS<sub>2</sub>/Gr photodetector measured under dark conditions. (d) I-V curves of the Gr/WS<sub>2</sub>/Gr photodetector measured under dark conditions.



**Figure S16.** (a) Typical AFM images of a multilayer h-BN flake. (b) height profiles along the scan lines.



**Figure S9.** (a) Raman spectra of the WS<sub>2</sub> and WS<sub>2</sub>/Ag structures. (b) Raman peak shifts of the WS<sub>2</sub> and WS<sub>2</sub>/Ag structures.

The slight shift of the A<sub>1g</sub> peak of WS<sub>2</sub> toward a lower wavenumber is primarily attributed to the n-type doping induced in WS<sub>2</sub> by the high-energy hot electrons generated via the LSPR effect [1].

**Note S1. Calculation of the built-in electric field strength.**

Based on the thermionic emission theory, the current-voltage characteristic of a Schottky junction can be described as:

$$I_{SC} = AA^*T^2 e^{\frac{-\varphi q}{kT}} \left( e^{\frac{V}{kT}} - 1 \right) \quad (1)$$

where  $I_{SC}$  is the device current,  $A$  is the device area,  $A^*$  is the effective Richardson constant,  $T$  is the absolute temperature,  $\varphi$  is the Schottky barrier height,  $k$  is Boltzmann's constant,  $q$  is the elementary charge, and  $V$  is the applied bias voltage. Under self-powered conditions, Equation (1) reduces to:

$$I_d = AA^*T^2 e^{\frac{-\varphi q}{kT}} \quad (2)$$

This simplified expression directly relates the dark current at zero bias to the Schottky barrier height. Therefore, the Schottky barriers of devices are calculated to be 0.456 eV, with corresponding built-in electric field strengths of  $4.56 \times 10^4$  V/m.

**Table S1.** Comparison of the photovoltaic performance between our work and previously reported self-powered blue photodetectors.

| Materials                                                                                        | Wavelengths(<br>nm) | R(mA/W) | EQE(%) | D*(Jones)             | Ref.      |
|--------------------------------------------------------------------------------------------------|---------------------|---------|--------|-----------------------|-----------|
| Bi <sub>2</sub> Te <sub>3</sub> /Bi <sub>2</sub> Se <sub>3</sub> /Bi <sub>2</sub> S <sub>3</sub> | 475                 | 3.5     | 26.17  | 8.96×10 <sup>9</sup>  | [2]       |
| p-Si/n-CdS                                                                                       | 442                 | 4.1     | 1.15   | 9.2×10 <sup>11</sup>  | [3]       |
| MAPbI <sub>3</sub>                                                                               | 490                 | 180     | 46     | 2×10 <sup>11</sup>    | [4]       |
| COF-5/n-Si                                                                                       | 400                 | 4.6     | 1.42   | 6.3×10 <sup>11</sup>  | [5]       |
| Bi <sub>2</sub> Te <sub>3</sub>                                                                  | 530                 | 51      | 65.51  | 2.51×10 <sup>11</sup> | [6]       |
| FA60-PVSK                                                                                        | 400                 | 280     | 84.9   | 5.08×10 <sup>12</sup> | [7]       |
| Rubrene/MoO <sub>3</sub>                                                                         | 455                 | 183     | 50     | 6.35×10 <sup>14</sup> | [8]       |
| Cs <sub>2</sub> AgBiBr <sub>6</sub>                                                              | 435                 | 130     | 37     | 2.4×10 <sup>10</sup>  | [9]       |
| InGaN                                                                                            | 405                 | 240     | 74     | 7.5×10 <sup>11</sup>  | [10]      |
| WS <sub>2</sub> /Ag                                                                              | 405                 | 244     | 74.4   | 5.2×10 <sup>11</sup>  | This Work |

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