

Highly anisotropic dual-heterostructure for multifunctional solar-blind polarization-sensitive photodetectors

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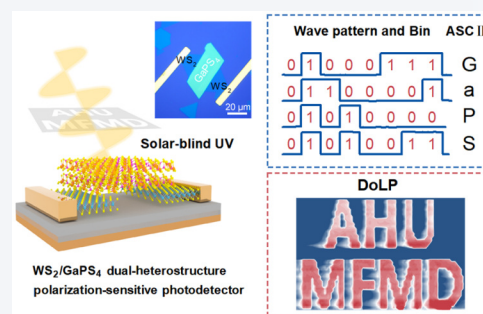
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ABSTRACT: Solar-blind ultraviolet photodetectors based on two-dimensional van der Waals heterostructures have emerged as indispensable components for high-precision warning systems, environmental monitoring systems, information encryption, and optical communication. However, the widespread use of solar-blind ultraviolet photodetectors is still constrained by the simultaneous lack of ultralow dark current, high specific photoresponse, sub-microsecond response time, and intrinsic polarization-sensitive photoresponse. Herein, a dual-heterostructure field-effect transistor composed of the ultra-wide bandgap semiconductor GaPS₄ and WS₂ nanosheets was fabricated. The WS₂/GaPS₄ dual-heterostructure devices exhibit high spike barriers at the dual-heterostructure interface owing to type-I band alignment, which can completely suppress charge transfer. Furthermore, the device exhibits an outstanding performance in the solar-blind ultraviolet (UV) region, stemming from the fast carrier separation and transfer enabled by the built-in electric fields at the dual-heterostructure interface. Critically, the two-fold rotational symmetry (*C*₂) of GaPS₄ breaks the original three-fold rotational symmetry (*C*₃) of WS₂ to simultaneously achieve unexpected linearly polarized Raman and anisotropic absorptions at the heterostructure interface. Moreover, the WS₂/GaPS₄ dual-heterostructure photodetector possesses high-resolution polarization imaging capability under 255 nm illumination owing to dichroic ratios as high as 3.4. These results suggest that the novel two-dimensional dual-heterostructure provides a foundation for developing next-generation polarization-sensitive multifunctional photodetectors.

KEYWORDS: WS₂/GaPS₄ dual-heterostructure, type-I band alignment, anisotropic optical response, solar-blind ultraviolet photodetector, high-resolution polarization imaging



1 Introduction

Polarization-resolved solar-blind ultraviolet photodetectors ($\lambda =$

200–280 nm) have become indispensable in the fields of high-precision warning and tracking, environmental monitoring, secure optical communications, and flame detection [1–4]. State-of-the-art photodetectors in the solar-blind ultraviolet (UV) range including Ga₂O₃ [5], AlN [6], and diamond [7] are technologically mature but remain encumbered by high-temperature epitaxy, sophisticated technology integration, interface defects, and concomitant performance degradation [8]. Recently, low-symmetry ultra-wide bandgap semiconductors, such as GaPS₄ [9], KNb₃O₈ [10], Rb₂CuCl₃ [11], and GaTe_{0.5}Se_{0.5} [12], have emerged as ideal charge-

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trapping layers owing to their large absorption coefficients, high saturation electron velocity, excellent thermal conductivity, and robust thermal and chemical stability [13–17]. However, insufficient optical absorption, pronounced carrier recombination, and limited intrinsic anisotropy restrict the use of high-resolution polarization photodetectors in the solar-blind UV regions.

The atomic-level thickness of WS_2 , a typical member of the transition metal chalcogenide (TMDC) family, results in an exceptional absorption coefficient and long carrier-diffusion length, making it attractive for high-performance photodetectors. The van der Waals (vdW) heterostructure band-alignment engineering further displays an atomically sharp interface, strong light-matter interactions, suppression of interfacial recombination, and ultrafast charge extraction [18–20]. Nevertheless, it remains challenging for two-dimensional heterostructure photodetectors to simultaneously exhibit the high dichroic ratios (DR), high photoresponsivity, ultralow dark current, and rapid responses. Consequently, dual-heterostructures and superlattices based on two-dimensional materials have been proposed to achieve high-performance multifunctional photodetectors [21–23]. Complete depletion of the intermediate absorbing layer in two parallel heterostructures not only reduces the transport distance of photocarriers but also significantly enhances the light absorption efficiency [22, 24]. Moreover, introducing a high electron barrier and large built-in electric field at the type-I dual-heterostructure interface ensures the rapid separation and transfer of photogenerated electron-hole pairs while effectively preventing the drift of minority carriers [25]. More importantly, the precisely engineered lattice mismatch at the dual interface breaks the intrinsic symmetry of the target material, giving rise to Moiré superlattices that substantially amplify the polarized electronic states. Consequently, synergistic band structure and symmetry engineering within wide-bandgap dual-heterostructures offer a potential pathway for realizing multifunctional solar-blind UV detectors with high performance and exceptional polarization sensitivity.

In this work, we constructed a novel vdW dual-heterostructure field effect transistor (FET) composed of a bottom monolayer WS_2 and middle multilayer GaPS_4 . Comprehensive characterization techniques, including optical microscopy (OM), photoluminescence (PL), atomic force microscopy (AFM), Kelvin probe force microscopy (KPFM), and high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM), were employed to evaluate the crystal quality. Ultrafast transient absorption (TA) spectroscopy and density functional theory (DFT) calculations were performed to elucidate the underlying carrier transport mechanisms in the $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure. Interestingly, two unique built-in electric fields were constructed at the dual-heterostructure interface, facilitating the efficient and rapid separation and transfer of photogenerated carriers. The device based on the $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure exhibits a low dark current, fast response time, and high dichroic ratio. Furthermore, this dual-heterostructure photodetector enables encoded optical communication and high-resolution polarization imaging. Our findings provide a promising route for developing high-performance and polarization-sensitive photodetectors in the solar-blind UV region based on two-dimensional dual-heterostructures.

2 Experimental

2.1 Preparation of GaPS_4 crystal and WS_2 nanosheets

High quality bulk GaPS_4 crystals were prepared by the chemical

vapor transport (CVT) method. Firstly, a mixture of gallium (Ga), phosphorus (P), and sulfur (S) powders with a molar ratio of 1:1:4 was loaded into a quartz tube. Subsequently, the temperature was gradually ramped up to 750 °C, held for one week, and finally was naturally cooled to room temperature to obtain GaPS_4 crystals. WS_2 nanosheets were synthesized by the chemical vapor deposition (CVD) method in tube furnace. The precursor S powder was placed in the center of the low temperature zone, while the WO_3 powder and substrate were placed in the center of the high temperature zone. The temperatures of the low and high temperature zones were simultaneously increased to 260 and 920 °C, respectively, within 30 min, maintained for 25 min, and then was naturally cooled to room temperature. WS_2 nanosheets were successfully obtained on the SiO_2/Si substrate.

2.2 Heterostructure fabrication

The vdW $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure was fabricated via an accurate transfer platform (Metatest, E1-G). The schematic diagram of the heterostructure fabrication process is displayed in Fig. S3 in the Electronic Supplementary Material (ESM). Multilayer GaPS_4 nanosheets were mechanically exfoliated from bulk GaPS_4 crystal using transparent tape. Next, a suitable GaPS_4 nanosheet was selected and was precisely transferred onto an adjacent monolayer WS_2 nanosheets on SiO_2/Si substrate. Finally, the $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure was heated at 80 °C for 10 min on a hot plate to enhance the interaction between the sample and substrate. Details regarding the STEM sample preparation are provided in Note S2 in the ESM.

2.3 Sample characterizations

The morphology of $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure was characterized by using OM (D4000M, Leica) and AFM (multimode 8, Bruker). Raman and PL spectroscopy of the $\text{WS}_2/\text{GaPS}_4$ heterostructure were investigated by a confocal Raman spectrometer (JY Horiba, HR Evolution). The phase structure, elemental composition, and valence states were analyzed by STEM (Talos F200X, Thermo Fisher) and X-ray photoelectron spectroscopy (XPS; ESCALAB 250Xi, Thermo Fisher Scientific). The micro-area UV-visible-near infrared (UV-Vis-NIR) absorption spectra were collected by a confocal spectrometer in the transmission mode (MStarter ABS, Metatest). The TA spectra were collected by using an integrated Helios setup (Ultrafast Systems). Femtosecond laser pulses were generated by a Ti:Sapphire amplifier at a repetition rate of 1 kHz (Spectra-Physics Solstice Ace). The 800 nm fundamental passed through a beam splitter, where most of the beam was sent into an optical parametric amplifier (Ultrafast Systems, Apollo-T) to generate 400 and 267 nm pulses for the pump. The rest of the fundamental was focused into a Vis continuum-generation crystal (Ultrafast, Helios) to generate the white-light continuum.

2.4 Device fabrication and measurements

The $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure devices were fabricated through a series of processes including laser direct writing lithography, metal thermal evaporation, and lift-off processes. The electrical and optoelectronic illuminated data were recorded using Keithley 2634B dual source meter. The optoelectrical measurement of $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure photodetector was conducted using the metatest scanpro laser scanning system (ScanPro Advance, Metatest) with a wavelength-adjustable light-emitting

diode (LED) (255–1550 nm) as the light source. Time-resolved photo-response data were collected with a lock-in amplifier (Dlpca200, Femto) equipped with an optical chopper. The degree of linear polarization (DoLP) can be calculated by using the following equations

$$S_0 = I_{0^\circ}(x, y) + I_{90^\circ}(x, y) \quad (1)$$

$$S_1 = I_{0^\circ}(x, y) - I_{90^\circ}(x, y) \quad (2)$$

$$S_2 = I_{45^\circ}(x, y) - I_{135^\circ}(x, y) \quad (3)$$

$$\text{DoLP} = \frac{\sqrt{S_1^2 + S_2^2}}{S_0} \quad (4)$$

where S_0 , S_1 , and S_2 represent the total intensity, the first Stokes parameter, and the second Stokes parameter, respectively. $I_{0^\circ}(x, y)$, $I_{45^\circ}(x, y)$, $I_{90^\circ}(x, y)$, and $I_{135^\circ}(x, y)$ denote the intensity images acquired through linear polarizers oriented at 0° , 45° , 90° , and 135° , respectively, where (x, y) indicates the spatial coordinates in the image plane.

2.5 DFT calculation

First-principles calculations were performed using the projector-augmented wave (PAW) method, as implemented within the Vienna *ab-initio* simulation package (VASP). The exchange-correlation interactions were modeled using the Perdew–Burke–Ernzerhof (PBE) functional, which is situated within the generalized gradient approximation (GGA) paradigm. To account for vdW interactions, Grimme's DFT-D3 approach was adopted. To mitigate artificial interactions between neighboring images in the simulations of two-dimensional and few-layer $\text{WS}_2/\text{GaPS}_4$ structures, a vacuum spacing of 15 Å was introduced along the z -axis. After conducting a series of convergence studies, an energy cutoff of 650 eV was selected for the plane waves, and the first Brillouin zone was sampled using a Monkhorst–Pack k -point mesh with a spacing of $2\pi \times 0.03 \text{ Å}^{-1}$. The structural relaxation process was concluded when both the total energy changes and the ionic Hellmann–Feynman forces fell below stringent thresholds of $5.0 \times 10^{-6} \text{ V}$ and 0.01 eV/Å , respectively.

3 Results and discussion

Schematics of the crystal model and an optical image of the $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure are shown in Fig. 1(a). A typical $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure is fabricated with GaPS_4 nanosheets bridging two neighbouring monolayer WS_2 nanosheets. Further details on the experimental growth and transfer exfoliation processes are presented in Section 2 and Figs. S1–S4 in the ESM. The optical image, AFM image, and corresponding height profiles of pure GaPS_4 nanosheets (Fig. S5 in the ESM) highlight the features of a typical layered crystal structure. Figures 1(b) and 1(c) show the Raman and PL spectra of the individual WS_2 , $\text{WS}_2/\text{GaPS}_4$, and GaPS_4 heterostructure regions. In the WS_2 region, the major Raman peaks at 175, 230 296, 323, 351, 355, and 419 cm^{-1} belong to the LA(M), A1(M)–LA(M), $E''(\text{G})$, $E''(\text{M})$, 2LA(M), $E'(\text{G})$, and A_{1g} vibration modes, respectively, which is consistent with previously obtained results [26]. For the heterostructure region, eight additional peaks, A_g^1 (71.9 cm^{-1}), A_g^2 (81.6 cm^{-1}), A_g^3 (110.6 cm^{-1}), B_g (119.8 cm^{-1}), A_g^4 (149.3 cm^{-1}), A_g^5 (189.8 cm^{-1}), A_g^6 (381.4 cm^{-1}), and

A_g^7 (427.7 cm^{-1}), are attributed to the Raman peak vibrational modes of GaPS_4 nanosheets [9]. The PL spectrum (Fig. 1(c)) shows a strong emission peak located at 1.96 eV, indicative of the direct bandgap of monolayer WS_2 [26]. However, the $\text{WS}_2/\text{GaPS}_4$ heterostructure shows a significant PL quenching effect and the peak is red-shifted, suggesting efficient separation and transfer of the photogenerated electron–hole pairs [27]. The thickness-dependence of the Raman and PL intensities of the $\text{WS}_2/\text{GaPS}_4$ heterostructure is shown in Fig. S6 in the ESM. The uniform Raman and PL intensity mappings (Fig. S7 in the ESM) further confirm the high quality of as-prepared $\text{WS}_2/\text{GaPS}_4$ heterostructure. The absorption spectra of WS_2 , $\text{WS}_2/\text{GaPS}_4$, and GaPS_4 are shown in Fig. 1(d). The blue arrows located at 617, 514, and 427 nm belong to the A, B, and C excitons of pure WS_2 , respectively, while the red arrow at 265 nm is attributed to the exciton peak of GaPS_4 , which is consistent with a previous report [9]. Notably, within the overlapping region of the heterojunction, the absorption is dramatically enhanced in the solar-blind UV region, implying the potential for high-performance solar-blind photodetection. The chemical composition and atomic-scale stacking structure of the dual-heterostructure were further investigated by aberration-corrected STEM (Ac-STEM). The HAADF images and elemental mapping (Fig. S8 in the ESM) show a uniform distribution of Ga, P, S, and W, indicating the high-quality stacking structure of the $\text{WS}_2/\text{GaPS}_4$ heterostructure. The selected area electron diffraction (SAED) pattern (Fig. 1(e)) reveals two sets of diffraction spots, corresponding to the hexagonal monolayer WS_2 and monoclinic GaPS_4 crystal, with an orientation relationship of GaPS_4 [201]// WS_2 [001], as verified by X-ray diffraction (XRD) and SAED. The atomic-scale HAADF-STEM image (Fig. 1(f)) of the $\text{WS}_2/\text{GaPS}_4$ heterostructure shows a lattice spacing of 0.76 nm, assigning to the (204) plane of the GaPS_4 crystal. Figure S9 in the ESM shows XPS spectra, confirming the binding energies of W 4f, Ga 2p, P 2p, and S 2p, which agree with those previously reported [9, 27]. Figures 1(g) and 1(h) exhibit an AFM image and the corresponding KPFM image, respectively. The corresponding height profiles (Fig. 1(i), top) indicate the thicknesses of 0.7 nm for WS_2 and 20.3 nm for GaPS_4 , while the surface potential of the GaPS_4 in the middle region is higher than that of the WS_2 and the difference is calculated to be 106 mV (Fig. 1(i), bottom). Notably, two distinct spike-shaped surface potential features are clearly observed in the $\text{WS}_2/\text{GaPS}_4$ dual-heterostructure region, implying the formation of a larger built-in potential at the $\text{WS}_2/\text{GaPS}_4$ interface. The thickness-dependent work function of GaPS_4 suggests the possibility of tuning the barrier height by controlling the number of layers (Fig. S10 in the ESM).

Ultrafast TA spectroscopy and DFT calculations were employed to further investigate the electron–hole pair transfer between WS_2 and GaPS_4 . The real-time photoexcited exciton dynamics within WS_2 and the $\text{WS}_2/\text{GaPS}_4$ heterostructure are directly monitored by TA spectroscopy using pump pulses lasers at 267 and 400 nm (a detailed schematic of the femtosecond TA experimental setup is shown in Fig. S11 in the ESM). As depicted in Figs. 2(a) and 2(b), the TA spectra of pristine WS_2 pumped at 267 nm exhibits two negative signals located at 617 and 514 nm of the ground state bleach (GSB), corresponding to the A and B excitons of WS_2 [28], respectively. A 400 nm pump pulse laser is chosen to selectively excite WS_2 while keeping GaPS_4 in an unexcited state. The GSB signal intensity of A and B excitons in the heterostructure region is significantly weaker than those observed for pristine WS_2 , as shown

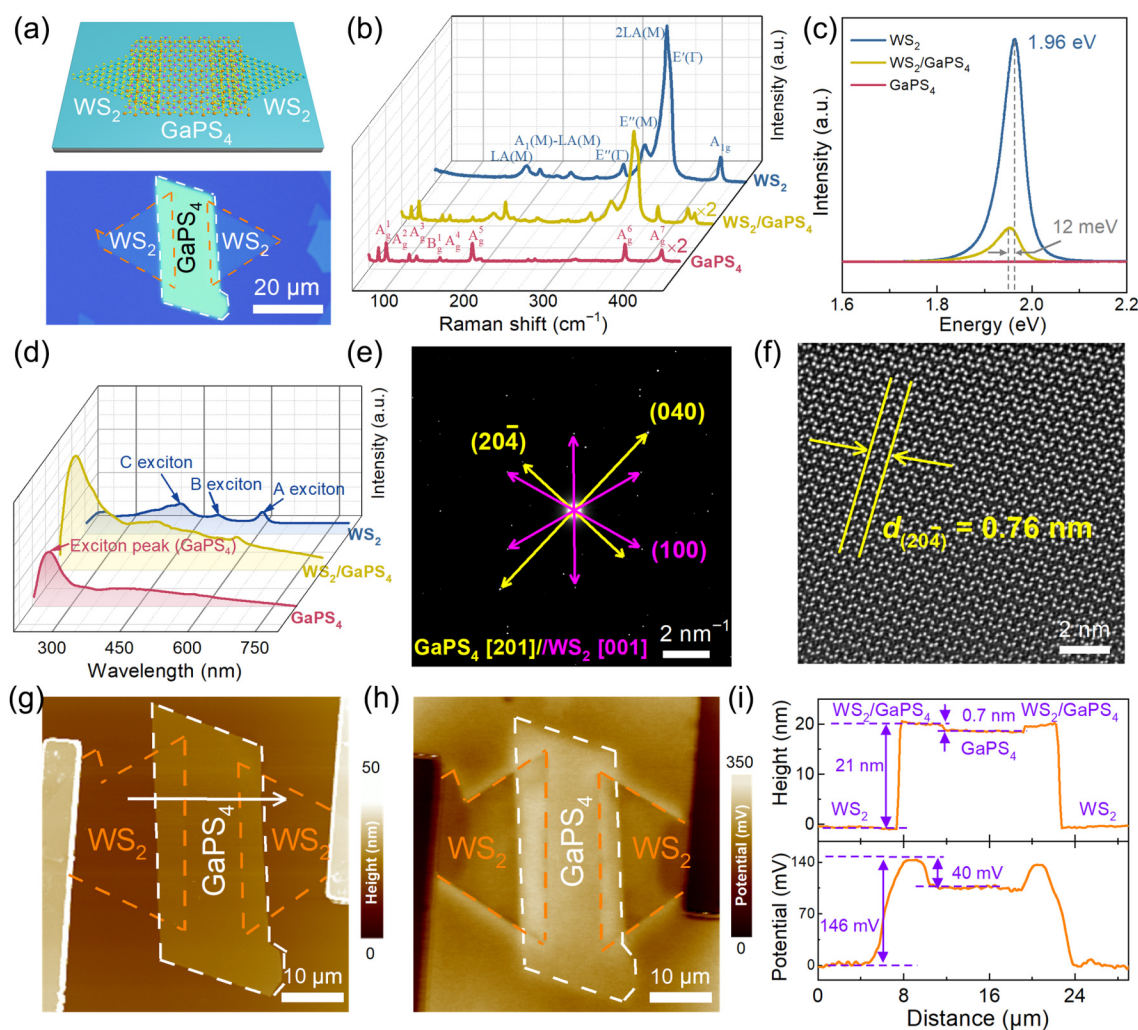


Figure 1 OM, Raman, PL, TEM, AFM, KPFM characterization of WS₂/GaPS₄/WS₂ heterostructures. (a) Schematic diagrams of (top) crystal model and (bottom) optical microscopy image for WS₂/GaPS₄ dual-heterostructure fabricated on the SiO₂/Si substrate. The GaPS₄ layer is located at top of two adjacent monolayer WS₂ nanosheets. ((b) and (c)) Raman and PL spectra collected from WS₂, WS₂/GaPS₄, and GaPS₄ under 532 nm laser excitation. The LA(M), A₁(M)-LA(M), E'(T), E'(M), 2LA(M), E'(T), and A_{1g} vibrational peaks are attributed to WS₂, whereas the A₁^s, A₂^s, A₃^s, B₂^s, A₄^s, A₅^s, A₆^s, and A₇^s are attributed to GaPS₄. The PL peak of the pure monolayer WS₂ is located at 1.96 eV, whereas the PL peaks in the heterostructure region are greatly diminished and shifted, indicating efficient photogenerated electron-hole pairs separation and transfer. (d) The absorption spectra of WS₂, WS₂/GaPS₄, and GaPS₄. The blue arrow marks located at 617, 514, and 427 nm belong to A exciton, B exciton, and C exciton of pure WS₂, respectively. The red arrow mark located at 265 nm is attributed to the exciton peak of GaPS₄. ((e) and (f)) The SADE pattern and corresponding HADDF-STEM image of WS₂/GaPS₄ heterostructure. ((g) and (h)) Typical AFM and KPFM images of WS₂/GaPS₄ dual-heterostructure, respectively. From the center to the sides: GaPS₄, WS₂/GaPS₄, WS₂, and Au. (i) (top) The corresponding height profiles and (bottom) the surface potential difference WS₂/GaPS₄ dual-heterostructure.

in Fig. S12 in the ESM. Conversely, the GSB signal intensity for A and B excitons pumped by 267 nm in the heterostructure region exhibits markedly higher intensities than that of pristine WS₂, providing unambiguous evidence that electrons and holes photogenerated in GaPS₄ are quantitatively transferred into the WS₂ layer. In addition, the TA spectra recorded 1 ps after 267 nm photoexcitation (Fig. 2(c)) show state-filling in GaPS₄, driven by fast electron-hole injection from GaPS₄ into WS₂. DFT calculations are used to explain the charge transfer mechanism between WS₂ and GaPS₄.

Figure 2(d) presents the electronic band structure of the WS₂/GaPS₄ heterostructure. The valence band maximum (VBM) is primarily composed of W and S atoms from WS₂, whereas the conduction band minimum (CBM) consists mainly of the Ga and S atoms from GaPS₄, with minor contributions from P atoms (Fig. 2(e)), suggesting that the orbital hybridization predominantly

occurs in the non-band-edge region [29]. Moreover, the CBM of the WS₂ is lower than that of the GaPS₄, whereas the VBM of WS₂ is higher than that of GaPS₄, confirming the type-I band alignment (Fig. 2(f)), which is consistent with the TA observations.

Moreover, the symmetry engineering strategy, achieved by incorporating GaPS₄ (C₂ symmetry) into monolayer WS₂ (C₃ symmetry), triggers pronounced interfacial effects of orbital hybridization and uniaxial lattice distortion, which breaks the intrinsic C₃-symmetric lattice of pristine WS₂ [27]. Angle-resolved polarization Raman spectroscopy (ARPRS) and angle-resolved polarization absorption spectroscopy (ARPAS) further reveal a markedly enhanced anisotropic optical response at the WS₂/GaPS₄ dual-heterostructure interface. Figure 3(a) illustrates the experimental geometry of the ARPRS results in parallel and cross configurations. Owing to the C₃ rotational symmetry of pristine WS₂, the Raman-active modes exhibit isotropic intensity

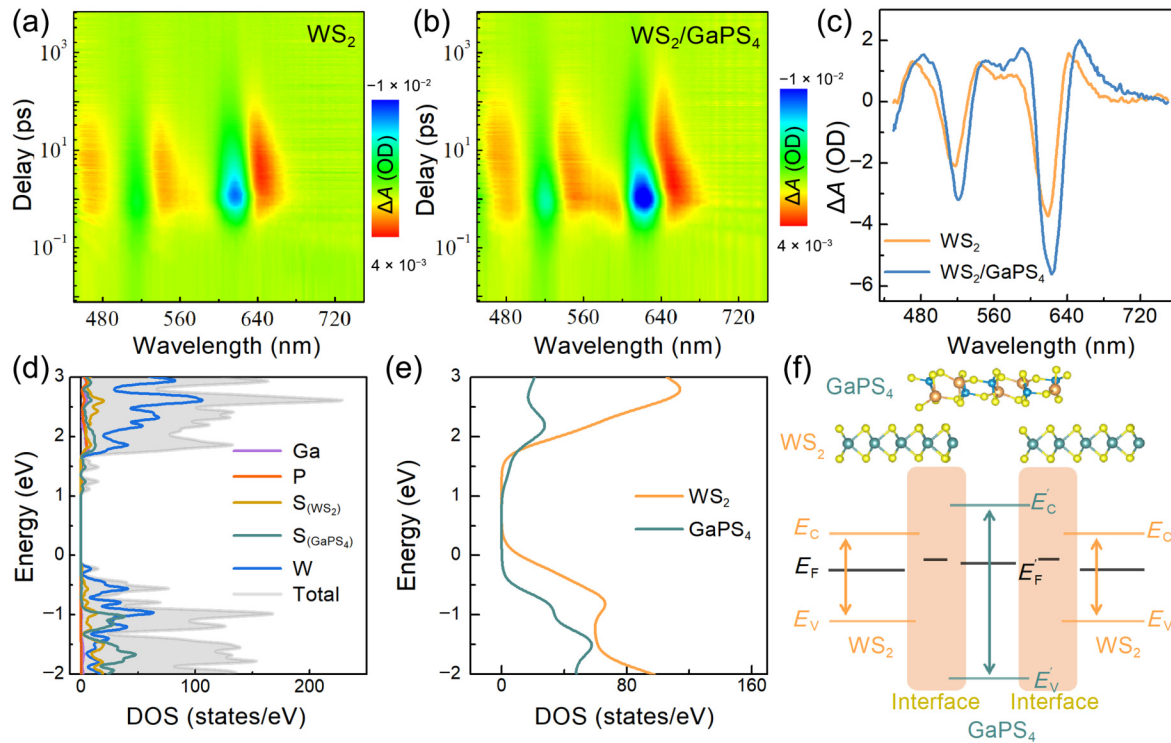


Figure 2 Transient absorption spectroscopy characterization and theoretical calculation of $WS_2/GaPS_4$ dual-heterostructure. ((a) and (b)) Pristine WS_2 and $WS_2/GaPS_4$ heterostructures under 267 nm pump pulses laser. (c) TA spectra extracted from the mappings in ((a) and (b)) WS_2 and $WS_2/GaPS_4$ heterostructures at 1 ps delay time. (d) Partial density of states (PDOS) based on Ga, P, S(WS_2), S($GaPS_4$) and W atoms. (e) DOS projected to $GaPS_4$ and WS_2 . (f) Band alignment of $WS_2/GaPS_4$ dual-heterostructure before contact.

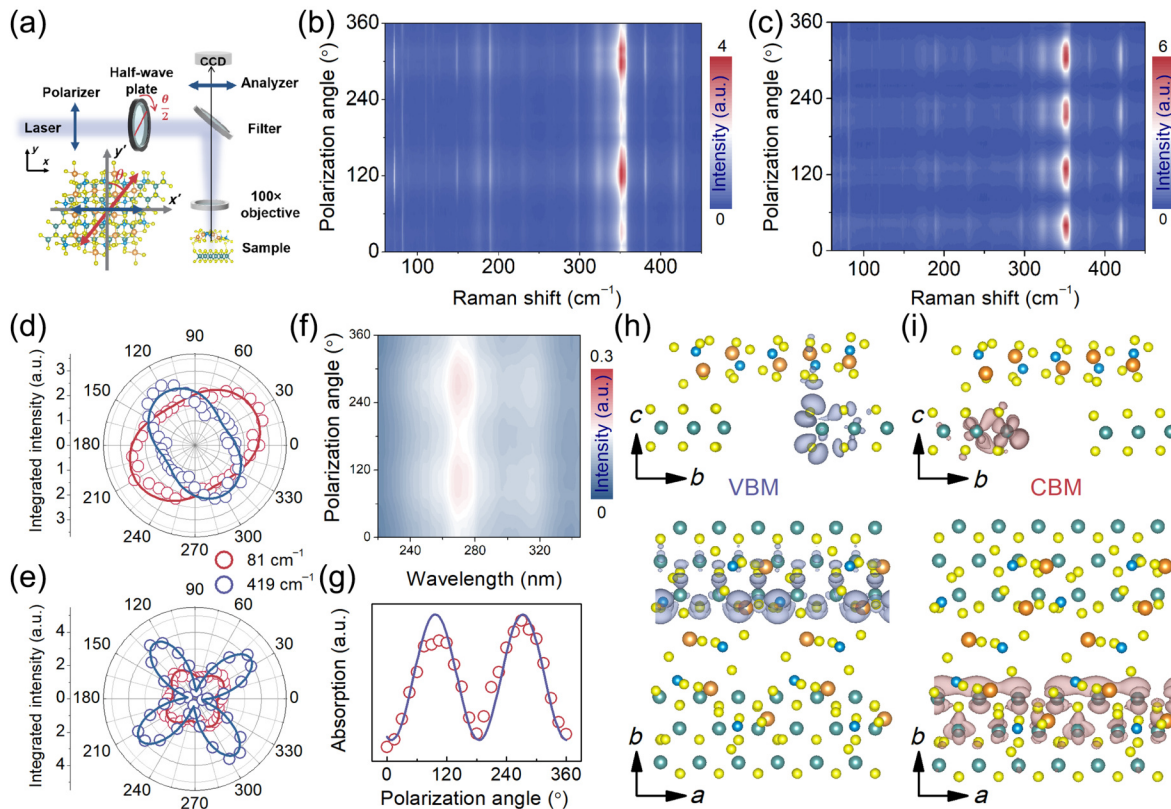


Figure 3 Anisotropic optical response of $WS_2/GaPS_4$ heterostructure interface. (a) Schematic diagrams of the angle-resolved polarized Raman spectroscopy. (b) and (c) Polarized Raman intensity mappings of $WS_2/GaPS_4$ heterostructure under parallel and cross configurations. (d) and (e) Polar plots of the measured and fitted peak intensities under parallel and cross configuration based on $WS_2/GaPS_4$ heterostructure in 81 and 419 cm^{-1} . (f) and (g) Contour mapping and extracted 267 nm exciton peak of ARPRS under parallel configurations. (h) and (i) The spatial distributions of the VBM and CBM wave functions in the bc - and ab -planes.

distributions as the polarization angle changed [27]. Figures 3(b) and 3(c) display the contour mapping of angle-dependent Raman intensity under parallel and cross configurations for WS₂/GaPS₄ heterostructure. By introducing the low-symmetry GaPS₄ layer (C₂ symmetry), the interfacial interaction breaks the three-fold symmetry of WS₂, rendering the Raman modes linearly polarized. Remarkably, the Raman characteristic peaks of WS₂/GaPS₄ heterostructure region display identical periodicities of π and $\pi/2$ (Figs. 3(d) and 3(e)), providing the evidence that the symmetry of WS₂ is effectively re-engineered by the adjacent GaPS₄ through tunable interlayer coupling. Detailed quantitative analyses are provided in Fig. S13 and Note S1 in the ESM. Similar results are observed in the ARPAS (Figs. 3(f) and 3(g)). The excitonic peak of GaPS₄ at 267 nm is significantly enhanced by the incorporation of WS₂ and exhibits π -periodic oscillations with polarization angle, indicating potential application solar-blind UV polarization photodetector. Moreover, the partial charge density (PCD) was calculated to evaluate the electron density distribution at the

WS₂/GaPS₄ dual-heterostructure interface. The spatial distributions of the VBM (Fig. 3(h)) and CBM (Fig. 3(i)) wave functions show clear distinctions between the *ab*- and *bc*-planes, suggesting the intrinsic in-plane anisotropic optical response. Notably, the asymmetrical and non-uniform charge density distribution at the dual-heterostructure interfaces effectively promotes the anisotropic charge separation in the built-in electric field, significantly enhancing the in-plane optical anisotropy of WS₂/GaPS₄ dual-heterostructure.

To investigate the optoelectronic properties of WS₂/GaPS₄ dual-heterostructure, a back-gate FET was fabricated on SiO₂/Si substrate (Fig. 4(a)). The transfer characteristics curve (Fig. 4(b)) reveals typically n-type conductive behavior with an on/off ratio up to 10⁶ and ultralow leakage currents (10⁻¹³ A). Figure 4(c) shows *I*_{ds}-*V*_{ds} (*I*_{ds} and *V*_{ds} are the source-drain current and voltage, respectively) curves under dark and 255 nm laser illumination at different power intensities. An increase of *I*_{ds} is observed as the power density increase from 2.8 × 10⁻¹ to 1.7 × 10³ mW/cm², suggesting that the

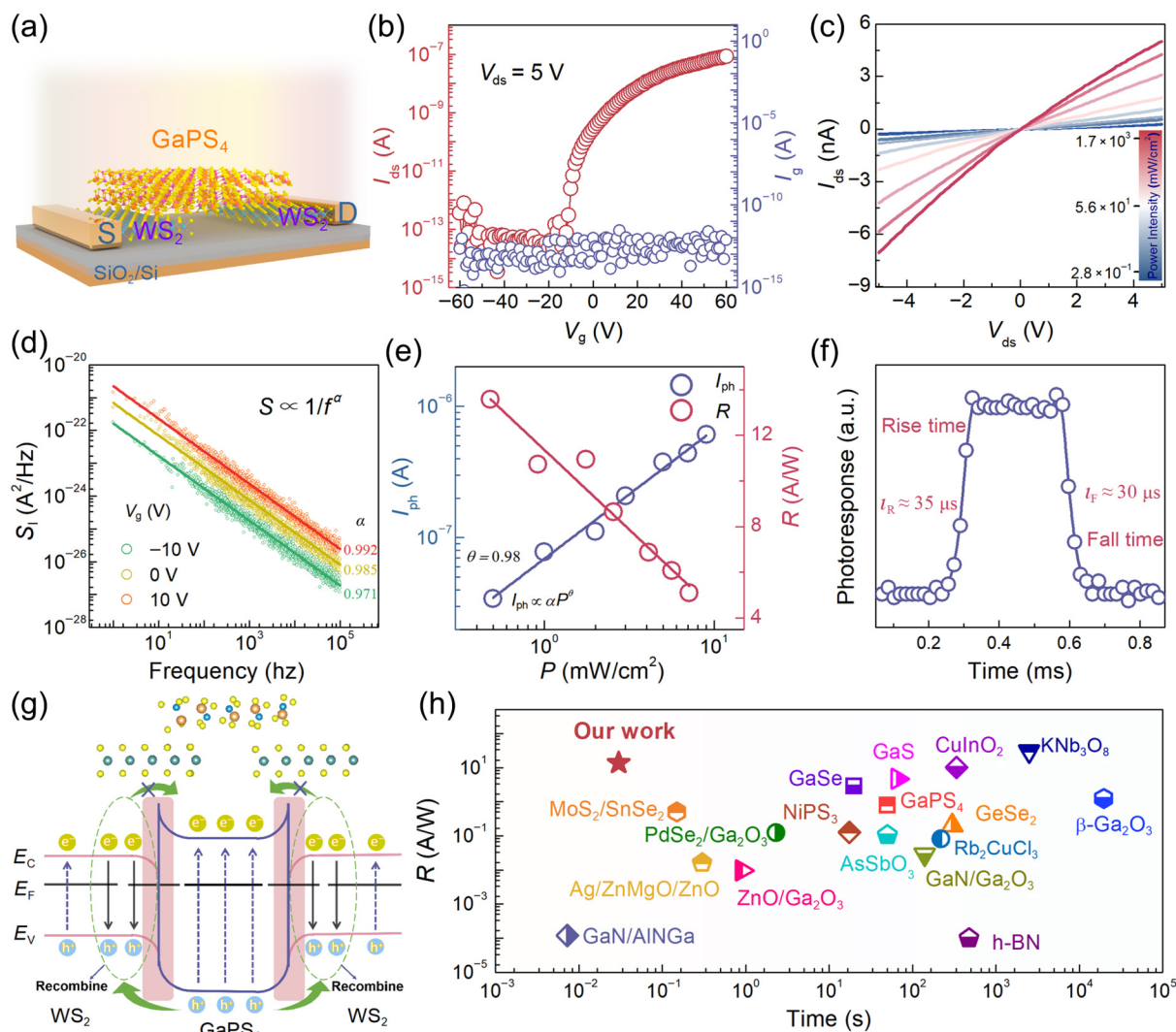


Figure 4 Photoelectric performance of WS₂/GaPS₄ dual-heterostructure photodetector. (a) Schematic illustration of WS₂/GaPS₄ dual-heterostructure device under illuminating with various wavelengths. (b) The representative plots of *I*_{ds} and leakage current (*I*_g) as function of *V*_g range from 60 to -60 V, implying high on/off ratios as 10⁶ and low leakage currents (10⁻¹³ A). (c) *I*_{ds}-*V*_{ds} curves of WS₂/GaPS₄ dual-heterostructure device in the dark and under irradiation with different power density under 255 nm illuminations. (d) Noise spectral density of the device under different *V*_g at *V*_{ds} of 5 V. (e) Photoelectric performances metrics of *I*_{ph} and *R* as function of power intensity under 255 nm light illuminations. (f) Rise and fall time of WS₂/GaPS₄ dual-heterostructure device. (g) Energy band diagram illustrating the photodetection mechanism of the WS₂/GaPS₄ dual-heterostructure. (h) Comparison of response time and responsivity in the solar-blind UV photodetector based on other materials.

generation of photogenerated carriers is proportional to the number of absorbed incident photons [30]. Time-dependent photoresponse of WS₂/GaPS₄ dual-heterostructure photodetector under different power intensities and wavelengths of 255 and 360 nm is displayed in Figs. S14 and S15 in the ESM, indicating a stable photoresponse in solar-blind UV region. Moreover, the frequency-dependent noise spectral density (S_i) is shown in Fig. 4(d), which can be fitted by the equation $S_i \propto (1/f)^\alpha$. The exponent α is calculated to be 0.971, 0.985, and 0.992 under gate voltages (V_g) of -10, 0, and 10 V, respectively, indicating that $1/f$ noise dominates the total noise of dual-heterostructure photodetector at low frequencies [31]. Figure 4(e) displays the photocurrent ($I_{ph} = I_{light} - I_{dark}$, where I_{light} and I_{dark} are the current under illumination and the dark current, respectively) and responsivity (R) as function of power intensity under 255 nm illumination. Notably, a nonlinear photoconductivity effect in WS₂/GaPS₄ dual-heterostructure photodetector is observed. Generally, I_{ph} can be fitted by the following equation: $I_{ph} \propto aP^\theta$, where a is a constant, P is laser power intensity, and θ is power factor ($\theta < 1$). The maximum I_{light}/I_{dark} , R , and external quantum efficiency (EQE) for the photodetector can be calculated as 3.6×10^3 , 1.3×10^4 A/W, and $6.6 \times 10^3\%$ under 255 nm illumination, respectively (Figs. S16(a) and S16(b) in the ESM), which are higher than those of pure GaPS₄ [9], WS₂/GaPS₄ heterostructure (Fig. S17 in the ESM), GaSe [32], and ZnO-Ga₂O₃ [33]. As depicts in Fig. 4(f), the device exhibits the rise and fall time of 35 and 30 μ s, respectively, which are attributable to the rapid separation of photocarriers and fast charge carrier transfer at the WS₂/GaPS₄ dual-heterostructure interface. Moreover, the photocurrent of WS₂/GaPS₄ dual-heterostructure photodetector shows no degradation after periodic on/off switching of the light for 10^4 cycles (Fig. S18 in the ESM), implying excellent durability and repeatability. Notably, the dual-heterostructure demonstrates remarkable operational stability across a wide temperature range from 163 to 473 K (Figs. S19 and S20, and Note S4 in the ESM). A comprehensive model with energy-band alignment is used to explain the mechanism underlying the nonlinear photocurrent behaviour (Fig. 4(g)). The WS₂/GaPS₄/WS₂ dual-heterostructure facilitates the efficient spatial separation of electron-hole pairs driven by the dual built-in electric fields. Here, GaPS₄ acts as a barrier layer, confining photogenerated carriers within the narrow-bandgap WS₂. Although holes flow unimpeded across the interface, electrons accumulate on the WS₂ side because of the potential barrier in the conduction band. This configuration significantly inhibits the electron-hole recombination and enhances the carrier collection efficiency, thereby boosting the photoelectric performance of the device. These results (Fig. 4(h)) highlight the superior performance of the WS₂/GaPS₄ dual-heterostructure photodetector compared to previously reported solar-blind UV photodetectors (see Table S1 in the ESM for a detailed comparison of merit values). Taking into consideration the ultra-low dark current, broad spectral sensitivity, and fast response time of the WS₂/GaPS₄ dual-heterostructure photodetector, its potential application in secure optical communications is explored through a photoelectric scanning system (see Fig. S21 and Note S3 in the ESM for details of optical communication application). Therefore, the outstanding performance of WS₂/GaPS₄ dual-heterostructure photodetector resulted in a strong double built-in electric field and high-spike barrier height, contributing to reduction in the

recombination rate of electron-hole pairs and preventing electron transfer from WS₂ to GaPS₄.

The breaking of the interfacial symmetry between C₃-symmetric WS₂ and C₂-symmetric GaPS₄ substantially amplified the anisotropic optical absorption, endowing it with strong potential for polarization-sensitive solar-blind photodetection. As schematically depicted in Fig. 5(a), angle-resolved photocurrent measurements for the WS₂/GaPS₄ dual-heterostructure were performed using a continuously rotated half-wave plate positioned between a 255 nm light source and the device to sweep the linear-polarization vector. Time-resolved photoresponse curves (Fig. 5(b)) reveal the periodic modulation of the photocurrent ratio (I_{ph}/I_{dark}) with polarization angle. The polar-coordinates of the angle-dependent photocurrent (Fig. 5(c)) exhibit a pronounced two-lobed pattern. Consequently, the dichroic ratio (I_{max}/I_{min}) is calculated to be 3.4 under 255 nm illumination, which is higher than that of pristine GaPS₄ (1.85) and those of some other previously reported anisotropic materials in UV range, as summarized in Fig. 5(d). Benefiting from the ultralow dark current, the ultrafast response speed, and the high dichroic ratio, the polarization-resolved imaging capability of WS₂/GaPS₄ dual-heterostructure is evaluated using a single-pixel scanning system (Fig. 5(e)). A mask engraved with the "AHUMFMD" pattern (AnHui University, Magnetic Functional Materials and Device Laboratory, Fig. S22 in the ESM) was positioned between a half-wave plate and the detector, and its linear motion was controlled by an X-Y stage. The reconstructed "AHUMFMD" (61 pixels $\times$ 44 pixels) generated from polarization photocurrent data is displayed in Fig. 5(f). Each pixel within this matrix represents a specific polarization photocurrent intensity, which is assigned to corresponding color blocks. Using image processing techniques, these color blocks are combined to create the "AHUMFMD" image, while the computer records the photocurrent data. Figure 5(g) illustrates a schematic of the polarization imaging mechanism for calculating DoLP (see the detailed analysis in Section 2). The polarization images acquired at angles of 0°, 45°, 90°, and 135° are displayed in Fig. S23 in the ESM. The extracted imaging results of WS₂/GaPS₄ dual-heterostructure photodetector exhibit significantly enhanced contrast (Fig. 5(h)), demonstrating its exceptional polarization imaging capability.

4 Conclusions

The WS₂/GaPS₄ dual-heterostructure photodetector was successfully constructed by bridging the wide-bandgap semiconductor GaPS₄ in two adjacent n-type semiconductor WS₂ nanosheets. This unique design introduces a high electron barrier and a large built-in electric field at the WS₂/GaPS₄ dual-heterostructure interface, which not only ensures the efficient separation and transfer of photogenerated electron-hole pairs but also effectively suppresses the drift of minority carriers. In addition, the optical anisotropy of WS₂/GaPS₄ dual-heterostructure in the solar-blind UV region is significantly enhanced via symmetry-breaking engineering. The optoelectronic performance of WS₂/GaPS₄ dual-heterostructure photodetector in the solar-blind UV region exhibits a high R of 13.5 A/W, an EQE of $6.6 \times 10^3\%$, and the photoresponse time of approximately 30 μ s. Additionally, the in-plane optical anisotropy of the WS₂/GaPS₄ dual-heterostructure exhibits a high dichroic ratio up to 3.4 under 255 nm illumination. These results indicate that the WS₂/GaPS₄ dual-heterostructure device with polarization-imaging capabilities

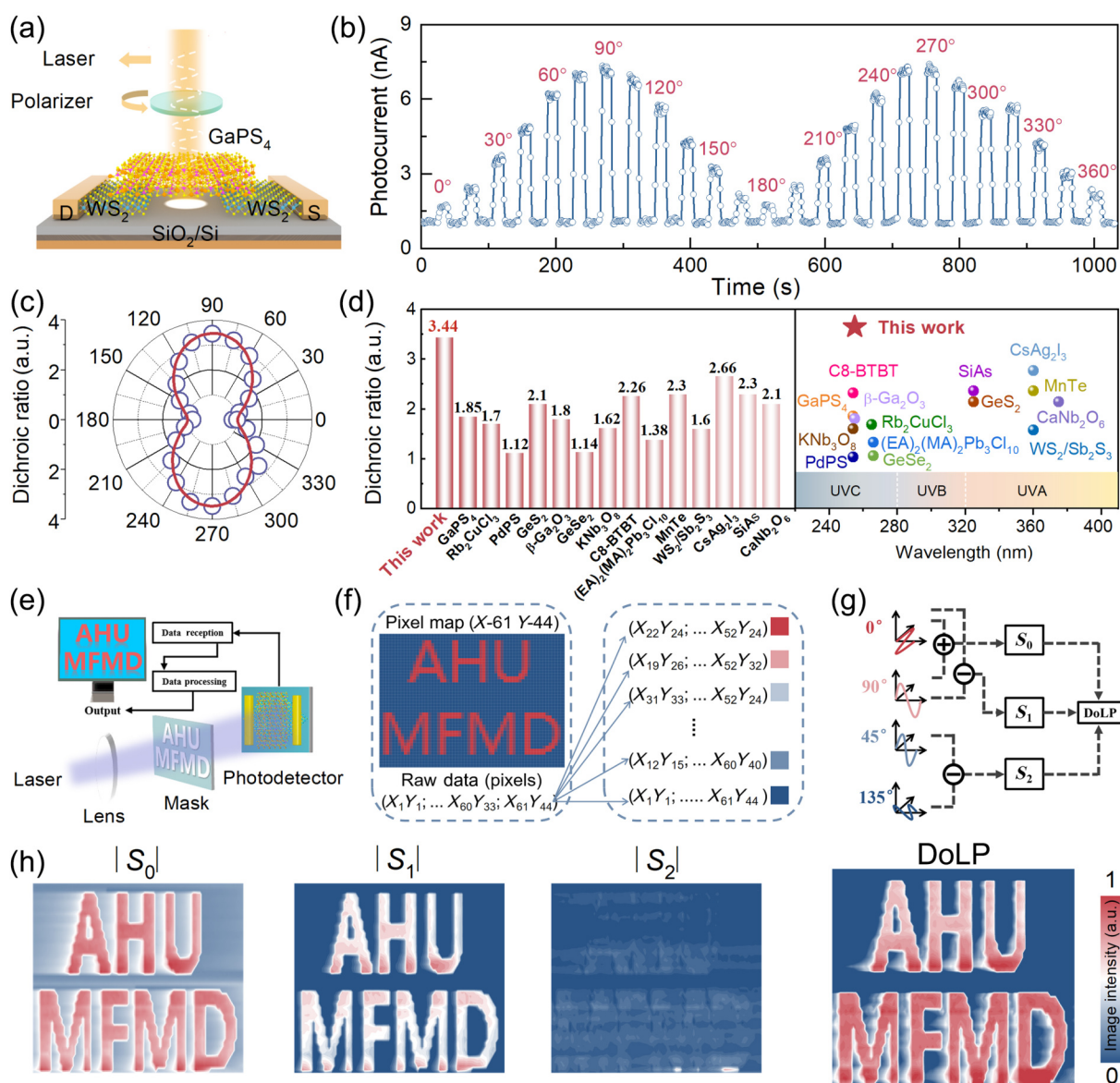


Figure 5 Polarized optoelectronic responses and polarization imaging applications of the WS₂/GaPS₄ dual-heterostructure device in solar-blind UV region. (a) The schematic view of WS₂/GaPS₄ dual-heterostructure polarized photodetection measurement. (b) Time-resolved photocurrent of WS₂/GaPS₄ dual-heterostructure device with different polarization angles with a 15° step under 255 nm illumination. (c) Plot of normalized dichroic ratio of the WS₂/GaPS₄ dual-heterostructure device. The DR is defined as $\frac{I_{ph, \max}}{I_{ph, \min}}$, where $I_{ph, \max}$ and $I_{ph, \min}$ represent the maximum and minimum photocurrents under polarization angles, respectively. (d) Photocurrent dichroic ratio comparison based on different materials, including GaPS₄ [9], KNb₃O₈ [10], Rb₂CuCl₃ [11], (EA)₂(MA)₂Pb₂Cl₁₀ [34], β-Ga₂O₃ [35], GeSe₂ [36], PdPS [37], C8-BTBT [38], GeS₂ [39], MnTe [40], WS₂/Sb₂S₃ [27], CsAg₂I₃ [41], SiAs [42], and CaNb₂O₆ [43]. (e) Schematic diagram of the measurement setup for an imaging sensor. (f) Real-time collection of raw photocurrent data and corresponding color mapping. (g) Schematic of the polarization imaging mechanism for the calculation of DoLP. (h) Imaging DoLP results for dual-heterojunction device using 255 nm laser illumination.

and polarization contrast enhancement has significant potential for applications in highly integrated electronics and optoelectronics in the solar-blind UV region.

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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 report no conflict of interests in this work.

Author contribution statement

Q. L., K. Y., and M. M. Y. designed the experiments, analyzed the data, and wrote the paper. J. X. Y., T. L. Z., and Q. L. grew the samples and fabricated the devices. K. Y., Y. H. G., Y. L., and B. H. conducted Raman, PL spectroscopy, and noise spectral measurements. Y. G., Q. G., and P. X. performed the XRD and AFM measurements. A. M. N. and J. X. Y. carried out the TEM characterizations. Q. Q. Y., R. Q. C., and Z. F. S. helped with the TA spectroscopy test. K. Y. and Q. L. conducted photo-detection testing. Q. G. and W. Y. conducted DFT calculation. M. M. Y., W. Y., Z. Y. L., and S. G. W. contributed to the discussion and analysis of experimental data. All the authors have approved the final manuscript.

Use of AI statement

None.

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