

Self-driven and ultrabroadband polarized photoresponse in BP/Nb₂SiTe₄ heterostructures for selective polarization-based encryption-decryption and recognition

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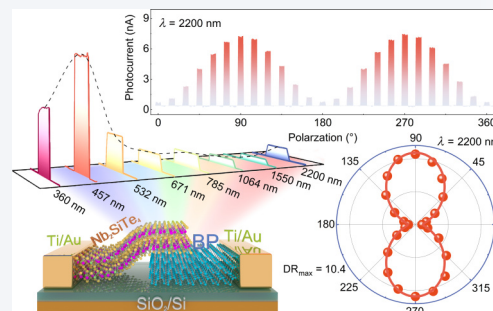
ABSTRACT: Being able to probe the polarization states in low-symmetry two-dimensional (2D) crystals is crucial for applications in polarization-encoded encryption and decryption. However, their polarization sensitivity is restricted by the inherent optical anisotropy. Here, we demonstrate a strategy where black phosphorus (BP) is stacked on Nb₂SiTe₄ to form a van der Waals heterostructure, realizing a self-powered, broadband, and highly polarization-sensitive photodetector. The atomically sharp and clean interface of BP/Nb₂SiTe₄ heterostructures forms a strong built-in electric field, which facilitates the separation of electron-hole pairs. Furthermore, angle-dependent Raman and absorption intensities reveal significant anisotropic optical response. The BP/Nb₂SiTe₄ photodetector exhibits a rapid response time and broadband detection capability from 360 to 2200 nm due to the efficient charge separation and strong interlayer coupling at the heterostructure interface. Notably, the BP/Nb₂SiTe₄ device also exhibits a large dichroic ratio (10.4) at zero voltage under 2200 nm light illumination. Furthermore, the proof-of-concept BP/Nb₂SiTe₄ photodetector possesses polarization-resolved imaging and polarization-encoded encryption/decryption capabilities, providing new insights for achieving low-power and multifunctional polarization-sensitive photodetectors.

KEYWORDS: van der Waals heterostructure, anisotropic optical response, self-driven and broadband photodetector, polarization sensitivity, polarization-encoded encryption and decryption

1 Introduction

Two-dimensional (2D) materials exhibit exceptionally high carrier

mobilities, tunable bandgaps, pronounced light-matter interactions, large specific surface areas, and abundant surface-active sites, making them compelling platforms for optoelectronics, sensing, photocatalysis, as well as energy storage and conversion [1, 2]. Critically, the realization of wafer-scale synthesis of vertically stacked and atomically thin 2D heterostructures via chemical vapor deposition (CVD) or physical vapor deposition (PVD) opens unprecedented avenues for developing ultralow-power and ultrabroadband photodetectors [3, 4]. Recently, the emergence of low-symmetry two-dimensional materials has introduced an



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additional polarization degree of freedom, offering a transformative paradigm for the miniaturization and functional diversification of polarization devices [5].

The low-symmetry 2D materials exhibit a rich structural polymorphism, encompassing monoclinic, orthorhombic, and triclinic lattices that manifest distinct point-group symmetries and correspondingly diverse anisotropic responses [6]. The puckered layered structures, such as black phosphorus (BP), GeS, and SnSe, are explicitly assigned to the D_{2h} point group [7–9]. In contrast, a distorted octahedral framework of ReS₂ or ReSe₂, stemming from Peierls instability is classified within the triclinic crystal structure (space group $P\bar{1}$) [10, 11]. These low-symmetry two-dimensional crystals, exhibiting pronounced polarization-resolved optical absorption and carrier mobility, are revealing the most promising candidates for next-generation polarization-sensitive electronics and optoelectronics. However, the intrinsic in-plane anisotropy of two-dimensional lattices is fundamentally limited by their native symmetry. To overcome this limitation, these novel strategies, including electric field tuning, strain engineering, surface plasmon resonance, and symmetry breaking engineering, have been systematically proposed to achieve markedly enhanced anisotropic responses [12–14]. Ferroelectric poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)) polarization generates an intense local electric field that spatially defines p- and n-type domains in BP, endowing the in-plane heterostructure with an extraordinary polarization ratio of 288 under 1450 nm illumination [15]. Besides, strain engineering in quasi-one-dimensional layered ZrSe₃, enabled by momentum- and band-dependent modulations of the electronic band structure enhances intrinsic polarization anisotropy as well as tunes operational energies in ultrafast all-optical switching [16]. Importantly, diverse symmetry van der Waals (vdW) interfaces can be constructed between two-dimensional materials with symmetry engineering by controllable stacking or twisting angles, making possible the design of high-performance, multifunctional, and low-power devices.

In this work, we designed a vdW vertical BP/Nb₂SiTe₄ heterostructure to realize a self-powered, broadband, and polarization-sensitive photodetector. Characterizations including optical microscopy (OM), atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), and high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) confirm the morphology, stacking orientation, and the atomically sharp BP/Nb₂SiTe₄ heterostructure interface. The pronounced in-plane anisotropy is further corroborated by polarization-resolved Raman and absorption spectra. Density functional theory (DFT) calculations reveal that deliberate symmetry reduction of the heterostructure markedly enhances the intrinsic anisotropy. Benefiting from the Type-II band alignment and built-in electric field, the BP/Nb₂SiTe₄ device displays a self-driven and broadband photoresponse. Notably, a dichroic ratio (DR) is calculated up to 10.4 under 2200 nm illumination due to the degeneracy of the in-plane optical response induced by symmetry engineering. Finally, the BP/Nb₂SiTe₄ heterostructure enables polarization-resolved imaging and encryption/decryption, presenting a viable strategy for developing low-power and multifunctional photodetectors.

2 Experimental section

2.1 Preparation of BP and Nb₂SiTe₄

Red phosphorus (99.999%) was used as the precursor for the

synthesis of BP crystals. BP crystals could be obtained under high-temperature and high-pressure environment. High-purity Nb (99.95%), Si (99.999%), and Te (99.99%) powders were uniformly mixed in a molar ratio of 2:1.5:4 with 10 mg of iodine. The mixture was then hermetically sealed under an ultra-high vacuum of 10 Pa. The sealed tube was positioned in a tubular furnace with its powder-bearing end (hot end) at 1000 °C and its counterpart cold end maintained at 850 °C, followed by a 10-day heat preservation. Afterward, the sample was allowed to cool naturally to room temperature over 18 h, then carefully removed and promptly stored under vacuum. The as-synthesized BP and Nb₂SiTe₄ crystals were kept in a glove box filled with Ar gas.

2.2 BP/Nb₂SiTe₄ heterostructure

Bulk BP and Nb₂SiTe₄ crystals were adhered to 3M tape, followed by repeated peeling and sticking of the tape for at least 10 cycles to thin the crystal layers, and then the target few-layer flakes were exfoliated. Subsequently, with polydimethylsiloxane (PDMS) serving as the transfer medium, the exfoliated few-layer BP nanosheets were transferred onto a silicon dioxide (SiO₂) substrate. The substrate was then placed on a hot stage and heated at 80 °C for 15 min to achieve stable adhesion of the few-layer BP to SiO₂ substrate. Prior to heterojunction lamination, the zone axes of BP and Nb₂SiTe₄ were precisely aligned. Finally, the few-layer Nb₂SiTe₄ nanosheets on PDMS were rotated to the desired orientation using a transfer stage and transferred onto the surface of BP flake. The assembled structure was further annealed in a vacuum tube furnace at 200 °C for 15 min to enhance interface bonding, thereby successfully fabricating the BP/ Nb₂SiTe₄ heterojunction.

2.3 Preparation of cross-sectional TEM sample

A high-precision focused ion beam (FIB) instrument was utilized to fabricate and thin BP/Nb₂SiTe₄ heterostructure samples. A series of procedures including electron beam positioning, platinum protective layer deposition, multi-angle trench milling, tungsten probe transfer, low-angle thinning, and surface cleaning were implemented to obtain ultra-thin specimens with a thickness of 50–70 nm for subsequent characterizations. Afterwards, the as-prepared sample was mounted on a double-tilt holder and loaded into a JEM-ARM300F2 transmission electron microscope operating at an accelerating voltage of 300 kV. Kikuchi lines were first used to identify the crystallographic zone axis to acquire selected-area electron diffraction patterns. The microscope was then switched to STEM mode. Spherical aberration correction was optimized via Ronchigrams and supporting software to acquire high-angle annular dark-field images and high-resolution atomic images for characterizing the atomic structure of the heterojunction. Finally, the microscope was equipped with an energy-dispersive X-ray spectroscopy (EDS) detector to conduct elemental mapping.

2.4 Characterizations

The morphology of BP/Nb₂SiTe₄ vertical heterostructure was investigated by OM (D4000M, Leica). Thickness and surface potential difference were measured via AFM and Kelvin probe force microscopy (KPFM, Multimode 8, Bruker), respectively. The phase structure and elemental composition of BP/Nb₂SiTe₄ heterostructure were characterized by XRD (Rigaku), XPS (ESCALAB 250Xi, Thermo Fisher Scientific), and STEM (JEM-ARM300F2, JEOL). The angle-resolved polarized Raman of BP/Nb₂SiTe₄ heterostructure was investigated by a confocal Raman spectroscopy equipped with a focused 532 nm laser (JY Horiba,

HR Evolution). The angle-resolved absorption spectra were recorded by confocal ultraviolet–near infrared (UV–NIR) spectrometer (Matatest, MStarter ABS).

2.5 Device fabrication and measurements

BP/Nb₂SiTe₄ heterostructure electrodes were fabricated via laser direct writing, followed by thermal evaporation of metal and lift-off processing. Photoelectric response data under illumination of various wavelengths (360, 457, 532, 671, 914, 1064, 1550, and 2200 nm) were acquired using a Keithley 2634B source-measure. The rise and fall time were defined as the intervals required for the increase of the photocurrent from 10% to 90% and the decrease from 90% to 10%, respectively. The laser polarization was fixed with its electric field initially aligned parallel to the device's longest edge, corresponding to the *b*-axis of Nb₂SiTe₄ and defined as 0°. All measurements were conducted at room temperature. The specific detectivity D^* and external quantum efficiency (EQE) were calculated using the following equations: $D^* = RS^{1/2}/(2qI_{\text{dark}})^{1/2}$ and $\text{EQE} = hcR/q\lambda$, where R is the responsivity of the device, q is the elementary charge, h is the Planck constant, c is the velocity of light, and λ is the laser wavelength.

2.6 DFT calculation

First-principles calculations were performed using the projector-augmented wave method, implemented within the Vienna *ab-initio* simulation package (VASP). The exchange–correlation interactions were modeled with the Perdew–Burke–Ernzerhof (PBE) functional, 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 BP/Nb₂SiTe₄ structures, a vacuum spacing of 20 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 probed using a Monkhorst–Pack *k*-point mesh with a spacing of $2\pi \times 0.02 \text{ \AA}^{-1}$. The structural relaxation process was concluded when both the total energy changes and the ionic Hellmann–Feynman forces fell below stringent thresholds of $1.0 \times 10^{-7} \text{ eV}$ and $0.01 \text{ eV/\AA}$, respectively. The electronic band structure of the crystal structures was investigated further by using the HSE06 hybrid functional method.

3 Results and discussion

3.1 Characterization of BP/Nb₂SiTe₄ heterostructure

Figure 1(a) illustrates the atomic structure models of the BP/Nb₂SiTe₄ vdW heterostructure. The crystal structure of Nb₂SiTe₄ is monoclinic with space group $P2_1/c$ whereas layered BP adopts an orthorhombic structure with the space group $Cmca$ (Fig. S1 in the Electronic Supplementary Material (ESM)). The detailed growth process of bulk Nb₂SiTe₄ crystals is provided in Section 2 and Fig. S2 in the ESM. Subsequently, the few-layer Nb₂SiTe₄ nanosheet prepared by mechanical exfoliation from the bulk crystal is vertically stacked onto the BP nanosheet to construct a BP/Nb₂SiTe₄ heterostructure. As shown in Figs. 1(b) and 1(c), the typical AFM image and the corresponding height profile of the BP/Nb₂SiTe₄ heterostructure confirm the vertical stacking of Nb₂SiTe₄ (21 nm) onto the BP nanosheet (44 nm). The interlayer coupling of the BP/Nb₂SiTe₄ heterostructure was investigated using Raman

spectroscopy. Raman spectra collected from pure BP, the overlapping heterostructure, and the Nb₂SiTe₄ region are shown in Fig. 1(d). In pure BP, Raman peaks at 362 and 467 cm^{−1} correspond to A_g vibrational modes and the peak at 440 cm^{−1} corresponds to B_{2g} vibrational mode, while in Nb₂SiTe₄, peaks at 94, 121, 132, 161, and 204 cm^{−1} are attributed to A_g modes. For the heterostructure, no significant peak shift is observed, but a substantial reduction in peak intensity occurs, attributed to strong interfacial coupling between BP and Nb₂SiTe₄, consistent with previous reports [17, 18]. Furthermore, Raman mapping of vibrational modes in the BP/Nb₂SiTe₄ heterostructure reveals uniform intensity distribution, confirming a high-quality vertical stacking heterostructure (Fig. S3 in the ESM).

The XPS was employed to examine elemental composition and valence states. As displayed in Fig. 1(e), the peaks located at approximately 206.7, 204.0, 101.1, 100.6, 583.7, 573.2, 133.9, and 129.8 eV, corresponding to Nb 2d_{3/2}, Nb 2d_{5/2}, Si 2p_{1/2}, Si 2p_{3/2}, Te 2d_{3/2}, Te 2d_{5/2}, P 2p_{1/2}, and P 2p_{3/2} orbitals, respectively, in agreement with the reported results [19, 20]. The stacking orientation and interface quality are assessed by XRD and aberration-corrected STEM (AC-STEM). As shown in Fig. 1(f), the XRD pattern exhibits diffraction peaks at 12.6°, 25.2°, 38.3°, and 51.8°, which are attributed to (002), (004), (006), and (008) planes of Nb₂SiTe₄, and peaks at 16.9°, and 30.5° correspond to the (020) and (040) planes of BP, respectively, indicating parallelism between the (002) plane of Nb₂SiTe₄ and the (020) plane of BP.

The cross-sectional HAADF-STEM images (Fig. 1(g)) reveal the atomically sharp interface with no interfacial disorder or chemical interdiffusion. High-resolution HAADF-STEM images reveal atomic structures of BP along the [020] zone axis and Nb₂SiTe₄ along the [002] zone axis, matching well with their crystal structure models. The interlayer spacings of BP and Nb₂SiTe₄ are 0.53 and 0.74 nm, respectively (Fig. S4 in the ESM). Additionally, EDS mapping further confirms a spatially uniform distribution of Nb, Te, Si, and P elements, indicating the high-quality vertical stacking of the BP/Nb₂SiTe₄ heterostructure.

3.2 Anisotropic optical properties

The crystallographic orientation and in-plane anisotropy of phonon vibrations can be further characterized by angle-resolved polarized Raman spectroscopy (ARPRS). Schematic illustrations of the ARPRS setup for parallel and cross polarization configurations are presented in Figs. 2(a) and 2(b). Contour maps of polarization angle-dependent Raman intensity for the BP/Nb₂SiTe₄ heterostructure exhibit pronounced periodic oscillations under parallel and cross configurations (Figs. 2(c) and 2(d)). Notably, the obtained polar plots of polarization-resolved intensity in parallel and cross configurations are further shown in Figs. 2(e) and 2(f). The main typical Raman peaks at 161 and 467 cm^{−1} exhibit a two-lobed pattern for the parallel polarization configuration and a four-lobed pattern under the cross polarization configuration, evidencing the intrinsic anisotropic phonon vibrations governed by the crystal symmetry of the BP/Nb₂SiTe₄ heterostructure. Further details about the BP/Nb₂SiTe₄ heterostructure in parallel and cross configurations are presented in Fig. S5 in the ESM. Polarization-resolved absorption spectroscopy (PRAS) is performed to assess the optical anisotropy of the heterostructure. Figure 2(g) presents contour maps of polarization angle-dependent absorption intensity, revealing distinct anisotropic optical responses in the wavelength range from 350 to 1600 nm. Two characteristic absorption peaks,

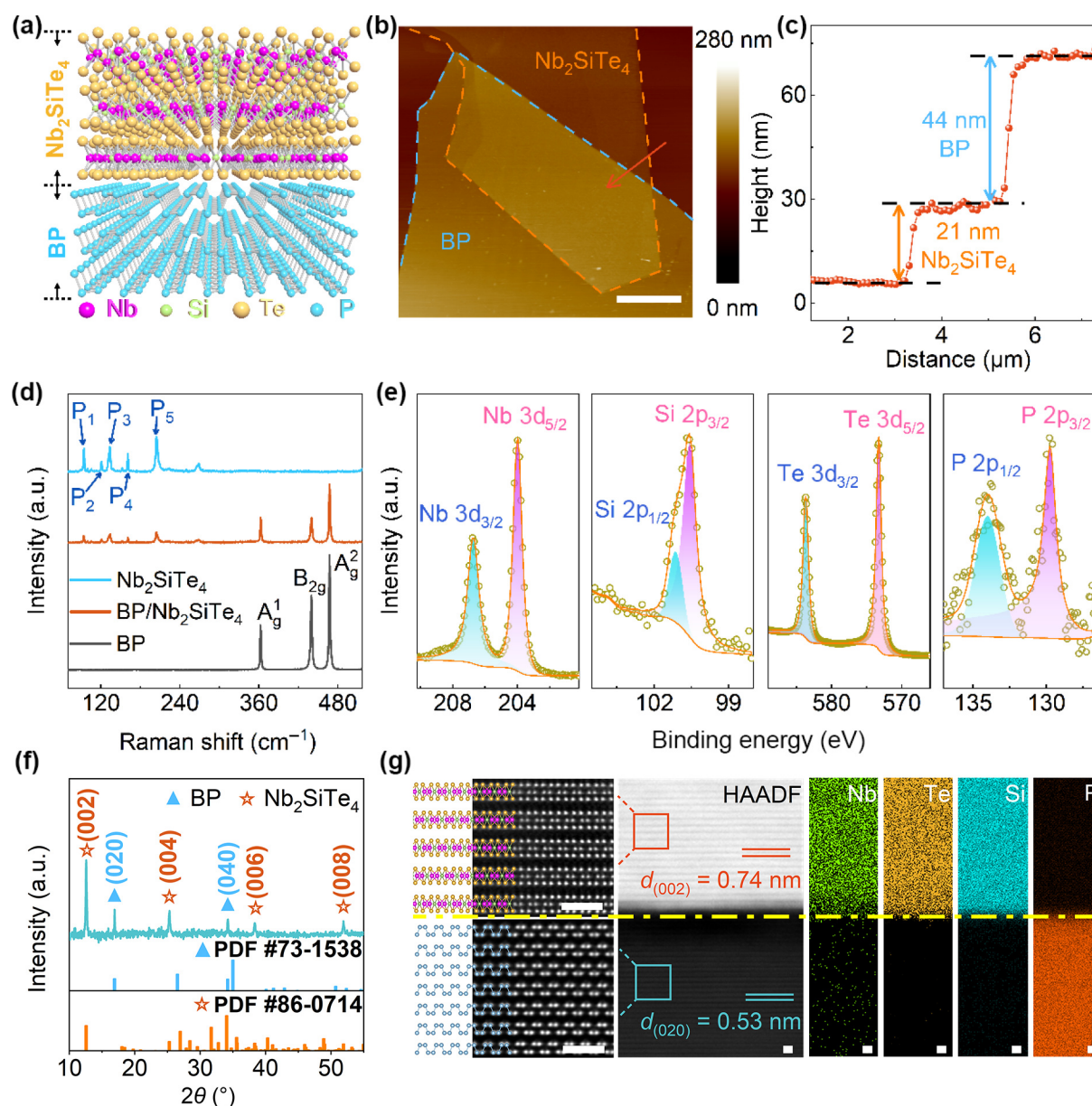


Figure 1 Crystal structure and characterization of BP/Nb₂SiTe₄ heterostructure. (a) The atomic structure model of BP/Nb₂SiTe₄ heterojunction. The purple, green, yellow, and cyan spheres represent Nb, Si, Te, and P atoms, respectively. (b) AFM image and (c) the corresponding height profile curve of BP/Nb₂SiTe₄ heterostructure (scale bar: 5 μ m). (d) Raman spectra of the Nb₂SiTe₄ (top), BP/Nb₂SiTe₄ heterostructure (middle), and BP (bottom). (e) The high-resolution XPS spectra of Nb 3d, Si 2p, Te 3d, and P 2p orbitals. (f) XRD pattern of BP/Nb₂SiTe₄ heterostructure. (g) High-resolution cross-sectional HAADF-STEM image and EDS mapping of BP/Nb₂SiTe₄ heterostructure (scale bar: 1 nm).

attributed to the B and C excitons of Nb₂SiTe₄ (Fig. 2(h)), exhibit a π -periodic modulation as a function of polarization angle, corroborating the intrinsic anisotropy in light absorption. Moreover, we further probed the influence of the heterostructure interface on the optical anisotropy through DFT calculations. The charge distribution at the valence band maximum (VBM), viewed from the top and side perspectives, predominantly accumulates near the Te and P atomic regions (Figs. 2(i) and 2(l)). The two-dimensional charge densities along the *c*- and *b*-axis are further illustrated in Figs. 2(j) and 2(m), indicating a spatially non-uniform charge distribution. The relative charge density analysis further elucidates the microscopic origin: Line profiles along the *c*- and *b*-axes display a stark contrast, and steep charge gradients and complete electron-hole separation along the *c*-axis generate a

strong interfacial dipole moment, whereas the *b*-axis shows only modulated transfer with residual carrier overlap. This pronounced directional anisotropy arises from the deliberate symmetry reduction achieved by stacking Nb₂SiTe₄ on BP, which enables selective hybridization of interfacial states with BP's armchair and zigzag crystallographic directions. The charge density curves extracted from the dark gray dashed lines (Fig. 2(j)) and dark gray/red dashed lines (Fig. 2(m)) further confirm the corrugated oscillatory charge distribution (Figs. 2(k) and 2(n)), which arises from the symmetry reduction caused by the stacking twist angle [21].

3.3 Self-driven and ultrabroadband photoresponse

Benefiting from the broadband absorption spectrum of the

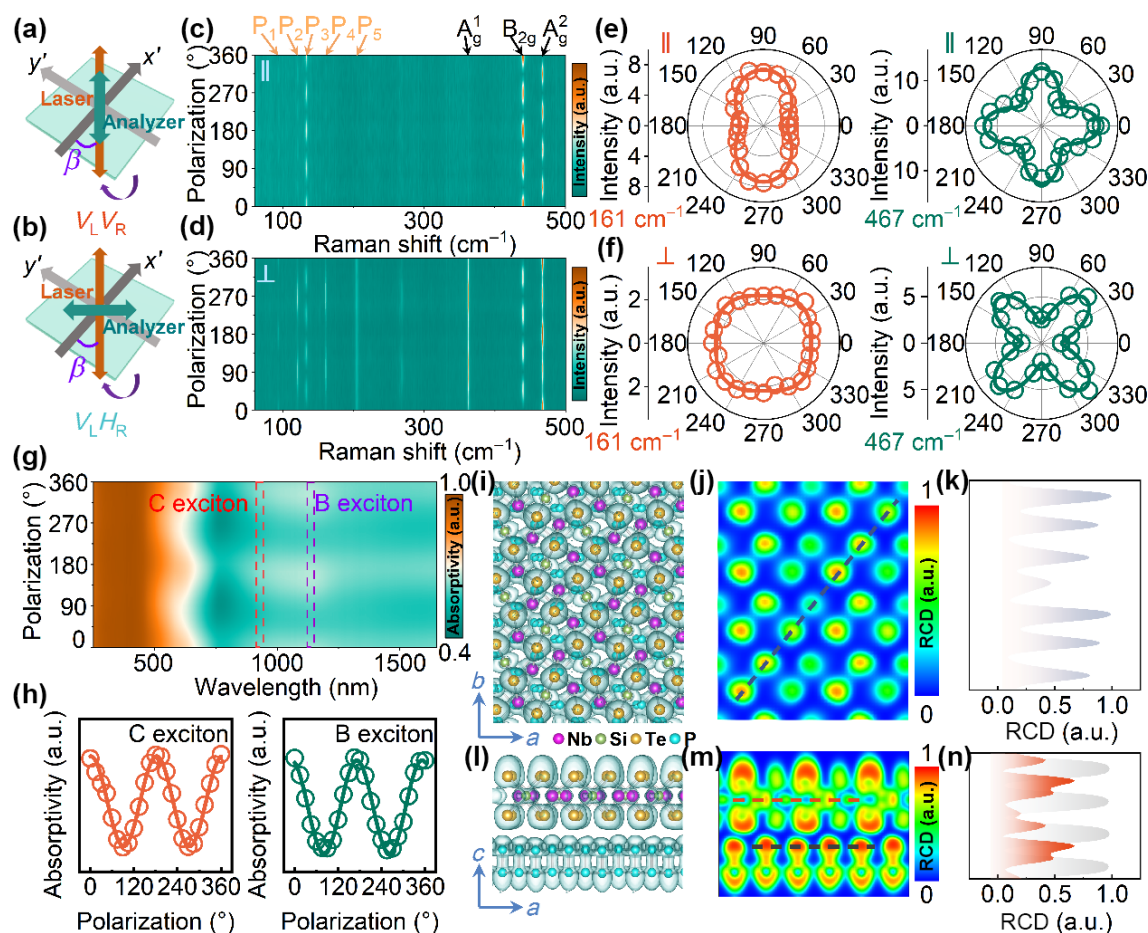


Figure 2 Anisotropic optical properties based on BP/Nb₂SiTe₄ heterostructure. ((a) and (b)) Schematic diagrams of polarization angle-dependent Raman intensity measurements in parallel and cross configurations. ((c) and (d)) Contour maps of polarization angle-dependent Raman intensity in parallel and cross configurations. The orange arrows indicate the P₁–P₂ vibrational modes of Nb₂SiTe₄. The black arrows indicate A_g¹, B_{2g}, and A_g² vibrational modes of BP. ((e) and (f)) Polar plots of polarization-resolved P₁ (Nb₂SiTe₄) and A_g² (BP) intensities in parallel and cross configuration. (g) Contour maps of polarization angle-dependent absorption intensity under different wavelengths. (h) The two typical absorption peak intensities (B and C excitons) as a function of polarization angle. (i) and (j) The calculated relative charge density (RCD). ((k) and (l)) 2D plot of relative charge density. (m) and (n) The corresponding line profiles taken along c- and b-axes. The densities of electrons and holes are shown in blue and red, respectively.

BP/Nb₂SiTe₄ heterostructure, the device is expected to deliver a sensitive and photoresponse over a wide spectral range. The typical OM and AFM images of the device are shown in Fig. S6 in the ESM. The thicknesses of the BP and Nb₂SiTe₄ are measured to be 48 and 19 nm, respectively, based on the height profile curves (Fig. S6(c) in the ESM). The representative I_{ds} – V_{ds} characteristic curves in the dark and under 457 nm light irradiation at different light intensities are displayed in Fig. 3(a). The positive open-circuit voltage (V_{oc}) and negative short-circuit current (I_{sc}) can be generated, unambiguously revealing a pronounced photovoltaic effect driven by built-in electric fields induced by band bending that efficiently separate photogenerated electron–hole pairs. The I_{sc} rises sharply from 6.2 to 24.9 nA as the power density increases from 0.57 to 3.40 mW/cm² due to the photo-induced abundant charge carriers.

The generated electrical power (P_d), calculated by equation $P_d = I_{ds} \cdot V_{ds}$, reaches a maximum value $P_{d,max} = 1.38$ nW under incident power of 3.40 mW/cm² (Fig. 3(b)). To systematically evaluate the self-powered photoresponse of the BP/Nb₂SiTe₄ device, time-resolved measurement was performed under 457 nm light illumination with different intensities at zero bias (Fig. 3(c)). The photocurrent gradually increases from 5.96 to 312 nA with

increasing incident light power intensity from 0.06 to 3.40 mW/cm². A series of I – T curves for the BP/Nb₂SiTe₄ photodetection show stable photoresponse at different wavelengths (Fig. 3(d)), suggesting a self-powered photoresponse and ultra-broad spectral capability. Notably, a sub-linear power-intensity-dependent photocurrent was quantitatively analyzed by the following equation: $I_{ph} \sim \alpha P^\delta$ (Fig. 3(e)). The extracted power factor (δ) was evaluated at 0.85, 0.91, 0.95, and 0.77 under wavelengths of 360, 457, 1064, and 2200 nm, respectively, implying a complex process of carrier recombination and defect trapping [22]. To elucidate the superiority of the BP/Nb₂SiTe₄ photodetector, we conducted a comparative analysis of its figure-of-merits, including the EQE, R , D^* , and response time. R , D^* , and EQE as a function of power intensity are shown in Fig. S7 in the ESM. The maximum R reached up to 4.0×10^{-4} , 3.0×10^{-3} , 3.0×10^{-6} , and 9.1×10^{-6} A/W at wavelength of 360, 457, 1064, and 2200 nm, respectively. A more detailed discussion of the optoelectronic performance parameters is presented in Fig. S7 in the ESM. The noise spectrum clearly indicates the decisive role of $1/f$ flicker noise (Fig. S8 in the ESM). As shown in Fig. 3(f) and Fig. S9 in the ESM, the rise and decay time of BP/Nb₂SiTe₄ devices were evaluated to be as high as 24 and 13 μ s, respectively, which are five orders of magnitude faster than

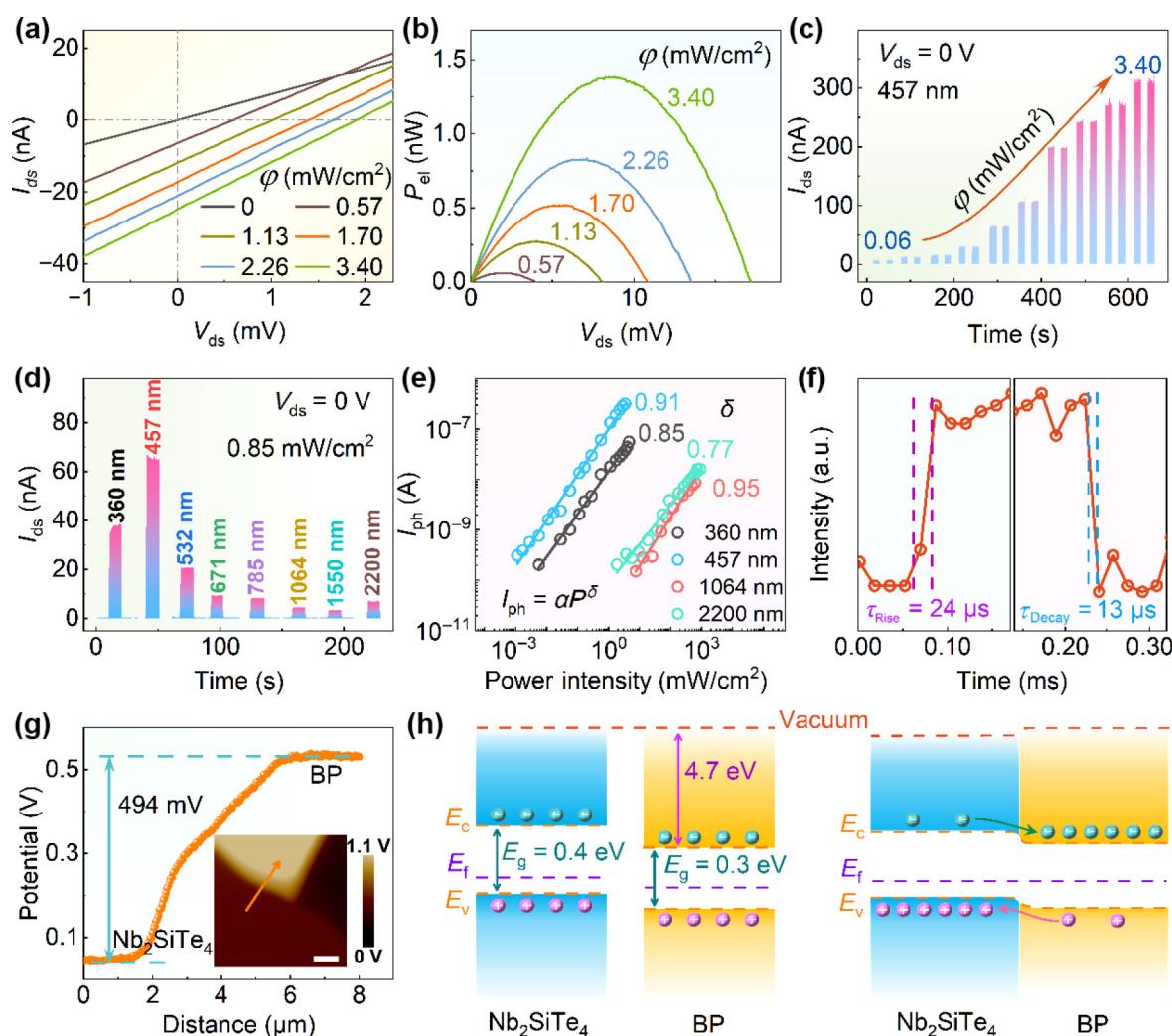


Figure 3 Self-driven and broadband optoelectronic performance and mechanism of the BP/Nb₂SiTe₄ photodetector. (a) Spatial mapping of I_{ds} as a function of incident light power density. (b) Relationship between electric power and bias voltage under varying optical power intensities of the BP/Nb₂SiTe₄ device. (c) Time-dependent photoresponse behavior of the heterojunction device under varied power intensities of 457 nm light at $V_{ds} = 0$ V. (d) Time-dependent photoresponse behavior of the device under varied wavelengths (360–2200 nm) of light illumination with a light power density of 0.85 mW/cm². (e) I_{ph} as a function of power density under varied wavelengths. (f) Response speed of the heterojunction device at 457 nm laser illumination under 0 V bias voltage. (g) The KPFM surface potential mapping and the corresponding line profile, suggesting the Fermi level difference between BP and Nb₂SiTe₄. (h) Band diagrams for the BP/Nb₂SiTe₄ heterostructure before (left) and after contact (right). The arrows indicate that electrons transfer from Nb₂SiTe₄ to BP, while holes transfer from BP to Nb₂SiTe₄.

the response time of pure Nb₂SiTe₄ ($\tau_{rise} = 1.03$ s and $\tau_{decay} = 2.14$ s). As shown in Fig. S10 in the ESM, the BP/Nb₂SiTe₄ photodetector exhibits negligible photocurrent degradation after approximately 4000 switching cycles under 2200 nm illumination, indicating superior stability and reproducibility of the heterostructure. The improvement in response time is attributed to the high carrier mobility in the heterostructure and the strong built-in electric field that rapidly separates photogenerated electron–hole pairs [23]. To further understand the outstanding self-driven response mechanism, the KPFM and band alignment were investigated in the BP/Nb₂SiTe₄ heterostructure. As depicted in Fig. 3(g), the KPFM image of the surface potential difference between BP and Nb₂SiTe₄ was evaluated to be about 494 mV with a depletion width of 2 μ m, evidencing a pronounced built-in electric field at the heterointerface. Figure 3(h) schematically illustrates the band arrangements of BP and Nb₂SiTe₄ before and after contact. Notably, when BP is vertically stacked with Nb₂SiTe₄ to form a vdW heterojunction, the work function difference induces spontaneous

carrier redistribution at the interface. Owing to the higher Fermi level of Nb₂SiTe₄ relative to BP, electrons diffuse from Nb₂SiTe₄ to BP while holes migrate in the opposite direction. As the Fermi levels equilibrate, a built-in electric field is established at the heterointerface directed from Nb₂SiTe₄ toward BP, resulting in a Type-II (staggered) band alignment where the conduction band minimum (CBM) of BP lies below the valence band maximum (VBM) of Nb₂SiTe₄. This band configuration creates a significant potential gradient that effectively suppresses recombination of photogenerated electron–hole pairs and facilitates their spatial separation: Electrons accumulate in the BP layer, whereas holes accumulate in the Nb₂SiTe₄ layer. When the incident photon energy exceeds the bandgap of either constituent material, photogenerated electron–hole pairs undergo directional drift driven by the aforementioned built-in electric field, with electrons migrating toward the BP side and holes toward the Nb₂SiTe₄ side, thereby achieving efficient spatial separation of photogenerated carriers. This charge separation process minimizes recombination losses

within the depletion region. Even in the absence of external bias, the separated carriers can be effectively collected by the respective electrodes, thereby continuously driving the separation and transport of photogenerated electron-hole pairs and enabling ultrafast self-powered photoresponse.

3.4 Broadband polarization-sensitivity photodetectors

The atomically sharp $\text{Nb}_2\text{SiTe}_5/\text{BP}$ heterostructure interface not only induces a strong built-in electric field for efficient carrier separation, but also enhances interfacial polarization selectivity, enabling a broadband and self-powered polarization-resolved photodetector. Figure 4(a) schematically depicts the polarization-resolved photocurrent configuration under different wavelength ranges from 360 to 2200 nm. A half-wave plate is rotated in steps of 15° to change the linear-polarization direction at wavelengths of 360, 532, 1064, and 2200 nm. As shown in Fig. 4(b), the time-resolved photocurrent recorded at zero bias displays periodic

oscillations as the polarization angle sweeps from 0° to 360° , evidencing self-driven and highly anisotropic photocurrent generation. Notably, as the polarization angle changes under 360, 532, 1064, and 2200 nm lasers, the maximum photocurrent values reached 37, 20, 4.7, and 7.42 nA at 90° (270°), while the minimum photocurrent values are 16.9, 8.09, 1.01, and 0.71 nA at 0° (180°), respectively. To further systematically and quantitatively investigate the polarization-angle-dependent photocurrent intensity with different laser irradiation, polar plots of the polarized photocurrent are presented in Figs. 4(c)–4(f) and Fig. S11 in the ESM, which are accurately fitted by the following equation: $I_\theta = I_{\text{ph,min}} \sin^2(\theta + \varphi) + I_{\text{ph,max}} \cos^2(\theta + \varphi)$, where $I_{\text{ph,min}}$ and $I_{\text{ph,max}}$ are the minimum and maximum photocurrent values, respectively. The DRs, defined as $I_{\text{ph,max}}/I_{\text{ph,min}}$, are calculated to be 2.2, 1.1, 2.5, 3.1, 4.3, 4.6, 3.6, and 10.4 at a power intensity of 1.13 mW/cm^2 at wavelengths of 360, 457, 532, 671, 914, 1064, 1550, and 2200 nm, respectively. Furthermore, the DR in $\text{BP}/\text{Nb}_2\text{SiTe}_5$ devices can be readily tuned from 2.3 to

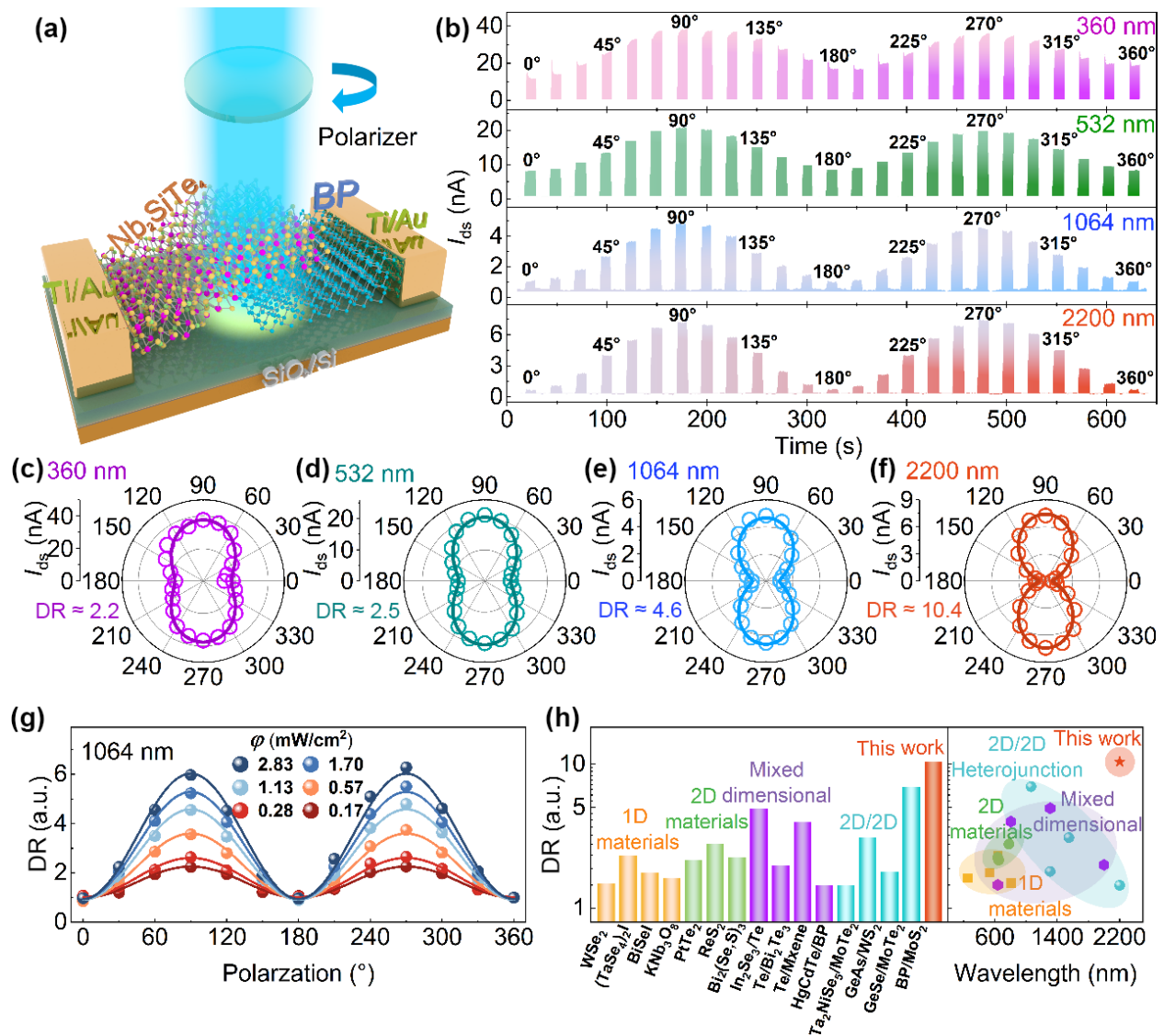


Figure 4 Polarization sensitivity photodetector of $\text{BP}/\text{Nb}_2\text{SiTe}_5$ heterostructure. (a) Schematic diagram for polarization-resolved photocurrent measurement under different wavelength ranges from 360 to 2200 nm. (b) The time-resolved polarization-angle-dependent photoresponse at various wavelengths (360, 532, 1064, and 2200 nm) under power intensity of 1.13 mW/cm^2 . (c)–(f) Polar plots of the photocurrent versus polarization angle under various wavelength. (g) Dichroic ratios as a function of power density under 532 nm illumination ($V_{\text{ds}} = 0 \text{ V}$). (h) Comparison of reported DR values of polarization-sensitive photodetectors based on 1D materials, 2D materials, mixed dimensional heterojunctions, and 2D/2D heterojunctions.

6.3 by increasing incident power density range from 0.17 to 2.83 mW/cm² of the 1064 nm irradiation (Fig. 4(g)). Notably, the DR of the BP/Nb₂SiTe₄ photodetector in the broadband range is superior to most photodetectors based on one-dimensional (1D) materials [24–27], 2D materials [28–30], mixed dimensional heterostructures [31–34], and 2D/2D heterostructures [17, 35–37] compared to previously reported results, as summarized in Fig. 4(h).

3.5 Polarization imaging

Traditional division-of-focal-plane polarimeters require a minimum of four neighboring pixels—each capped with an oriented micropolarizer, to reconstruct the first three Stokes parameters and hence one degree of linear polarization (DoLP) value. Consequently, producing an $m \times n$ DoLP image necessitates a $2m \times 2n$ pixel array, halving the effective spatial resolution. In contrast, the intrinsic anisotropic photoresponse of BP/Nb₂SiTe₄ devices, combined with their high dynamic range and ultrafast response time in the short-wave infrared (SWIR) region, enables direct linear polarization state detection at the single-pixel level. This capability allows for full-resolution DoLP imaging without spatial undersampling, while preserving the integrity of polarization information to discriminate subtle polarization signatures among objects with unprecedented fidelity. The schematic of the contrast-

enhanced polarimetric imaging system is depicted in Fig. 5(a). In the test setup, a digital device sequentially projects a structured light mask onto the object (YSU-CHIPS target, 88 pixel $\times$ 57 pixel) while the incident polarization is rotated to 0°, 45°, 90°, and 135° (Fig. 5(a)). Photocurrent maps $I_0(x, y)$ are acquired by scanning the mask in the x - y plane and represented by a color bar. Then, the raw data are processed off-chip to reconstruct the DoLP (Figs. 5(b) and 5(c)). Figure 5(d) illustrates the schematic diagram of the polarization imaging procedure for DoLP calculation, given by 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 $I_\theta(x, y)$ is the photocurrent intensity under different polarization angles.

To highlight the advantages of polarization imaging based on the

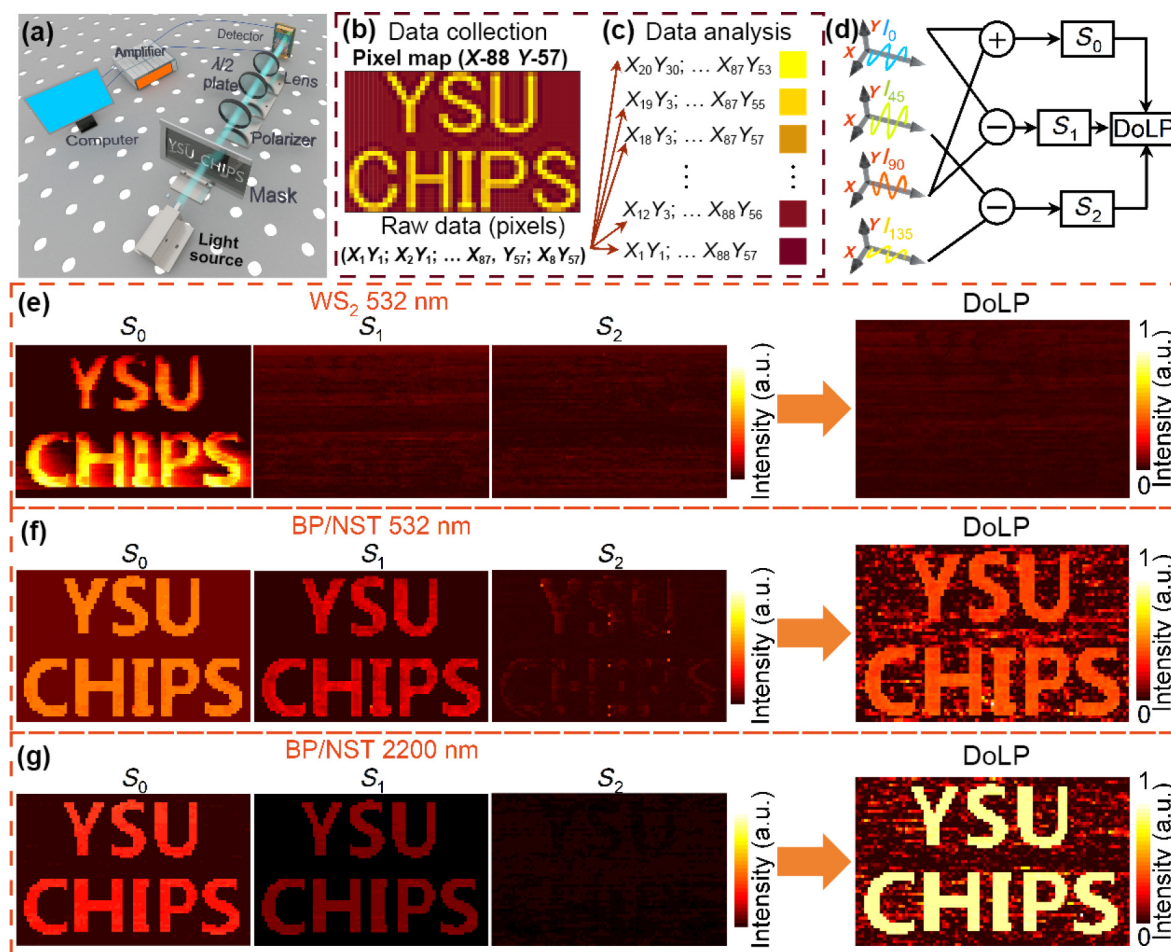


Figure 5 Single-pixel polarization imaging of BP/Nb₂SiTe₄ photodetector under 2200 nm illumination. (a) Schematic representation illustrating the working principle of single-pixel polarization imaging. (b) and (c) Raw data collection (an 88 $\times$ 57 matrix array) of real-time photocurrent and corresponding position coordinate color information. (d) Theoretical DoLP results obtained by different polarization state processing. ((e)–(g)) The corresponding calculating DoLP imaging results of polarization image with four polarization states (0°, 45°, 90°, and 135°) information under 532 (pure WS₂), 532 (BP/Nb₂SiTe₄), and 2200 nm (BP/Nb₂SiTe₄) illumination, respectively.

anisotropic BP/Nb₂SiTe₄ detector, we first employed a non-polarized 3R-stacked WS₂ photodetector to image the YSU-CHIPS target under varying polarization angles. As shown in Fig. 5(e) and Fig. S11(a) in the ESM, the non-polarized WS₂ device exhibits a DoLP mapping in which the target is barely discernible. In sharp contrast, after the DoLP calculation the BP/Nb₂SiTe₄ detector delivers a markedly enhanced image contrast for the same target (Fig. 5(f) and Fig. S12(b) in the ESM). Under 2200 nm illumination, the larger absorption anisotropy ratio affords even sharper polarization imaging (Fig. 5(g) and Fig. S12(c) in the ESM), underscoring the great potential of BP/Nb₂SiTe₄ detectors for advanced polarimetric imaging applications.

3.6 Near-infrared polarization-encoded encryption and decryption

Taking advantage of the distinct NIR polarization responses at 1550 and 2200 nm, we introduce a proof-of-concept sensitive NIR-polarization encryption/decryption scheme in which both modalities are monolithically integrated within a single BP/Nb₂SiTe₄ device. As illustrated in Fig. 4(f) and Fig. S11 in the ESM, polarization photocurrents extracted at wavelengths of 1550 and 2200 nm exhibit 180°-periodic mirror-symmetric oscillations with respect to the polarization angle. At polarization angles of 0°, 180°, and 360°, the two photocurrent intensities overlap, whereas the two photocurrent signal intensities are clearly separated under

polarization angles of 90° and 270°. This angle-selective contrast provides the physical basis for image encryption and decryption in the near-infrared region. Figure 6(a) presents the polarization-encoded virtual image reconstructed simultaneously at wavelengths of 2200 and 1550 nm. The “pumpkin” image is interrogated by these two spectral channels, whose raw photocurrent maps are intentionally contaminated with stochastic noise extracted from the actual BP/Nb₂SiTe₄ device (Fig. 6(b)). Importantly, we define the wavelength of 2200 nm illumination as the information-bearing channel (content), whereas the wavelength of 1550 nm channel serves as a device-intrinsic noise source (background). When the incident polarization angle is rotated to 0°, the photocurrent distributions of the two channels becomes statistically indistinguishable. Consequently, the image content is cryptographically obscured. Upon rotating the polarization angle to 90°, the differential contrast between channels become monotonic, culminating in complete isolation of the content-bearing 2200 nm signal and unambiguous decryption of the latent image. Furthermore, we conducted a comparative analysis of the recognition performance between encrypted and decrypted images using a convolutional neural network (CNN) method (Fig. 6(c)). As shown in Fig. 6(d), compared with the pristine images, letters drawn from the EMNIST dataset become visually unrecognizable upon dual-wavelength encryption, while letters processed with polarization remain clearly discernible. Figure 6(e) further shows a

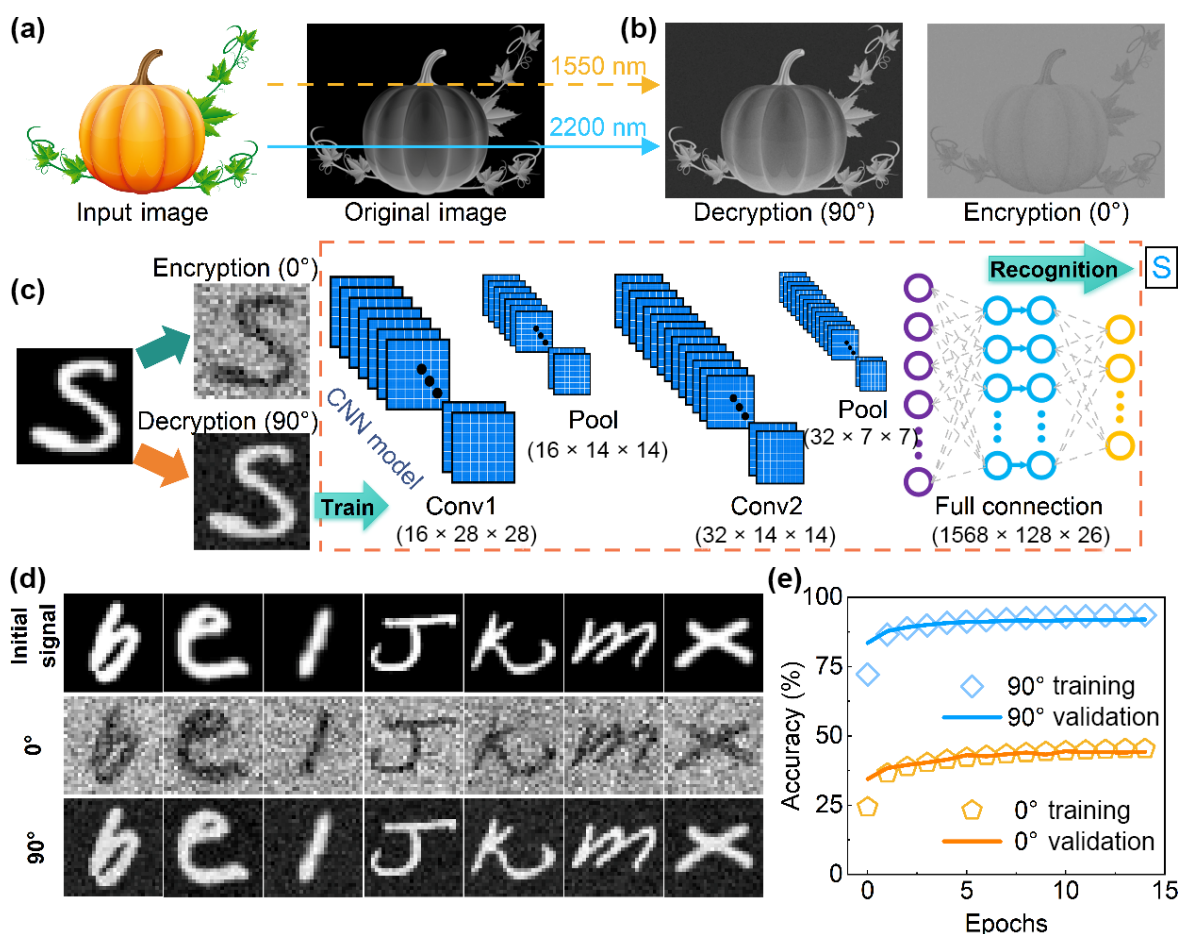


Figure 6 Polarization-encoded encryption and decryption. (a) Cryptographic imaging application of BP/Nb₂SiTe₄ photodetector. Polarization virtual imaging of the “pumpkin” image under 2200 and 1550 nm illumination. (b) Decryption (0°) and encryption (90°) based on “pumpkin” image. (c) Schematic diagram of image processing by CNN. (d) CNNs for recognizing encrypted and decrypted images in the English letters EMNIST dataset. (e) Accuracy comparison at 0° and 90° encrypted images as a function of epochs.

comparison of the accuracy of encrypted and unencrypted images as the number of epochs increases. Encrypted images retain only 45.33% (training) and 44.28% (validation) classification accuracy after 15 epochs, whereas decrypted images recover 93.68% and 92.01% accuracy, showing polarization encryption and decryption capabilities. These results demonstrate that the dual-wavelength polarization strategy creates significant response contrast, substantially enhancing encryption/decryption capabilities and providing a multifunctional and low-power prototype for secure near-infrared image transmission and classification.

4 Conclusions

In summary, the novel strategy of stacking BP onto Nb₂SiTe₄ to form a vdW heterostructure provides a platform versatile that simultaneously enhances photoresponse performance and encodes near-infrared polarization-selective encryption/decryption functionalities. Atomically sharp and contamination-free heterointerfaces generate a built-in electric field at the BP/Nb₂SiTe₄ heterostructure interface due to the work function difference and Fermi level equilibration between the two components, facilitating ultrafast electron-hole pair separation. Consequently, BP/Nb₂SiTe₄ photodetector exhibits self-powered and a broadband spectral response range from 360 to 2200 nm. Notably, intrinsic anisotropy of the BP/Nb₂SiTe₄ heterostructure shows polarization angle-resolved photocurrent whose maximum DR reaches 10.4 under 2200 nm light illumination. Moreover, a proof-of-concept based on the BP/Nb₂SiTe₄ photodetector confirms polarization-resolved imaging and polarization-encoded encryption/decryption, providing fresh perspectives for the realization of low-power and multifunctional polarization-sensitive photodetectors.

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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

C. P. M., S. G. W., and K. Y. initiated the project. J. X. Y., B. C. Y., and L. X. L. grew the samples and fabricated the devices. J. X. Y., Q. L., Y. H. S., and K. Y. conducted Raman and PL spectroscopy measurements. Q. L. and T. L. Z. performed the XRD and AFM measurements. X. Q. G. and Z. Y. J. performed the XPS measurements. A. M. N. and J. X. Y. carried out the TEM characterizations. J. X. Y. and K. Y. conducted photodetection testing. Q. G. conducted DFT calculation. C. P. M. and K. Y. wrote the manuscript. All the authors have approved the final manuscript.

Use of AI statement

None.

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