

Application of chiral WS₂ rolls in circular/linear polarization-sensitive photodetectors

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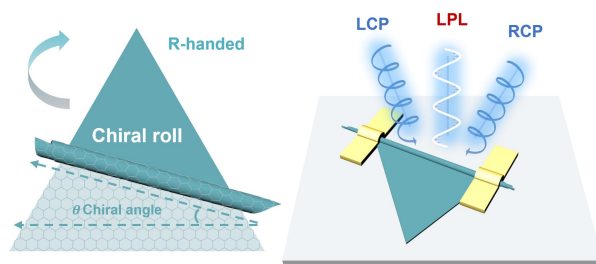
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This work fabricates chiral WS₂ rolls that respond uniquely to linearly and circularly polarized light. Photodetectors built on these rolls demonstrate efficient detection of both polarization types, enabling circular/linear polarization-sensitive photodetectors and showcasing excellent imaging capabilities. This highlights their significant potential for superior optoelectronic applications.

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ABSTRACT: We have fabricated chiral WS₂ rolls that utilize a chiral superstructure to address the lack of chirality in traditional two-dimensional materials. This structure exhibits responsiveness to both linearly polarized light and circularly polarized light. This unique combination enables the development of circular/linear polarization-sensitive photodetectors. Our data demonstrate that chiral WS₂ rolls exhibit robust linear/circular polarization responsiveness. Compared to traditional circular/linear polarization-sensitive photodetectors, this approach simplifies device fabrication and miniaturization, making it a promising optoelectronic material. The ability to effectively detect both linearly polarized light (LPL) and circularly polarized light (CPL) enhances the versatility of these devices, paving the way for innovative applications in optical sensing and communication. Overall, chiral WS₂ rolls represent a significant advancement in the field of optoelectronics, offering potential improvements in performance and functionality.

KEYWORDS: transition metal dichalcogenides (TMDs), chiral WS₂ rolls, linearly polarized light, circularly polarized light

1 Introduction

Two-dimensional (2D) transition metal dichalcogenides (TMDs) have emerged as a pivotal class of materials in the development of optoelectronic devices. Owing to their unique properties, such as a direct bandgap, high absorption coefficient, and remarkable mechanical flexibility, TMDs are being extensively explored for applications in photodetectors, light-emitting devices, and other optoelectronic phenomena [1-8]. The ability of TMDs to interface seamlessly with various materials enhances their potential for revolutionizing electronic and optoelectronic systems.

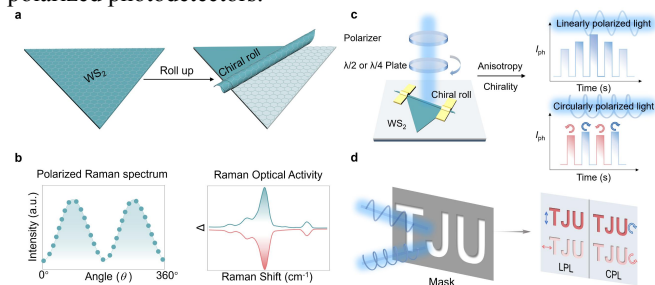
Among the various applications of TMDs, polarization-sensitive detectors play a crucial role in capturing the nuanced behavior of light. LPL detection is particularly important for improving signal clarity and enhancing imaging resolution across diverse fields, including telecommunications, remote sensing, and biomedical imaging [9-17]. The ability to distinguish between different polarization states can significantly enhance photodetection performance, allowing the extraction of complex information from optical signals. However, despite their

promising attributes, traditional non-chiral chemical molecule-modified 2D materials are inherently achiral and thus incapable of responding to CPL.

CPL detection represents another frontier in optical sensing technology, with implications for advanced communication systems and optical processing. Devices that can effectively detect CPL are crucial for applications in chiral molecule detection, which is invaluable in fields such as drug discovery and enantiomeric analysis [18-22]. Unfortunately, the lack of chirality in conventional TMD materials restricts their functionality, as they cannot differentiate between left-handed and right-handed circularly polarized light, ultimately limiting their application.

In response to these challenges, we propose the fabrication of chiral roll-shaped TMD materials. With the chiral TMD roll materials providing robust responses to various polarization states, we aim to pave the way for more versatile and efficient optoelectronic devices, including circular/linear polarization-sensitive photodetectors, which can precisely detect light polarization (both LPL and CPL) and possess excellent polarization imaging capability (Scheme 1). Our work explores

the synthesis and characterization of these chiral superstructures of TMD materials, focusing on their optical properties and the mechanisms facilitating polarization-sensitive photodetectors. This approach leverages the unique properties of 2D materials while overcoming the limitations posed by achiral counterparts, enabling advanced applications in both linearly and circularly polarized photodetectors.



Scheme 1. (a) Chiral WS₂ roll. (b) Polarized Raman spectrum and Raman Optical Activity of chiral WS₂ roll. (c) LPL and CPL detection of the chiral WS₂ roll photodetector. (d) Linearly and circularly polarized imaging.

2 Results and discussion

In this article, we selected WS₂ as the foundational TMD material and WS₂ flakes were synthesized via chemical vapor deposition (CVD). The schematic diagram of the synthesis

process, Raman spectra and atomic force microscopy (AFM) images of WS₂ flakes are shown in Figures S1–S3. First, we introduce the preparation process for chiral WS₂ rolls, as illustrated in Figure 1a. The monolayer isolated WS₂ single-crystal flakes grown on a Si/SiO₂ substrate [23, 24] were vertically immersed in a mixed acetone-ammonia solvent at a constant speed [25]. Due to the strong interaction between the flakes and the substrate, ammonia in the solution slightly etched the Si/SiO₂ surface, facilitating the release of the flake edges from the substrate. Simultaneously, the solvent intercalated between the WS₂ flakes and the substrate, promoting their separation [26]. Driven by surface tension, the detached flakes subsequently rolled up to form rolls, which were then dried using a flow of nitrogen. The study of the factors affecting the curling behavior is shown in Figures S4 and S5. Figure 1b presents a schematic of the two-dimensional chiral structure of TMD materials. Similar to the chiral rolls of graphene, a chiral roll structure is formed when the curvature angle (θ) of the two-dimensional WS₂ transitions between the zigzag and armchair edges. Figures 1c and 1d provide schematic images of left/right-handed WS₂ rolls and their chiral angle θ ($\pm 15^\circ$) as for instance, respectively. The multi-view schematic illustration of the chiral roll structure and statistical analysis of the chiral angles for the synthesized WS₂ rolls are provided in Figures S6 and S7.

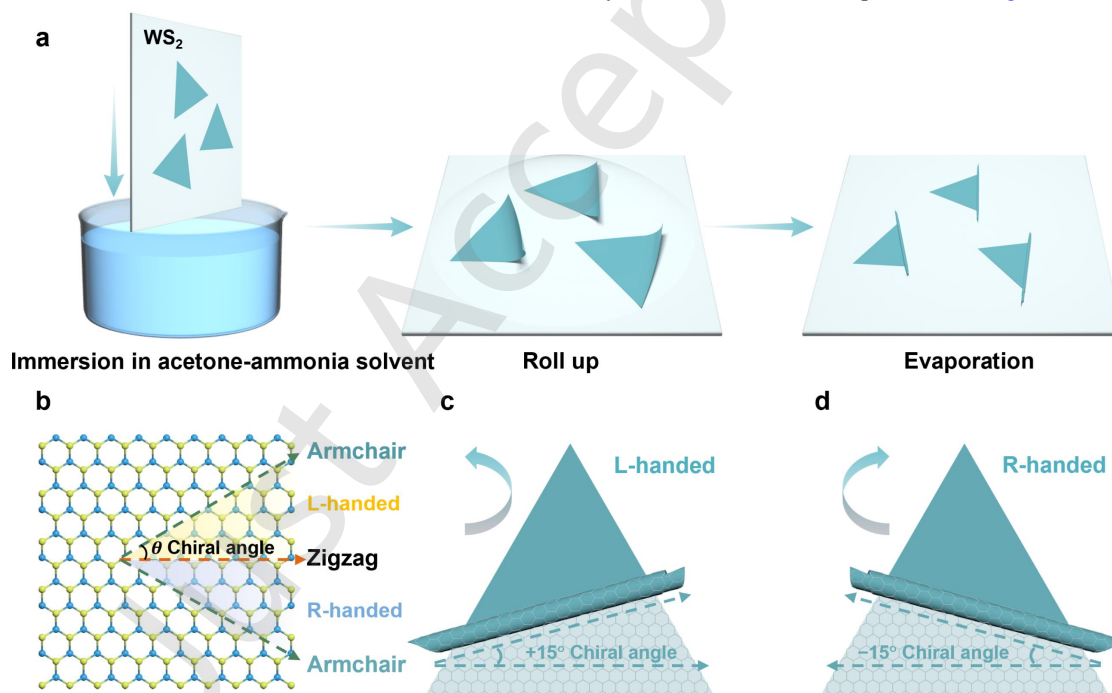


Figure 1. Schematic diagram of the preparation process and chirality origin of chiral WS₂ rolls. (a) Schematic illustration of the fabrication process for WS₂ rolls. (b) Defining the chirality of WS₂ rolls. The zigzag and armchair axes are oriented along the respective crystal lattice chains (red and blue dashed lines). Rolls with their scrolling axes positioned in the yellow and blue regions are classified as left-handed (L-handed) and right-handed (R-handed), respectively. (c, d) Schematics of the left-handed and right-handed WS₂ rolls.

Next, we investigate the morphology, thickness, and microstructure of the chiral WS₂ roll material. Figure 2a presents our optical microscopy (OM) image of the chiral WS₂ roll $\theta = +8^\circ$. And the AFM image (Figure 2b) shows a uniform tube-shaped structure with a height of about 50 nm. Transmission electron microscopy (TEM) characterization reveals a tightly stacked multilayer structure (Figure S8). Subsequently, we explore the unique optical properties of the chiral WS₂ roll. Figure 2c displays stronger and more distinct peaks for the

planar WS₂ (A) compared to the WS₂ roll (B). As the number of roll layers increases, the intensity of the Raman peaks diminishes, which is similar to that observed in multilayer WS₂. The distance between the E_{12g} and A_{1g} peaks of WS₂ is 67 cm⁻¹, and this distance remains unchanged when the WS₂ layers are rolled up. This indicates that rolling only alters the topological structure of WS₂ without changing its chemical structure. Figure 2d illustrates the photoluminescence (PL) spectra of rolled and planar WS₂. Consistent with the Raman spectra, the

photoluminescence intensity of the planar structure is significantly higher than that of the roll structure. This trend

mirrors the lower fluorescence intensity observed in multilayer WS₂ compared to planar WS₂.

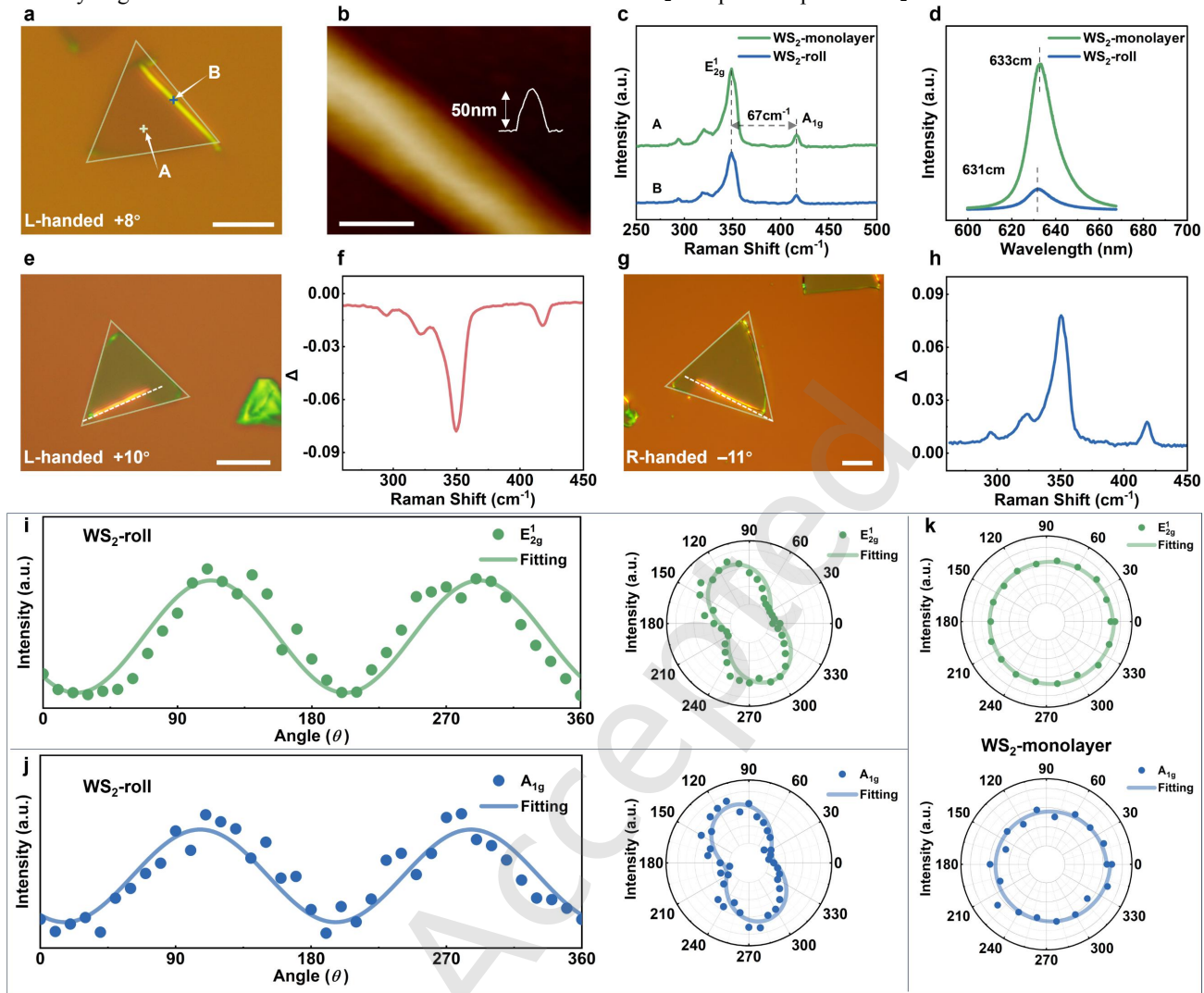


Figure 2. Characterization of the chiral WS₂ rolls. (a) Optical microscopy image of the chiral WS₂ roll. Scale bar, 5 μm. (b) AFM image of the chiral WS₂ roll. Scale bar, 100 nm. (c) Raman spectra of the monolayer WS₂ (A) and WS₂ roll (B) under 532 nm excitation. (d) Photoluminescence (PL) spectra of the WS₂ monolayer and roll under 532 nm excitation. (e, f) OM image (e) and the Raman optical activity (ROA) spectrum (f) of a left-handed WS₂ roll. Scale bar, 10 μm. (g, h) OM image (g) and the ROA spectrum (h) of a right-handed WS₂ roll. Scale bar, 10 μm. (i, j) Angle-resolved polarized Raman spectrum and polar plots of the E'₂g and A₁g modes of the chiral roll. (k) Polar plots of the angle-resolved polarized Raman spectrum for the E'₂g and A₁g modes of the WS₂ monolayer.

Studies on the optical activity of chiral rolls have attracted widespread attention [27]. Herein, we use Raman optical activity to measure the chiral structure of the rolls (Figure S9). Raman optical activity (ROA) is highly sensitive to chiral microstructures. According to classical light scattering theory [28], the parameter Δ is defined as the ratio of the difference to the sum of scattering intensities for left (L) and right (R) circularly polarized incident light:

$$\Delta = \frac{I_L - I_R}{I_L + I_R} \#(1)$$

Figures 2e and 2g provide OM images of left-handed ($\theta = +10^\circ$) and right-handed ($\theta = -11^\circ$) WS₂ rolls, respectively, with the annotations marking their chiral angles. Their corresponding active Raman spectra are displayed in Figures 2f and 2h [29–31]. In order to better compare the optical activity of WS₂ rolls, we define the S factor using the Δ obtained from E'₂g peak as the standard. Their S factors are -0.0785 and 0.0786 , respectively in Figures 2f and 2h. Thus, the S factor data confirms that the left-handed and right-handed WS₂ rolls indeed possess chiral chemical structures and optical activity. These indicate that

chiral rolls exhibit higher optical activity than planar two-dimensional materials, reflecting their inherent chiral superstructure.

To investigate the anisotropic optical properties of chiral WS₂ rolls, the angle-resolved polarized Raman spectra of the chiral rolls were measured. In the experiment, Raman spectroscopy measurements were performed on WS₂ roll structures using a 532 nm linearly polarized laser, and the polarization-dependent behavior was systematically investigated by continuously rotating the incident polarization direction from 0° to 360° . In the angle-resolved Raman intensity maps, two prominent Raman peaks are clearly observed at approximately 349 cm^{-1} and 416 cm^{-1} , corresponding to the E'₂g and A₁g vibrational modes, respectively. The intensities of both Raman modes exhibit pronounced and reproducible periodic variations with the polarization angle (Figures 2i and 2j). When the polarization direction is parallel to the axial direction of the WS₂ roll, the Raman intensity reaches a maximum, whereas it is significantly reduced when the polarization is perpendicular to the roll axis. A clear 180° periodic angular dependence is observed. In contrast,

the Raman intensity of monolayer WS₂ shows no obvious variation (Figure 2k), indicating that the WS₂ roll structure exhibits a strong anisotropy. Repeated measurements for the angle-resolved polarized Raman and ROA characterizations confirmed the stability and reproducibility of the results (Figure S10).

To investigate the optoelectronic properties of chiral WS₂ rolls, a photodetector was fabricated on the rolls, as illustrated in Figure 3a. Device fabrication is described in the Experimental Section. The thickness of the gold electrode was 50 nm (Figure S11). Figure 3b displays the time-resolved photocurrent under illumination across a broad spectral range from 450 nm to 808 nm. The responsivity (*R*), external quantum efficiency (*EQE*) and specific detectivity (*D*^{*}) extracted from these measurements are shown in Figure 3c and Figure S12, confirming its broadband response capability.

Furthermore, we investigated its power-dependent photoresponse characteristics. The time-resolved photocurrent under 450 nm illumination is shown in Figure 3d. The photocurrent exhibits a strong dependence on the incident power

density, increasing with higher power levels, as further supported by the *I-V* characteristics under varying power densities (Figure S13). Figure 3e shows the photocurrent and the on/off ratio as a function of the incident power density. The power-dependent photocurrent was fitted with the power-law equation $I_{ph} \propto P^\alpha$. $\alpha = 1.1$, indicating a near-linear response. Under 450 nm illumination at a power density of 47.2 mW cm⁻², the photocurrent reaches 3.7 nA with an on/off ratio of 1.4×10^2 . Key performance metrics extracted from these measurements (Figures 3f and 3g) exhibit excellent device performance. The maximum *R*, *EQE*, and *D*^{*} values reach 210 mA W⁻¹, 58% and 4.7×10^8 Jones. Detailed calculations are provided in Note S1 and the Experimental Section. Under 450 nm illumination temporal photoresponse measurements reveal a rapid rise time of approximately 84 ms and a decay time of approximately 86 ms (Figure 3h). Moreover, the device exhibits excellent operational stability, with its photoresponse remaining stable during continuous testing under 450 nm illumination at a bias voltage of 20 V (Figure 3i). These results demonstrate that chiral WS₂ rolls exhibit good photodetection performance.

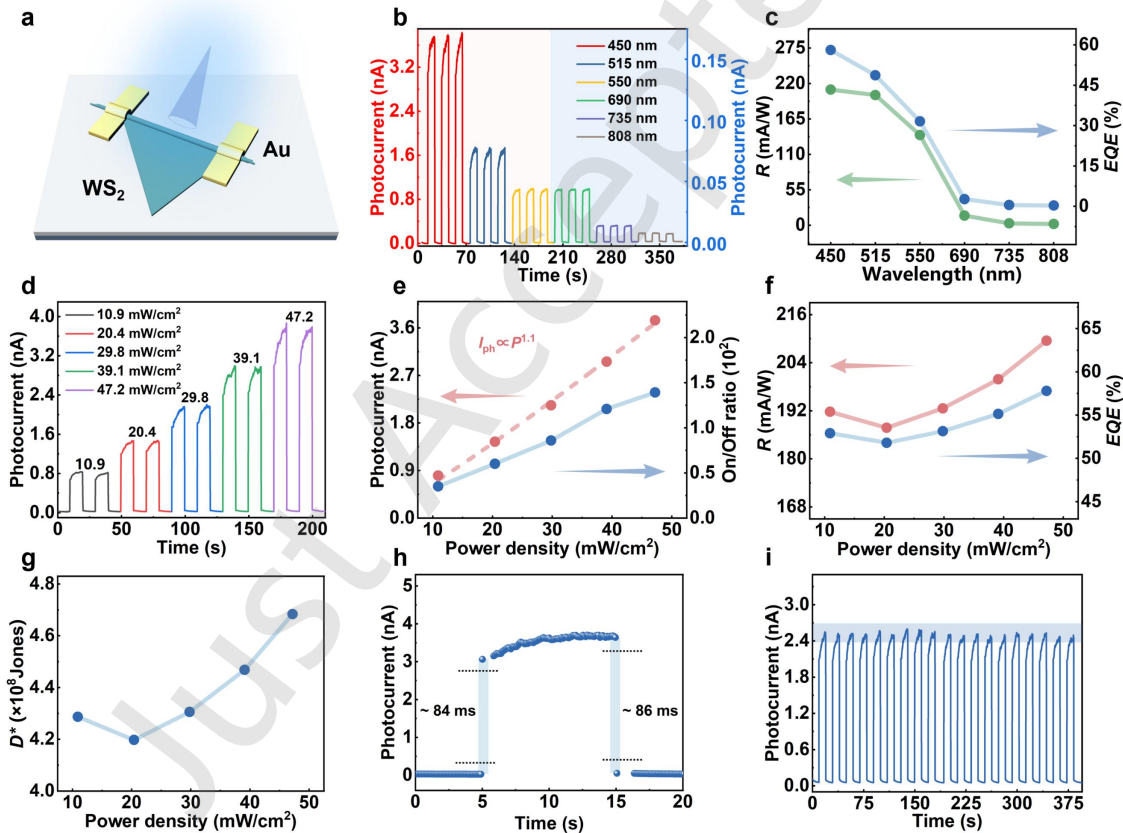


Figure 3. Optoelectronic characterization of the chiral WS₂ roll photodetector. (a) Schematic illustration of the chiral WS₂ roll photodetector. (b) Broadband photoresponse of chiral WS₂ roll photodetector from 450 to 808 nm at 20 V bias. (c) Wavelength-dependent responsivity and external quantum efficiency. (d) Photocurrent under various light power densities at 450 nm illumination. (e) Power-dependent photocurrent and on/off ratio of the photodetector. (f) *R* and *EQE* of the photodetector. (g) Specific Detectivity (*D*^{*}) of the photodetector. (h) Response time of the photodetector under 450 nm illumination at 20 V bias. (i) Time-resolved photocurrent stability of the photodetector under 450 nm illumination at 20 V bias.

TMD rolls have demonstrated great potential in linearly polarized light detection [32]. However, in this work, we prepared chiral WS₂ rolls. The chiral structure endows them with both anisotropic and chiral properties, allowing us to realize a photodetector for circularly/linearly polarized light, simultaneously discriminating linearly and circularly polarized light. Based on these advantages, we investigated the CPL and LPL detection performance of the chiral WS₂ roll photodetector.

A schematic of the polarization measurement system is presented in Figure 4a, where a polarizer was used to linearly polarize the 450 nm light and the polarization state was modulated using a half-wave ($\lambda/2$) plate or a quarter-wave ($\lambda/4$) plate [33, 34]. Figures 4b–d display the polarization-dependent photocurrent responses of non-chiral roll, right-handed roll, and left-handed roll photodetectors, respectively, under 450 nm illumination at 50 V bias. The measured photocurrent exhibited

a periodic cosine variation. The corresponding angle-resolved polar plots of photocurrent (Figures 4e–g) exhibit a typical “8” pattern, indicating pronounced LPL detection capability. The photocurrent anisotropy ratios ($I_{ph,max}/I_{ph,min}$) for the non-chiral, right-handed, and left-handed devices reach 1.60, 1.59, and 1.57, respectively. This result demonstrates that the photodetector based on rolls exhibits an anisotropic photoresponse to LPL. Then, we discuss the mechanism underlying the LPL response of the chiral rolls. We believe that quasi-1D TMD roll structures are crucial for the emergence of the anisotropic electrical response under LPL. As evidenced by the polarized Raman measurements for planar WS₂ in Figure 2k, planar WS₂ exhibits essentially no detectable polarization anisotropy. This contrast suggests that the polarization selectivity observed in the rolled structures originates from the structural and symmetry evolution induced by rolling.

Previous literature emphasizes that polarization is inherently a symmetry-defined property of light: linear polarization carries anisotropic symmetry. In the photoelectric conversion process, light absorption couples to relevant electronic degrees of freedom such as charge, spin, and/or valley, and subsequently

generates scalar or vector currents. Importantly, this conversion is fundamentally governed by the symmetry of both the material and the device. By invoking symmetry principles, one can transcend material-specific explanations and identify universal selection rules [17]. For instance, scalar responses to linearly polarized light require C_1 or C_2 symmetry, reflecting broken rotational symmetry, whereas vector responses arise from broken inversion symmetry, independent of the specific material platform.

In our case, planar WS₂ possesses D_{3h} symmetry and therefore does not show polarization-sensitive behavior (Figure 2k). When planar WS₂ is rolled into a quasi-1D structure, its symmetry is reduced. For chiral roll structures, the symmetry becomes C_1 ; for non-chiral roll structures, it becomes C_2 . This symmetry reduction enables the chiral WS₂ rolls to become polarization-sensitive. In summary, roll-shaped TMD materials exhibit low intrinsic crystalline symmetry characteristic of quasi-1D systems (C_1 or C_2). As a consequence, anisotropic electronic transition dipole moments lead to polarization-dependent absorption, thereby producing a sensitive electrical response to linearly polarized light.

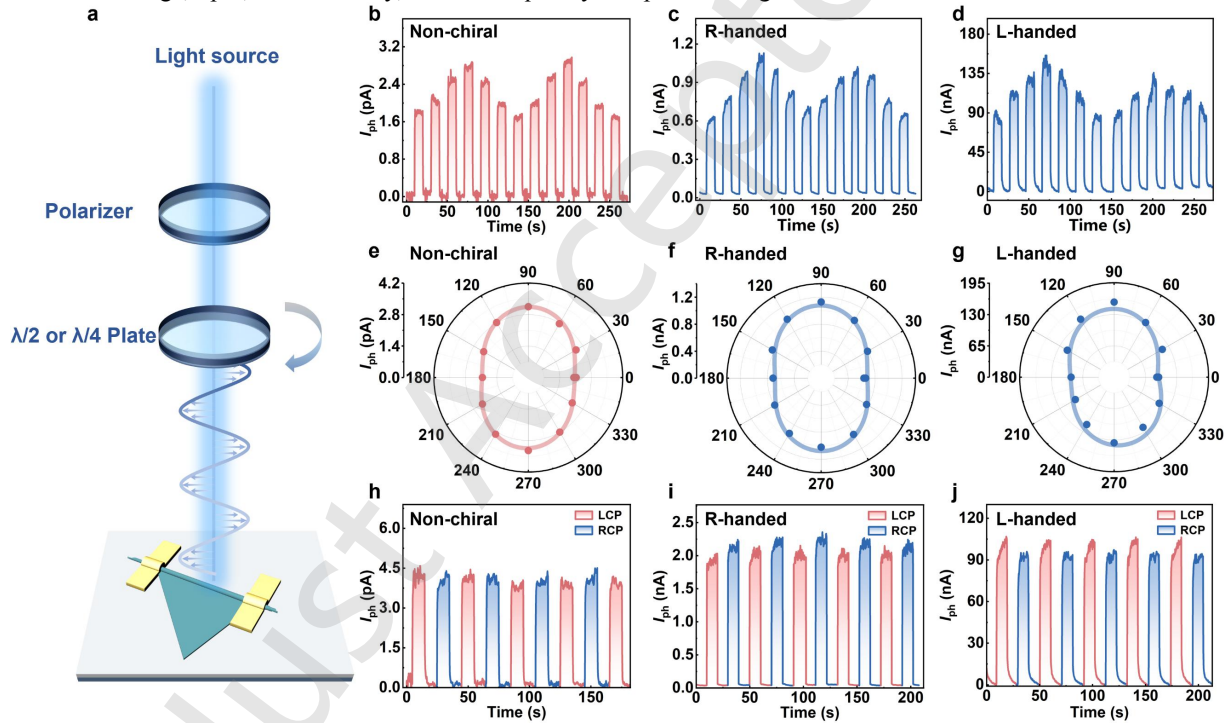


Figure 4. LPL and CPL detection capabilities of the chiral WS₂ roll photodetector. (a) Schematic illustration of the LPL and CPL detection measurement. (b–d) Polarization-dependent photocurrent responses of non-chiral roll (b), right-handed roll (c), and left-handed roll (d) photodetectors under 450 nm illumination. (e–g) Angle-resolved polar plots of photocurrent of non-chiral roll (e), right-handed roll (f), and left-handed roll (g) photodetectors. (h–j) Photocurrent of non-chiral roll (h), right-handed roll (i), and left-handed roll (j) photodetectors under LCP and RCP illumination (450 nm).

We further evaluated the CPL detection performance of the three photodetectors. Figures 4h–j show the photocurrent responses of non-chiral roll, right-handed roll, and left-handed roll photodetectors under 450 nm illumination at 50 V bias. The non-chiral roll photodetector showed no significant difference in photocurrent under LCP and RCP illumination (Figure 4h). In contrast, the right-handed roll photodetector exhibited a stronger photocurrent under RCP illumination compared to under LCP illumination (Figure 4i). The photocurrent asymmetry factor g_{ph} was calculated using the expression

$$g_{ph} = 2 \frac{I_{LCP} - I_{RCP}}{I_{LCP} + I_{RCP}} \#(2)$$

The right-handed roll photodetector exhibited a g_{ph} of -0.09 . Conversely, the left-handed roll photodetector showed the opposite trend (Figure 4j), with a g_{ph} of 0.10 . This can be explained by Raman optical activity (Figures S14 and S15). The non-chiral WS₂ roll showed almost no chiral optical activity, and thus exhibited no differential photocurrent response to LCP and RCP. In contrast, rolls with a chiral angle exhibited significant optical activity due to their chiral structure. Their unique chiral optical activity leads to differential photocurrents under CPL illumination, enabling circular polarization detection. Furthermore, we studied the CPL detection capability of several chiral WS₂ rolls (Figure S16). The devices showed differential

photocurrent responses under LCP and RCP illumination, which confirmed the reliability of chiral WS₂ rolls for CPL detection. Overall, the chiral WS₂ roll devices not only retain the linear polarization detection capability of other TMD rolls (Table S1), but also achieve circularly polarized light detection. Although the polarization detection performance of chiral WS₂ roll devices still requires enhancement relative to other materials possessing both linear and circular polarization capabilities (Figure S17), their simple structure and facile fabrication provide significant benefits for miniaturized system integration. In accordance with our results, chiral WS₂ rolls exhibit similar performance in LPL detection comparable to that reported in previous literature [32, 35]. Furthermore, they also exhibit a degree of CPL detection capability. This linear/circular polarization sensitivity enhances the utility of chiral WS₂ rolls in various applications, enabling them to effectively respond to different polarization states of light, thereby supporting both linearly and circularly polarized light imaging. Thus, we investigated the imaging performance of the WS₂ rolls photodetectors for LPL and CPL. The imaging setup is shown in Figure 5a. Upon polarized light illumination, different photocurrent signals are generated as the “TJU” mask moves. This process enables the recording of photocurrent at each position, and the “TJU” pattern is displayed under both LPL and CPL. Under 450 nm illumination, images under different polarizations were obtained by changing the polarization state of the incident light. As shown in Figure 5b, the linearly polarized imaging results of non-chiral (1) and chiral (2) WS₂ roll detectors are presented. Both detectors show clearer images at a polarization angle of 90° than at 0°. Figure 5c further shows the

circularly polarized imaging results of non-chiral and chiral WS₂ roll detectors. The non-chiral detector (1) exhibits relatively consistent imaging clarity under left-handed and right-handed circularly polarized light. The chiral detector (2) shows differentiated imaging results under left-handed and right-handed circularly polarized light. These results demonstrate the application capability of WS₂ roll detectors in the field of linearly and circularly polarized imaging.

Overall, our results show that chiral WS₂ rolls exhibit remarkable efficacy in linear polarization detection while seamlessly incorporating circular polarization sensitivity. This unique combination positions them as a formidable alternative within the optoelectronics field. The polarization sensitivity not only broadens the spectrum of potential applications but also underscores the critical role of chirality in optimizing optical detection mechanisms.

Chiral WS₂ rolls signify a substantial leap forward in the evolution of next-generation polarization-sensitive devices. Their ability to respond effectively to various polarization states exemplifies their versatility and adaptability. Furthermore, when compared to other dual sensitive linear/circular polarization photodetectors harnessing chiral materials [36-39], chiral WS₂ rolls stand out due to their compact size and cost-effectiveness. This intrinsic advantage facilitates their seamless integration with existing semiconductor technologies, enhancing functionality without the need for complex fabrication methods. Ultimately, this positions chiral WS₂ rolls as not just a technological innovation but as a transformative component in the future of optoelectronic devices.

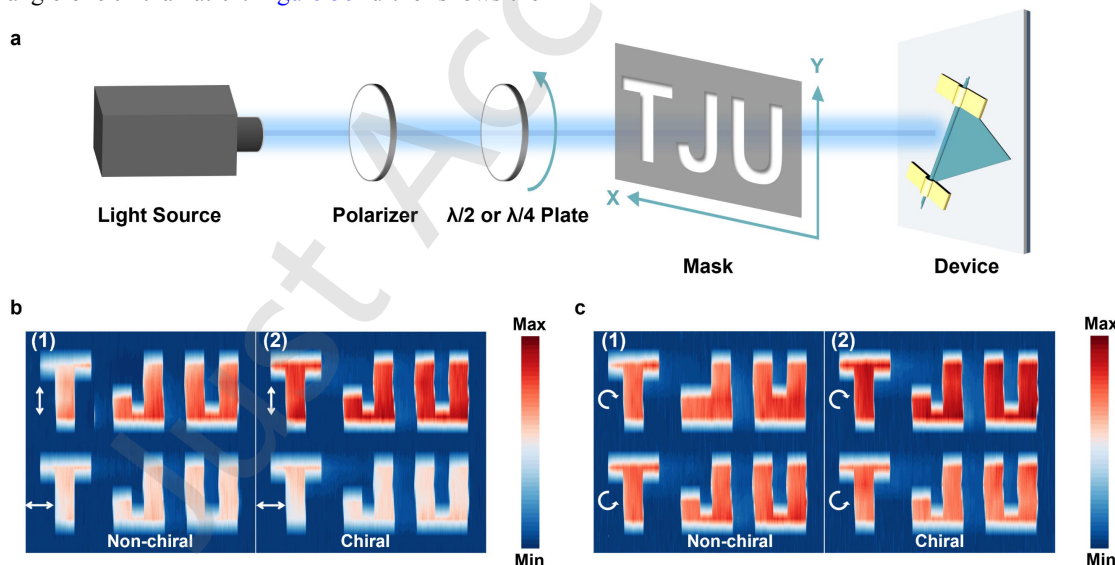


Figure 5. Imaging applications based on WS₂ roll photodetectors. (a) Schematic diagram of linearly polarized and circularly polarized imaging systems. (b) Imaging results of the “TJU” pattern for the non-chiral roll photodetector (1) and chiral roll photodetector (2) at polarization angles of 0° and 90° under 450 nm illumination. (c) Imaging results of the “TJU” pattern for the non-chiral roll photodetector (1) and chiral roll photodetector (2) under 450 nm CPL illumination.

3 Conclusions

In this study, we explored the unique capabilities of chiral 1D WS₂ rolls, which can detect both LPL and CPL. This sensitivity enhances versatility in optical sensing and communication applications and improves the detection of chiral molecules. In the future, the integration of chiral WS₂ rolls into circular/linear polarization-sensitive photodetectors significantly boosts the performance of optoelectronic devices by enabling precise polarization detection. This advancement not only expands the range of potential applications but also showcases innovative

uses across various fields, highlighting the transformative potential of chiral WS₂ rolls in advancing technology.

4 Experimental

CVD growth of WS₂

The Si/SiO₂ substrate spin-coated with a 1 mg/mL Na₂WO₄ precursor solution was placed on a quartz plate at the center of the chemical vapor deposition (CVD) furnace. Another clean Si/SiO₂ substrate was placed face-down on top of the precursor-coated substrate. Sulfur powder (0.08 g) was placed upstream in the furnace at a distance of 19 cm from the Si/SiO₂ substrate.

Before the heating program, the quartz tube was flushed with Ar gas (1000 sccm) for 2 minutes. Then the furnace was heated to 850°C in 25 minutes, and annealed at this temperature for 10 minutes. Finally, the sulfur powder (pre-heated to 200 °C) was introduced into the furnace. The reaction proceeded for 20 minutes under a gas flow of 200 sccm Ar and 10 sccm H₂. After the reaction, the system was naturally cooled to room temperature.

Optical characterization

Raman optical activity was conducted using a custom ultra-low wavenumber Raman system with 532 nm excitation. To prevent thermal damage to the chiral WS₂ rolls, the laser power was restricted to 5 mW with a 10 s acquisition time. The polarization states were modulated sequentially using a combination of waveplates, linear polarizers and a quarter-wave plate in the excitation path. Specifically, the final quarter-wave plate was employed to ensure that left and right circularly polarized light irradiated the sample with uniform intensity. This setup allowed us to systematically investigate whether the material exhibits Raman responses to circularly polarized light. The backscattered signal was collected through a linear analyzer and dispersed by an Andor Shamrock 303i spectrograph equipped with a 1200 grooves per millimeter grating, yielding a spectral resolution of approximately 3.5 cm⁻¹.

Device fabrication

Two parallel Au electrodes were patterned onto the WS₂ roll through lithography and lift-off processes. The sample on a Si/SiO₂ substrate was placed on a spin coater. A layer of LOR1A was first spin-coated to create an undercut structure. The substrate was then baked at 195°C for 5 minutes to remove solvent and harden the resist, to prevent collapse after development. Next, a layer of S1805 photoresist was spin-coated and baked at 115 °C for 1 min. A maskless laser direct writer was used to align and expose two parallel electrode patterns. The exposed substrate was developed in ZX-238 developer for 1 min at room temperature, rinsed in deionized water for 1 min to remove residual developer, and finally dried with N₂ flow. A 50 nm-thick Au electrode layer was deposited by magnetron sputtering. The substrate was immersed in N-methylpyrrolidone (NMP) for lift-off, then rinsed in acetone to remove residual NMP, followed by isopropanol to remove acetone. The substrate was dried with N₂ flow, completing device fabrication.

Performance metrics for photodetectors

Photoresponsivity $R = (I_{\text{light}} - I_{\text{dark}})/(PS)$, where I_{light} and I_{dark} are the currents under illumination and dark, P is the power density of the incident light, S represents the effective radiation area. External quantum efficiency $EQE = hcR/\lambda e$, Specific Detectivity $D^* = R/(S/2eI_{\text{dark}})^{1/2}$, where h is Planck's constant, c is the speed of light in a vacuum, λ is the incident light wavelength, and e is the electron charge.

Electronic supplementary material

Schematic illustration of CVD growth of WS₂, optical microscopy image, Raman spectra and AFM image of the monolayer WS₂, the factors affecting the curling behavior, multi-view schematic illustration of the chiral roll structure and statistical analysis of the chiral angles, TEM image of a WS₂ roll, schematic of the ICP ROA spectrometer system, optical microscopy image and Raman Optical Activity (ROA) spectrum of WS₂ roll, AFM image of the Au electrode, Wavelength-

dependent D^* and I - V characteristic curves of the WS₂ chiral roll photodetector, CPL detection capability of several chiral WS₂ rolls, comparison of the detection capability.

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

Yi Zhen: Experimental investigation, formal analysis, visualization, writing original draft, writing review & editing.

Xianfeng Shen: Experimental investigation, formal analysis, visualization, writing review & editing. Ying Xu: Experimental investigation, formal analysis. Wei Gao: Experimental investigation, writing original draft. Xiangyun Ma: formal analysis, writing original draft, writing review & editing. Yi Sun: Experimental investigation, formal analysis, writing review & editing. Zhuwei Zhang: Conceptualization, supervision. Qifeng Li: Conceptualization, supervision. Hechen Ren: Conceptualization, supervision. Rongjin Li: Conceptualization, supervision. Yongtao Shen: Conceptualization, supervision, writing review & editing, funding acquisition.

Use of AI statement

None.

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Electronic Supplementary Material

Application of chiral WS₂ rolls in circular/linear polarization-sensitive photodetectors

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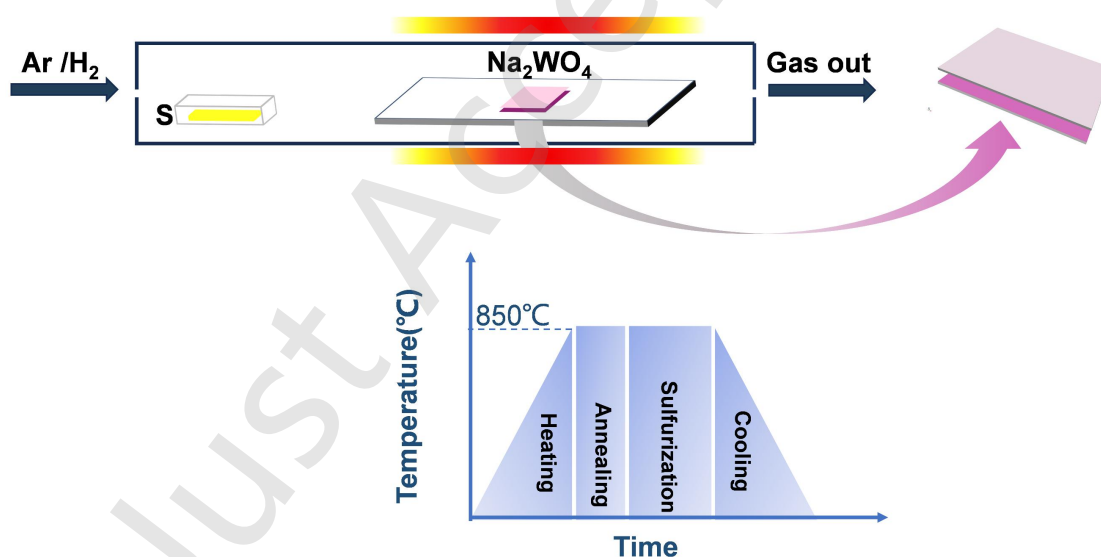


Figure S1. Schematic illustration of CVD growth of WS₂.

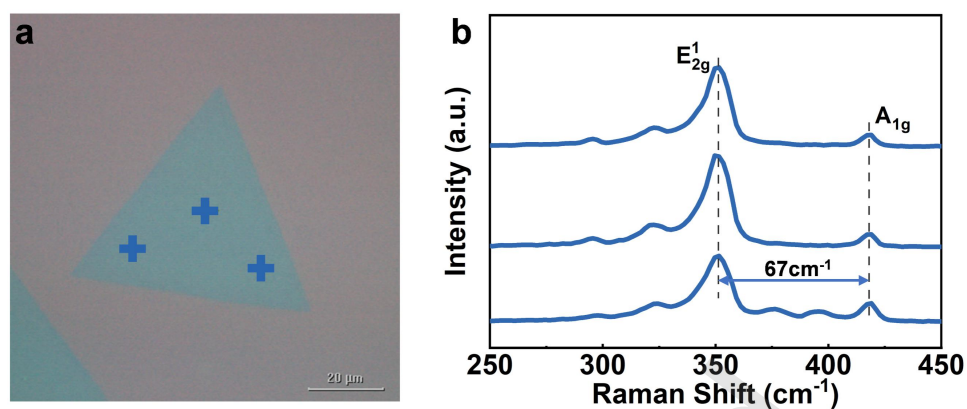


Figure S2. Optical microscopy image (a) and Raman spectra (b) of the monolayer WS₂.

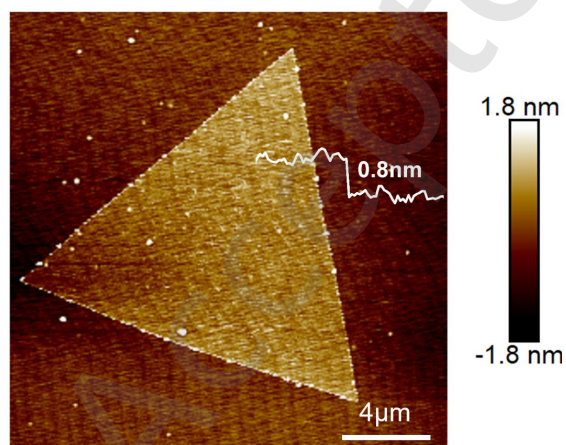


Figure S3. AFM image of the monolayer WS₂.

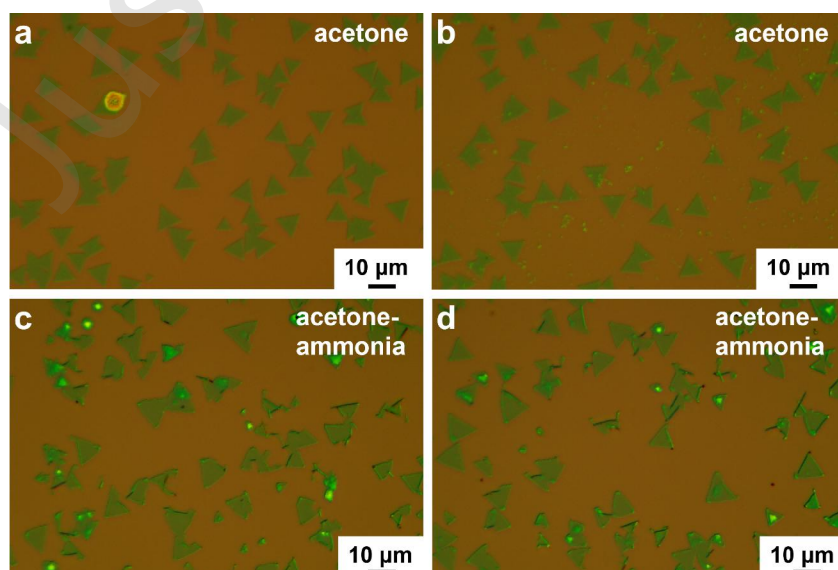


Figure S4. (a, b) Rolling of monolayer WS₂ treated with acetone; (c, d) Rolling of monolayer WS₂ treated with acetone and ammonia solution.

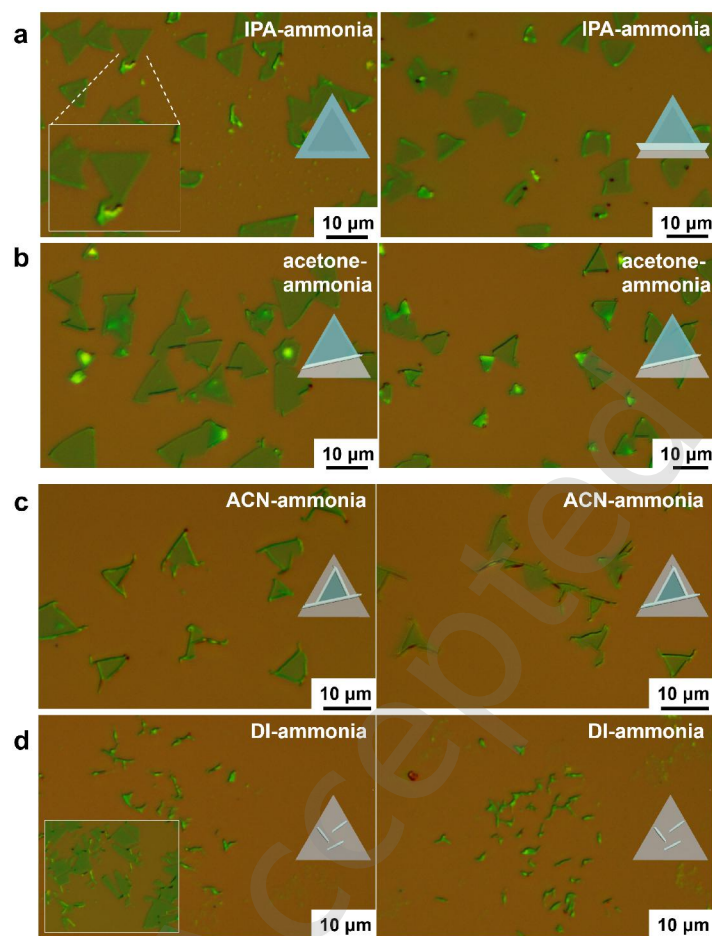


Figure S5. (a–d) Rolling images of monolayer WS₂ treated with isopropanol (IPA)-ammonia, acetone-ammonia, acetonitrile (ACN)-ammonia, and deionized (DI) water-ammonia, respectively.

As shown in Figure S4 (a, b), without the addition of ammonia solution and under the action of acetone solvent alone, the sample exhibited almost no curling behavior. The edges of the flakes were difficult to detach from the substrate surface, resulting in difficulty in the occurrence of curling. In contrast, when a small amount of ammonia solution was introduced into acetone, the edges of the flakes were more easily released, making curling prone to occur, and as shown in Figure S4 (c, d), a large number of WS₂ flakes underwent curling and rolling. This is because the dangling bonds at the growth edges of WS₂ flakes may form disordered bonding with the substrate, or adsorb small particle impurities, and these factors can significantly enhance the interfacial force between the WS₂ domain boundaries and the Si/SiO₂ substrate, causing the flake edges to be “pinned” on the substrate, thereby inhibiting the occurrence of curling. The mild etching effect of ammonia solution facilitates the release of the edges, thereby driving curling. Furthermore, from the curling images under solutions of different polarities (Figure S5), it can be seen that isopropanol with lower polarity cannot effectively drive curling, and as the polarity increases, the curling driving force increases, and the curling degree is enhanced to some extent. When water with the highest polarity was used as the curling solvent, it was found that curling occurred to a greater extent, and there were obvious traces of WS₂ detachment from the substrate. In order to better determine the chiral angle, acetone-ammonia mixed solution was ultimately selected as the curling solution.

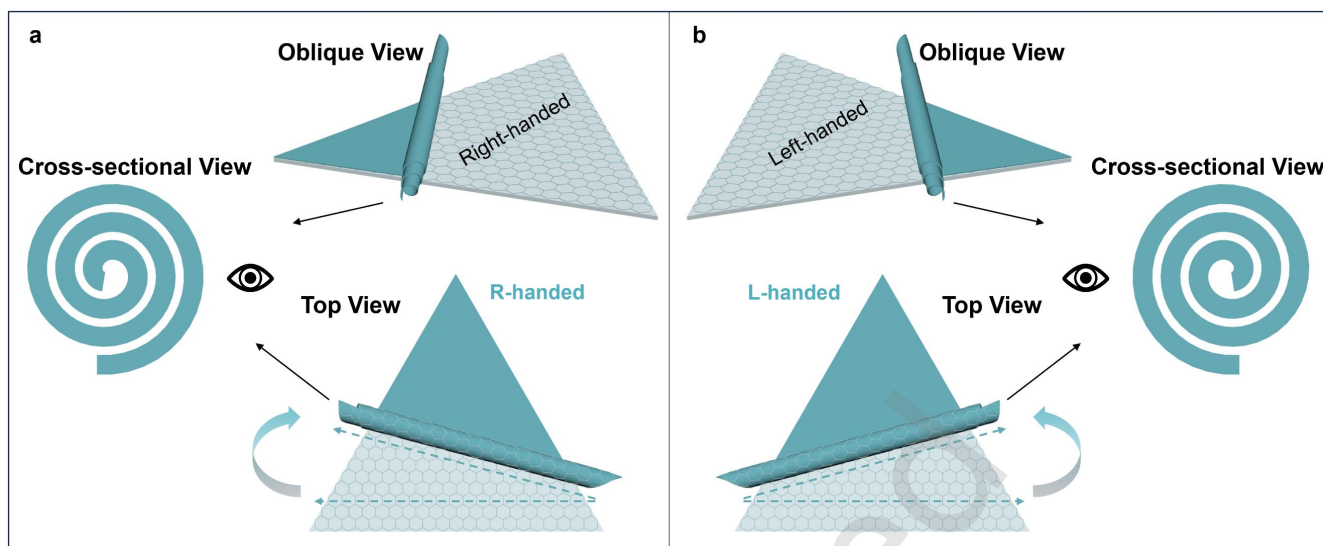


Figure S6. Schematic illustration of chiral WS₂ roll structures. (a, b) Oblique view, top view, and cross-sectional view of right-handed (a) and left-handed (b) chiral WS₂ rolls. The eye symbol indicates the viewing direction from which the cross-sectional view is obtained.

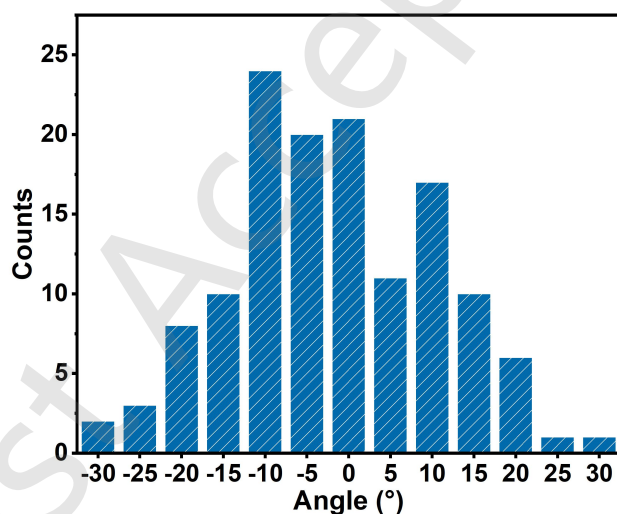


Figure S7. Statistical distribution of chiral angles for the WS₂ rolls.

As shown in Figure S7, statistical measurements of chiral angles were conducted for a large number of WS₂ rolls. Negative values denote right-handed chirality (-30° to 0°), positive values denote left-handed chirality (0° to 30°) and 0° and ±30° are considered achiral. According to the statistical results, chiral angles are mainly concentrated in the ranges of -15° to -5° and 5° to 15°. A considerable proportion is also observed near 0°, while large angles ($|\theta| > 20^\circ$) constitute a minor fraction.

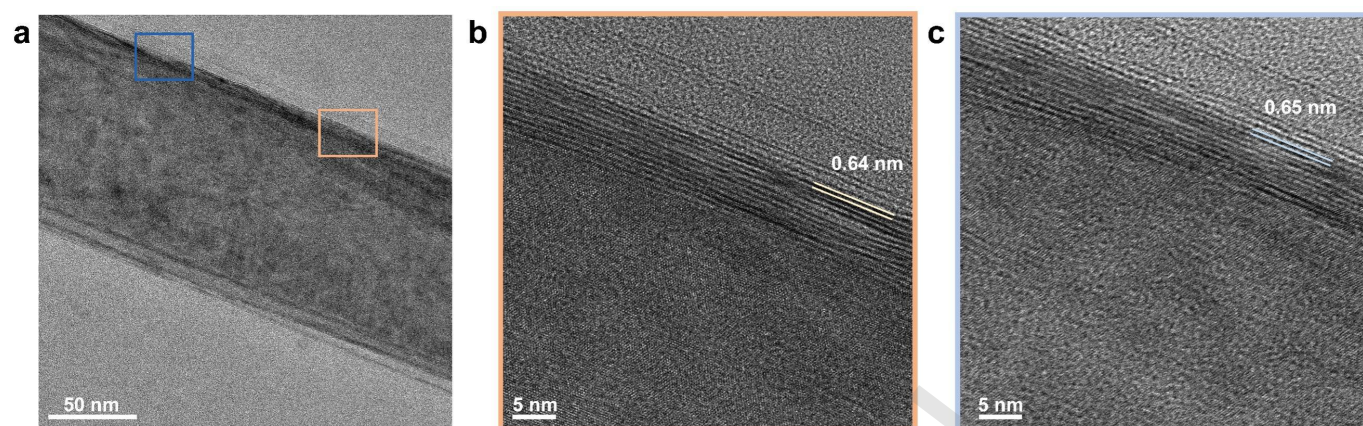


Figure S8. (a) TEM image of a WS₂ roll. Magnified images of the (b) orange and (c) blue-boxed areas in a.

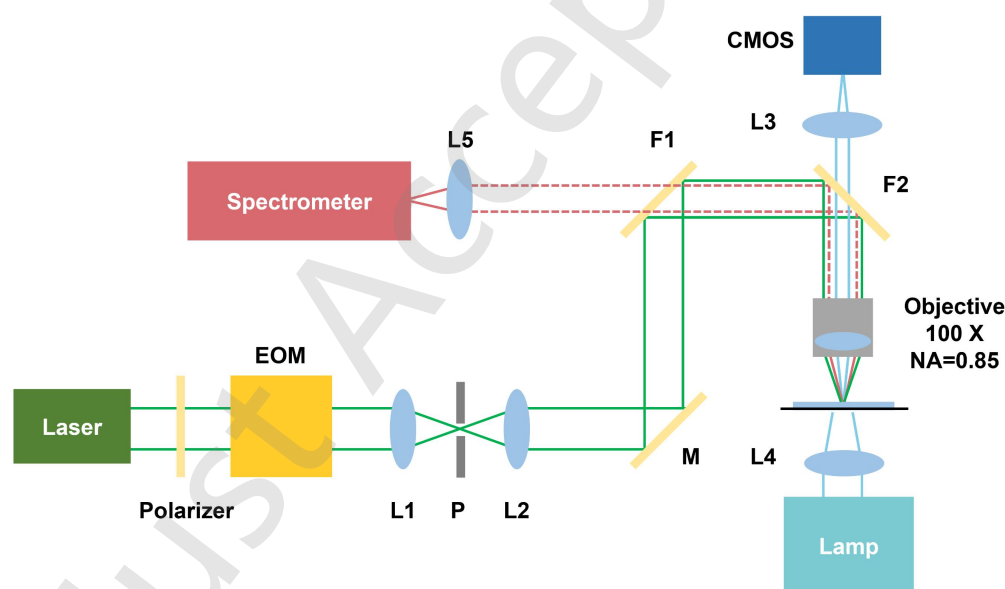


Figure S9. Schematic of the ICP ROA spectrometer system. Key optical elements are labeled in the schematic. The optical paths are simply indicated by coloured lines. The elements labeled L1 to L5 are achromatic doublets and M is a mirror. F1 is a long-pass dichroic mirror with a cutoff wavelength of 540 nm. F2 is a short-pass dichroic mirror with a cutoff wavelength of 500 nm. The size of the pinhole P is 40 μm . The numerical aperture (NA) of the objective lens is 0.85.

The polarization state of the laser beam was first defined by a linear polarizer, providing a linearly polarized excitation with high polarization purity. An electro-optic modulator (EOM), driven by a high-voltage linear differential amplifier, was employed to achieve electrical control of the incident light polarization state. By precisely adjusting the driving voltage applied to the EOM, continuous modulation of the incident linear polarization direction, as well as controlled conversion from linear polarization to left- and right-handed circular polarization, was realized without altering the optical path, thereby enabling angle-dependent linearly polarized Raman and Raman optical activity measurements.

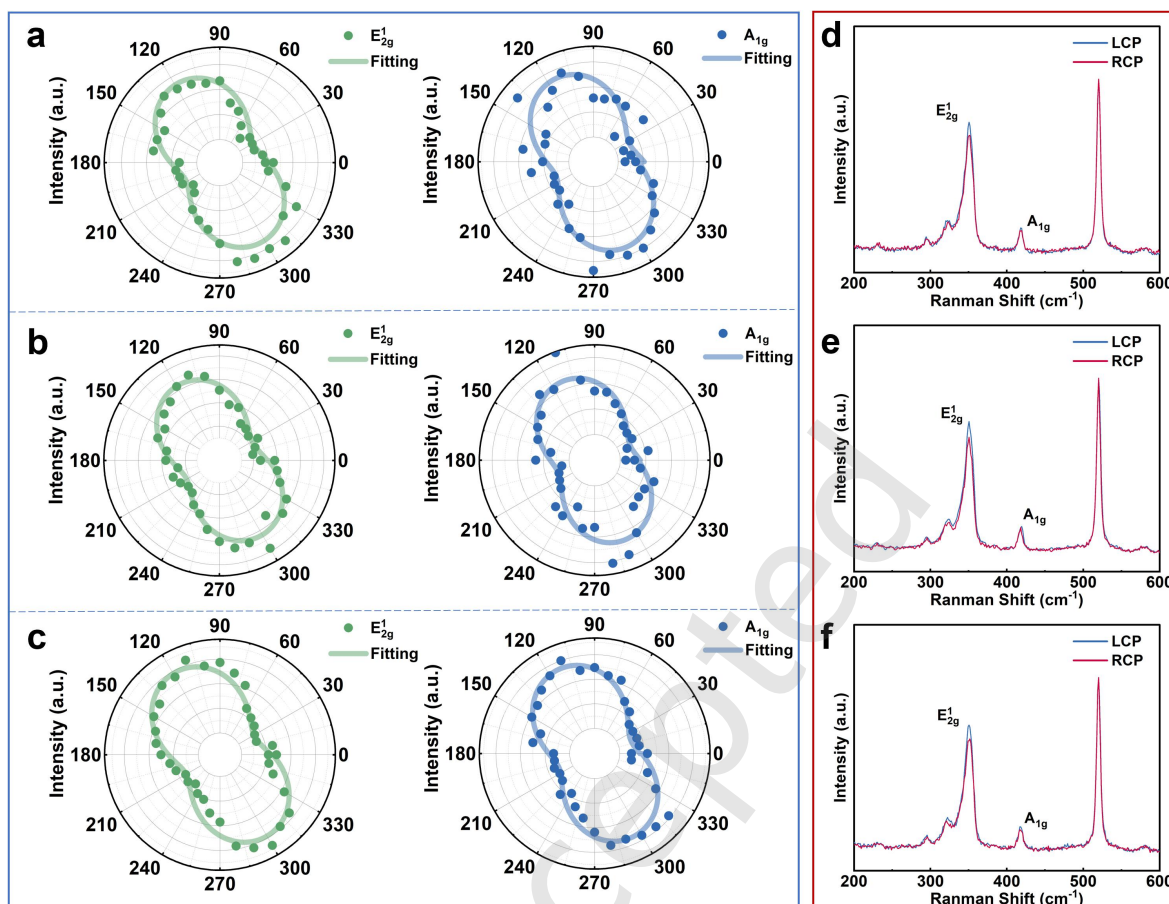


Figure S10. (a–c) Polar plots and the corresponding fitting of the Raman E_{12g}^1 and A_{1g} modes of the WS₂ roll from three measurements, and (d–f) three Raman spectra measurements of the WS₂ roll under 532 nm left- and right-handed circularly polarized light excitation.

Multiple repeated measurements were conducted for the polarization-resolved Raman and Raman optical activity (ROA) characterizations, as presented in Figure S10. Specifically, the polarization-resolved Raman measurements all exhibited a 180° periodic variation (Figure S10a–c). For the ROA characterization, the repeated Raman spectra under left- and right-handed circularly polarized excitation showed differential peak intensities with good trend consistency, with deviations remaining within 10% (Figure S10d–f). The overall results demonstrate satisfactory stability and reproducibility.

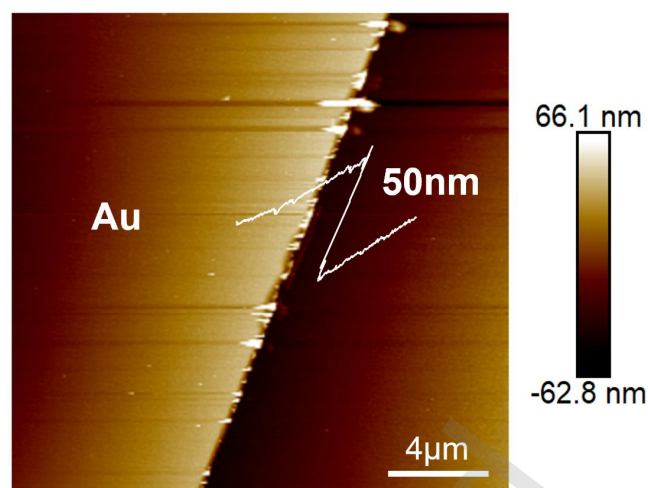


Figure S11. AFM image of the Au electrode.

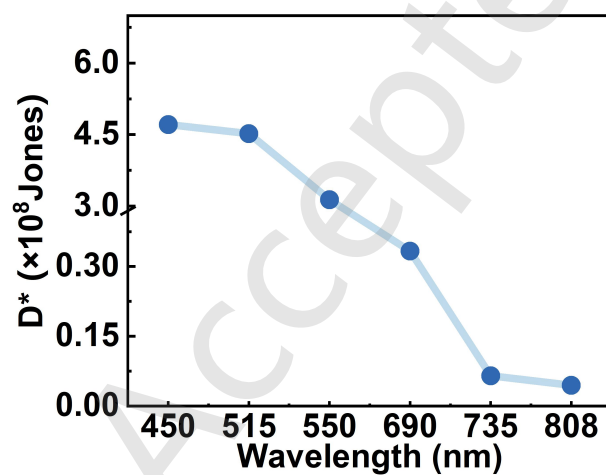


Figure S12. Wavelength-dependent Specific Detectivity of the WS_2 chiral roll photodetector at 20 V bias.

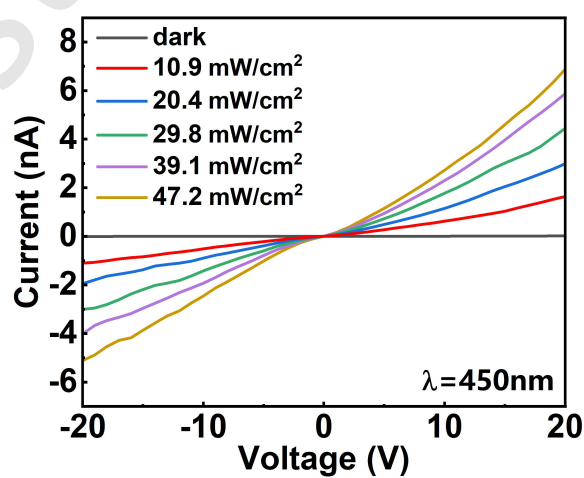


Figure S13. I - V characteristic curves of the WS_2 roll photodetector under 450 nm illumination.

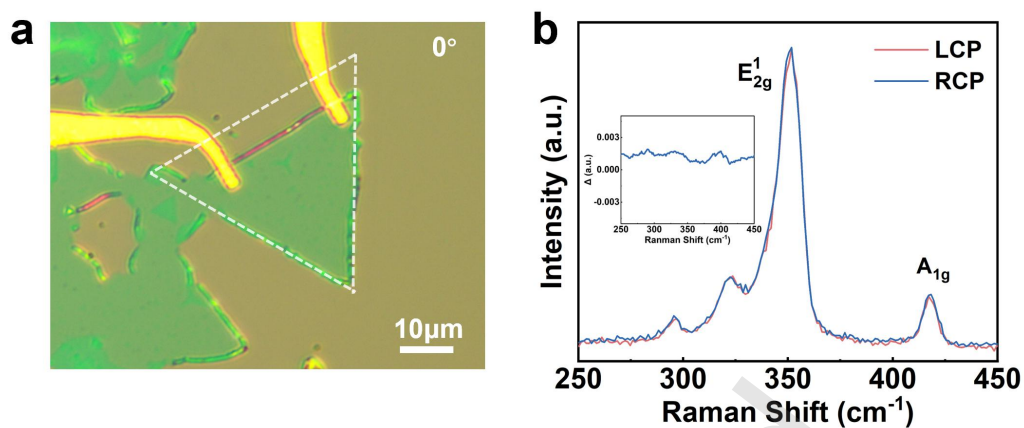


Figure S14. (a) Optical microscopy image of the 0° WS₂ roll. (b) Raman spectra of the 0° WS₂ roll excited by 532 nm left- and right-handed circularly polarized light. The inset shows the Raman optical activity (ROA) spectrum.

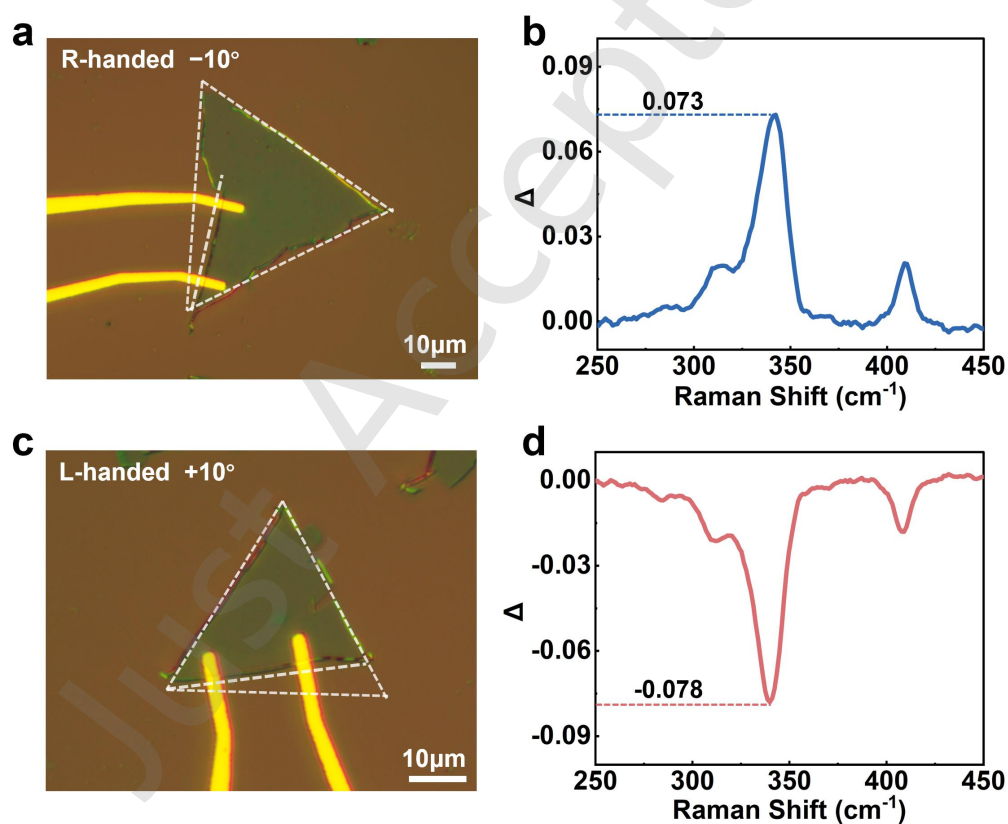


Figure S15. (a,b) Optical microscopy image and Raman optical activity (ROA) spectrum of the -10° WS₂ roll. (c,d) OM image and ROA spectrum of the $+10^\circ$ WS₂ roll.

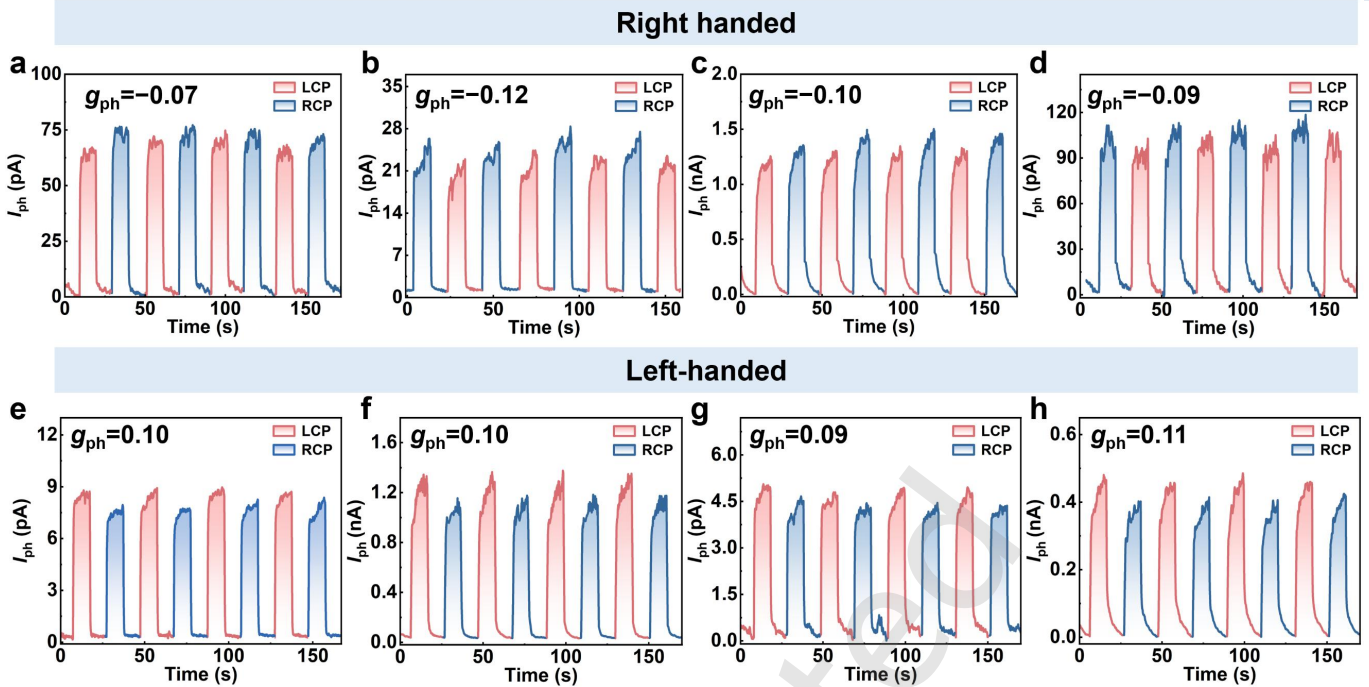


Figure S16. Photocurrent responses of four right-handed (a–d) and four left-handed (e–h) WS₂ roll devices under LCP and RCP light illumination.

Table S1. Dichroic ratio (*DR*) of TMD roll-based photodetectors.

TMD rolls	<i>DR</i>	Ref.
WSe ₂ Nanoscroll-2D WSe ₂ Homojunctions	1.96	Zhang et al. ^[1]
WSe ₂ Nanoscroll	1.5	Ao et al. ^[2]
This work (WS₂ roll)	1.6	

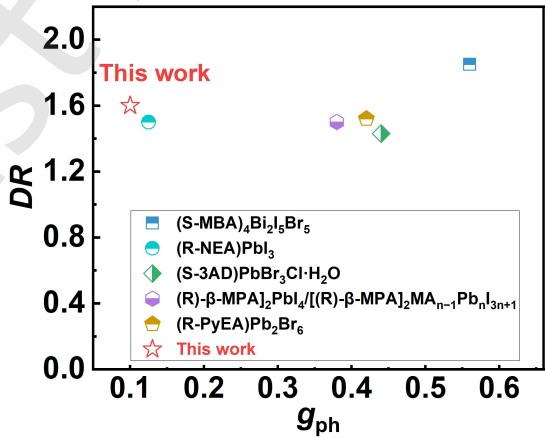


Figure S17. Comparison of the dichroic ratio (*DR*) and photodetection anisotropy factor (g_{ph}) between this work and reported chiral perovskite materials ^[3–7].

Note S1. Calculation of specific detectivity (D^*)

The specific detectivity (D^*) was calculated as:

$D^* = R(S/2eI_{\text{dark}})^{1/2}$, where R is the responsivity, I_{dark} is the dark current, S is the effective radiation area, and e is the elementary charge.

The responsivity R was determined by:

$R = (I_{\text{light}} - I_{\text{dark}})/(PS)$, where I_{light} and I_{dark} are the photocurrent and dark current, respectively, P is the incident light power density, and S is the effective radiation area. The D^* versus power density curve in Figure 3g was obtained from the above equations using the measured data at different power densities.

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