SUPPORTING INFORMATION

Circular Dichroism Control of Tungsten Diselenide (WSe_2) Atomic Layers with Plasmonic Meta-Molecules

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Monolayer Tungsten Diselenide (WSe_2) Characterization

The luminescence of two-dimensional transition mental dichalcogenide monolayers (TMDCs) are layer dependent, and the circular polarization selective absorption in the K_1 and K_2 valleys occurs only in TMDCs with an odd number of layers. Therefore, being able to recognize a single layer of tungsten diselenide (WSe_2) is valuable. This study adopted Raman spectroscopy to identify the properties of a single WSe_2 layer. Figure S1 displays the Raman spectra for WSe_2 excited by a 532-nm laser, showing the characteristic peaks E^1_{2g} and A_{1g}, which are consistent with previous reports[1, 2]. When moving to high-energy bands, we could not observe the characteristic B^1_{2g} peak at 308 cm^{-1}, related to interlayer interaction[1, 2]. These results indicate that the WSe_2 observed in this study was indeed a monolayer.

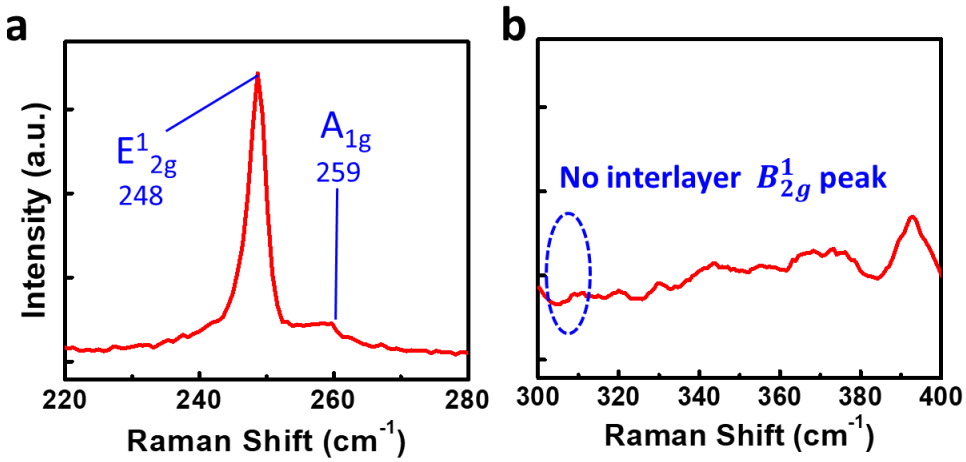

Figure S1. Measured Raman spectra of the WSe_2; (a) and (b) display different wavenumber regions. In (a), the two peaks at 248 and 259 cm^{-1} represent the characteristic peaks of WSe_2. In (b), no B^1_{2g} peak is shown at 308 cm^{-1}, indicating this to be a monolayer WSe_2.

Circularly Polarized Extinction Measurement

Figure S2 displays the extinction measurement setup to characterize the localized surface plasmon resonance (LSPR) of the chiral metasurface. Nonpolarized white light from a tungsten–halogen source was backside-focused on the devices by a 20× objective lens, and the output signal was collected by a 10× objective lens and then sent to the spectrometer by a fiber. To further obtain the polarized properties of the chiral metasurface, a broad-band quarter-wave plate (QWP) was included in the setup, followed by a linear polarizer (LP) on the light path before the fiber to analyze the polarization information.

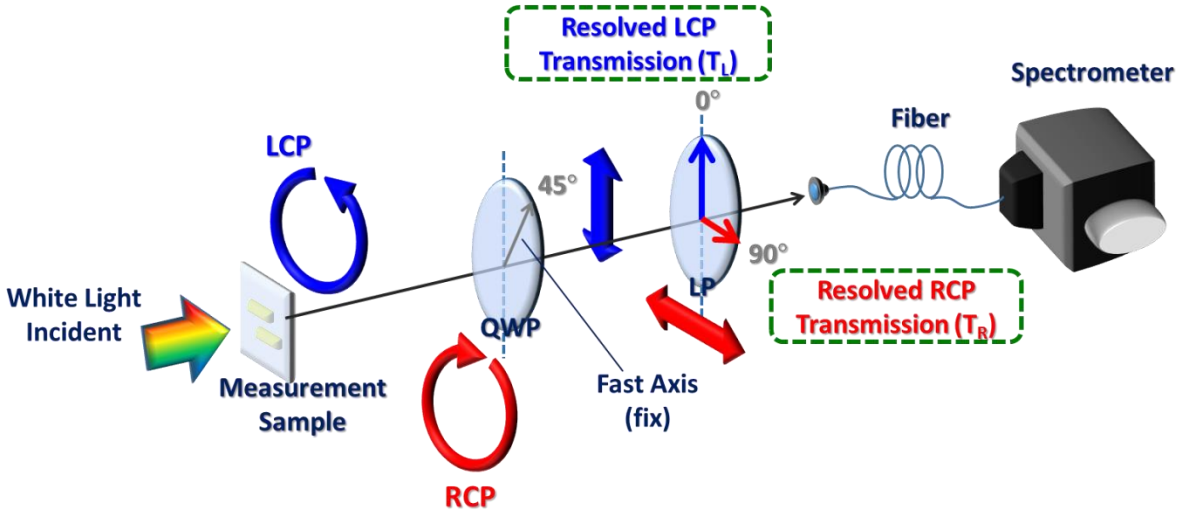

Figure S2. Schematic of the extinction measurement system.

Micro-Photoluminescence Measurement System with Cryogenic System

Figure S3 presents a schematic of the micro-photoluminescence (micro-PL) system used to analyze the circular dichroism (CD) of the PL from the monolayer WSe_2. To access the valley selective absorption, the devices were loaded in a Janis ST-500 cryostat and the operating temperature was lowered to 77 K by flowing liquid nitrogen. A 532-nm continuous wave (CW) laser, which served as the excitation source, was modulated as a left-hand/right-hand circular polarizer (LCP/RCP) by a QWP (to convert linear polarization to circular polarization) and a half-wave plate (to switch between LCP and RCP). The circularly polarized excitation was focused on the devices at normal incidence through a 100× objective lens, and the output PL was collected by the same objective lens. The output PL was then imported to the monochromator and detected by the Andor iDus 416 CCD. To obtain the CD of the output PL, a broad-band QWP and an LP were applied to analyze the LCP and RCP components [*I(R)* and *I(L)*] of the output PL. The CD value (CD_{PL}) could then be calculated as

$$CD_{PL} = \frac{I(R) - I(L)}{I(R) + I(L)} \times 100\%$$

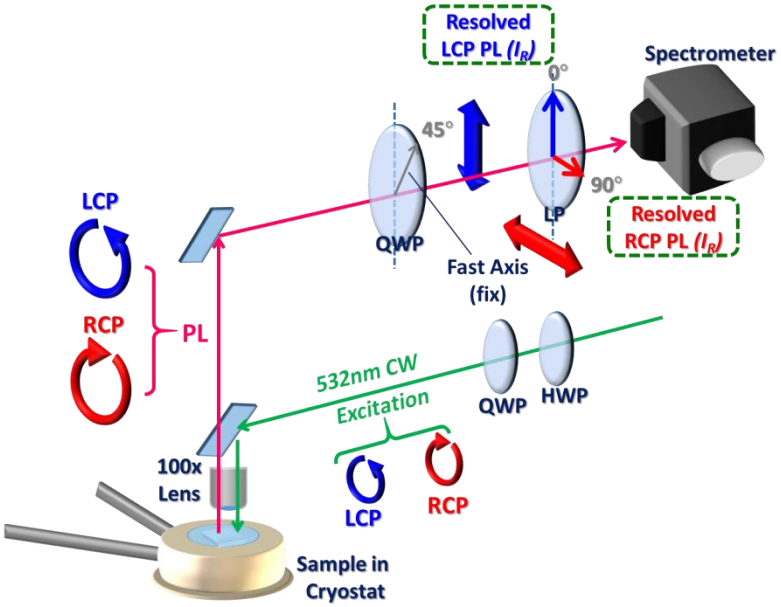

Figure S3. Schematic of the micro-PL system.

Manipulation of Circular Polarization of Chiral Metamolecules

To manipulate the circular polarization of the emission from the monolayer WSe_2 with the chiral metasurface, a series of shift dimer nanorods (SDNRs) with different lengths were investigated. The gold nanorod length *L* varied from 120 nm to 210 nm, and the other geometric parameters were fixed. Because the LSPR mode in the SDNRs was evolved from the long axis mode in each nanorod, SDNRs with different lengths exhibited their LSPR at different wavelengths.

Following the aforementioned characterization method, the CD_T spectra of these SDNRs were obtained (Figure S4). The CD_T peaks and dips red shifted from approximately 680 nm to over 900 nm when *L* increased. In other words, the LSPR modes of these SDNRs could provide different spectral mode matching with the PL from the monolayer WSe_2. We focused the CD_T values around 721 nm, which is consistent with the PL peak wavelength of a bare monolayer WSe_2 at 77 K (Figure S4). These SDNRs covered a range in CD variation from $\pm 5\%$ to 13%, providing a solid platform for manipulating the circular dichroism of the PL from the monolayer WSe_2.

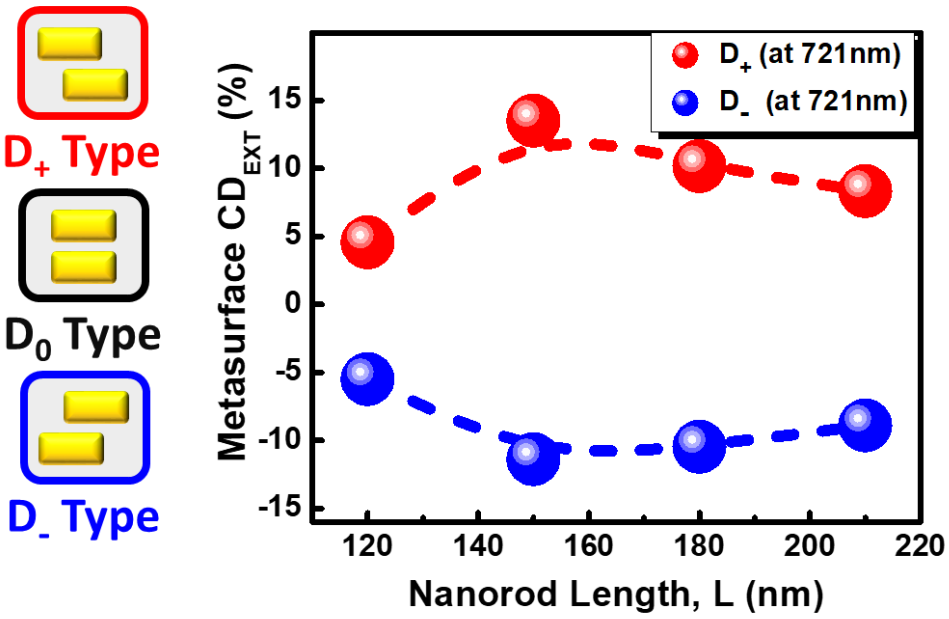

Figure S4. CD_T value of each D+ and D− type SDNR when wavelength = 721 nm. The insets represent CD_T spectra of the D0, D+, and D− type SDNRs for L = 150 nm and L = 210 nm. The other geometric parameters of the nanorods were fixed: width = 80 nm, height = 50 nm, gap between two nanorods = 40 nm.

The Photoluminescence (PL) Spectra under Different Temperatures

To characterize photoluminescence (PL), a 532-nm continuous wave (CW) laser was applied as the excitation source. Figure S5 show the PL spectra from the monolayer WSe_2 was recorded in a Janis ST-500 cryostat and the operating temperatures are 300K (red) and 77 K (black). The emission from the neutral exciton and the charged exciton are labeled with X^0 and X^-.

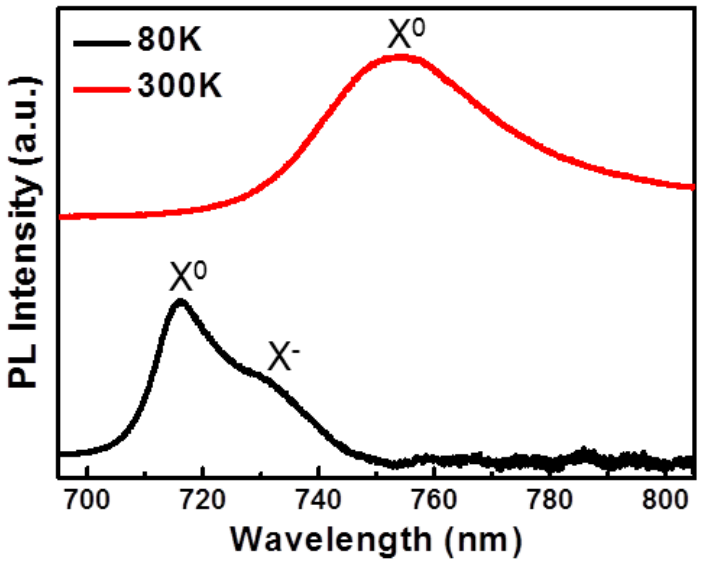

Figure S5. The photoluminescence (PL) spectra from a WSe_2 monolayer at room temperature (red) and 80K (black). The emission from the neutral exciton and the charged exciton are labeled with X^0 and X^-.

Reference

(1) Huang, J.-K.; Pu, J.; Hsu, C.-L.; Chiu, M.-H.; Juang, Z.-Y.; Chang, Y.-H.; Chang, W.-H.; Iwasa, Y.; Takenobu, T.; Li, L.-J. Large-Area Synthesis of Highly Crystalline WSe_2 Monolayers and Device Applications. *ACS Nano* **2014**, *8* (1), 923–930.

(2) Li, H.; Lu, G.; Wang, Y.; Yin, Z.; Cong, C.; He, Q.; Wang, L.; Ding, F.; Yu, T.; Zhang, H. Mechanical Exfoliation and Characterization of Single- and Few-Layer Nanosheets of WSe_2, TaS_2, and $TaSe_2$. *Small* **2013**, *9* (11), 1974–1981.