Monolayer transition metal dichalcogenides (TMDCs) have intrinsic spin-valley degrees of freedom, making it appealing to exploit valleytronic and optoelectronic applications at the nanoscale. Here, we demonstrate that a chiral plasmonic antenna consisting of two stacked gold nanorods can modulate strongly valley-polarized photoluminescence (PL) of monolayer MoS$_2$ in a broad spectral range at room temperature. The valley-polarized PL of the MoS$_2$ using the antenna can reach up to $\sim$47%, with approximately three orders of PL magnitude enhancement within the plasmonic nanogap. Besides, the K and K$'$ valleys under opposite circularly polarized light excitation exhibit different emission intensities and directivities in the far field, which can be attributed to the modulation of the valley-dependent excitons by the chiral antenna in both the excitation and emission processes. The distinct features of the ultracompact hybrid suggest potential applications for valleytronic and photonic devices, chiral quantum optics, and high-sensitivity detection.

INTRODUCTION

Broken inversion symmetry and strong spin-orbit interaction in monolayer transition metal dichalcogenides (TMDCs) lead to spin-valley locking at K and K$'$ valleys (1–3). Energetically degenerate excitons in different valleys can be optically addressed and detected using different circularly polarized light because of their valley-dependent optical selection rule (4–6). As a result, the spin-valley effect lays the foundation for the exploration of valleytronics (7–9). Metallic nanostructures [e.g., nanowires (10), gratings (11), and metasurfaces (12–16), etc.] have been used to improve the light-matter interaction and steer valley-polarized emission of TMDCs. These nanostructures can spatially separate the emission of valley excitons through the near-field interference of circularly polarized dipoles, resulting in asymmetric emission. These waveguide structures provide a feasible way to design valleytronic devices by combining TMDCs with nanophotonic structure.

Nevertheless, previous metallic nanostructure–based surface plasmon polaritons used to control valley emission have had large footprints on the microscale. It is well known that localized surface plasmon resonance (LSPR) can concentrate the electromagnetic field considerably in a tiny volume to improve the light-matter interaction on the nanoscale. Nano-objects such as metallic core-shell, V-shaped, and Yagi-Uda antennas have been shown to control the emission directivity of nanoemitters (17–20); however, these antennas have seldom been applied to valleytronic devices. Chiral light-matter interactions in valleytronic applications need a chiral structure to generate chiral electromagnetic near fields (21, 22). Chiral nanostructures have been widely applied in the study of biomolecules through the analysis of circular dichroism spectra, providing valuable information with high sensitivity (23–25). Recently, chiral MoS$_2$-metasurface heterostructures have been used to tailor the valley-polarized photoluminescence (PL) of monolayer MoS$_2$ (12). However, the chiral performance was only successfully achieved at low temperatures down to 87 K because of the low coupling efficiency between the metasurfaces and valley excitons in MoS$_2$, limiting its application at room temperature.

In this study, we constructed a stereoscopic antenna consisting of two gold nanorods (GNRs) with a small footprint ($\sim$0.02 $\mu$m$^2$) and an ultrasmall “hot-spot” volume. We experimentally assembled two GNRs in a corner-stacked configuration to form the stereoscopic antenna via atomic force microscopy (AFM) manipulation. The monolayer MoS$_2$ was sandwiched inside the nanogap to greatly enhance the interaction between the LSPRs and excitons. Compared with previous works, this stereoscopic antenna having chiral properties can more efficiently tailor valley-polarized emission of monolayer MoS$_2$, reaching up to a degree of valley polarization (DVP) of $\sim$47% at room temperature (Fig. 1A). We observed that the PL valley polarization of the MoS$_2$ was dependent on the configurations of the antennas. In addition, the chiral antenna can modulate the emission directions of the MoS$_2$ over a broad spectral range, and even unidirectional emission could occur. The PL emission of valley excitons under different polarized excitations can be separated spatially. Combining the finite-difference time-domain (FDTD) method and the dipole-dipole model, we revealed that the far-field interference between the radiation fields from the MoS$_2$ and the out-of-phase dipoles induced in the GNRs gave rise to this unidirectional emission effect. The valley-polarized modulation originates from the near-field interaction between excitons of the monolayer MoS$_2$ and the chiral field of the antenna. The calculated results, such as the electromagnetic distribution field, antenna quantum efficiency, and optical chirality, supported the experimental observations.

RESULTS

The valley-dependent selection rule can explain the chiral optical valley selectivity of the PL of MoS$_2$. In a MoS$_2$ monolayer, the coupling of right-/left-handed circularly polarized (RCP/LCP) light with the K/K$'$ valley to generate valley excitons leads to opposite-handed
circularly polarized emission. Because of phonon-assisted inter-valley scattering, excitons can partly scatter into the opposite valley. In the present experiment, the pristine monolayer MoS$_2$ without antenna exhibited 18% DVP at room temperature. For symmetric metal structures such as a single GNR, the near-field enhancement is chirality independent. The excitons of opposite valleys couple equally to the local field of such achiral plasmonic structures that cannot modify the DVP. To obtain a chiral antenna, we had to break the symmetry of the nanostructures. The MoS$_2$-antenna hybrid was assembled using the AFM manipulation method. The top-right illustration in Fig. 1B presents a schematic of the chiral stereoscopic plasmonic antenna in the present experiment, which consists of two GNRs with corner-stacked configuration and a MoS$_2$ monolayer sandwiched in the nanogap. First, the GNRs were sparsely deposited on a glass coverslip with separations of several micrometers, as confirmed by AFM and optical images. Then, the chemical vapor deposition (CVD)–grown MoS$_2$ monolayer was transferred onto the coverslip. Last, with the aid of AFM manipulations, we assembled the chiral antenna by moving a GNR from the bare glass onto a GNR covered by monolayer MoS$_2$. We then adjusted the relative position and intersection angle between the two stacked GNRs, which determined the chirality and optical response of the antenna (26–28). During the manipulation, we measured their optical properties in situ, which was convenient for studying the optical response of the hybrids with different configurations. Representative images of the AFM manipulation during the assembly process are presented in fig. S1A. We defined this structure as a left-handed (LH) antenna. Figure 1C presents the PL spectra for typical MoS$_2$ monolayer (dots) and an LH antenna–MoS$_2$ hybrid (lines) under circularly polarized excitation. a.u., arbitrary units. (D) Measured far-field PL patterns and corresponding normalized angular radiation distributions in polar coordinates for the bare MoS$_2$ monolayer and the LH antenna–MoS$_2$ hybrid.
opposite side in the BFP patterns: the ratio between the maximum emitted power on a side and its cross-sectional area of the antenna and the area of the laser spot, respectively. We introduce the directivity, D

The ratio between the maximum emitted power on a side and its cross-sectional area of the antenna and the area of the laser spot, respectively.

The modulation of chiral emission occurred mainly within the near field around the antenna, where excitons interacted strongly with plasmon modes. To quantify the unidirectional emission induced by the hybrid, we accounted for the background emission and subtracted the raw data with the background signal using a ratio of 1 – S0/S1, where S1 and S0 are the extinction cross-sectional area of the antenna and the area of the laser spot, respectively. S0 was estimated to be 11.4% in the present experiment (details in section S1 and fig. S3). We introduce the directivity, D, as the ratio between the maximum emitted power on a side and its opposite side in the BFP patterns: D = 10 * Lo / Plo. The directivity values S1 and S2 were obtained by integrating the PL intensities over two different areas shown in the first image of Fig. 2C (dashed lines) after background correction. From the quantitative values in fig. S4, we observed that the directional emission performance changed with opposite circular polarization excitation and the intersection angle. Moreover, the PL patterns of the hybrid structure under different linear polarized excitation (fig. S5) also changed because of the structural chirality of the antenna.

In addition to the experiments, we calculated the far-field emission patterns by using the FDTD method (29) and the near-field–to–far-field (NFTFF) transformation method. The emission from the MoS2 monolayer originates solely from the in-plane excitons at K and K’ valleys (30). Thus, the valley excitation can be treated as dipole pairs at a wavelength of 675 nm (A exciton of MoS2) polarized perpendicular to each other in the X-Y plane with a phase difference of ±90° for σ+/− polarized emission. As shown in Fig. 3A, we placed a dipole pair in the hot spot for simplicity. We determined the physical size of the antenna shown in Fig. 3A from AFM measurements and FDTD simulations (as discussed in section S1). The calculated angular emission patterns for different configurations are presented in Fig. 2 (C and D). The difference in the far-field angular distribution under oppositely circular-polarized excitation results from the different chiral near-field response in the nanogap. We note that there are some deviations between the experimental and simulated results, which can be attributed to the deviations between the simulation and experiment, such as the exciton distribution and quantity and the antenna configuration resulting from different separation and orientation of the GNRs. For example, the top GNR was oblique in the experiment (as discussed in fig. S1), whereas the GNR was set as horizontal in the simulations.

To understand the unidirectional emission, we used the FDTD method and the dipole-dipole model (31) to simulate the processes. Such unidirectional emission can be explained as the interference of the direct radiation from the monolayer MoS2 with the radiation from the induced dipolar mode of the antenna. The dipole moment can be written as Pj = Pjx+Pjy, where j = 1 and 2 refer to the two dipoles, and x and y refer to the component in the X and Y directions. Thus, the constructive or destructive interference depends on the relative amplitude (Pj/Pk) and phase difference (∆ø ± kl) of the two dipole moments, where k is the wavelength vector and l is the dipole-dipole separation distance. Figure 3B presents a schematic illustration of the model for the analysis of directional emission. In this case, the constructive or destructive interference in the X/Y component is the bright or dark region, respectively, in the angular distribution in the X or Y direction. For destructive interference, the two conditions, i.e., Pj/Pk = 1 and |∆ø ± kl| = π, should be satisfied in the same direction, resulting in unidirectional emission (the calculations are described in detail in section S2). For the antenna in Fig. 3A, the surface charge-density distributions around two GNRs were transferred to the plane of the dipole pair by adding a phase of kl1 and kl2, where d1 = 45 nm and d2 = 32.5 nm. The surface charge-density distribution of the σ– dipole pair is shown in Fig. 3C. The calculated results indicate that the total phase difference in the Y direction is ∆ø = kl1 = 1.026π and P1/P2 = l = 1.27, which suppresses the radiation in the −Y direction (resulting in unidirectional emission D of ~8.4 dB). In the X direction, the parameters were far from the destructive interference conditions, with equal radiation in both the ±X directions (~1.6 dB). For comparison, the surface charge-density distribution and calculated results with a σ+ dipole pair in the same plane in Fig. 3 (E and F) explain
the directional emission pattern. These results illustrate the origin of the directional emission under opposite polarization excitation.

Furthermore, we compared the valley-polarized PL spectra of the pristine MoS$_2$ and MoS$_2$-antenna hybrid. The representative polarized PL spectra of the pristine MoS$_2$ show similar behaviors for the LCP and RCP excitations in fig. S6. The obtained DVP (~18%) is consistent with that in previous works, with typical values of DVP of 15 to 25% at room temperature (32). This result was expected under the valley-dependent optical selection rule. As discussed above, the excitons directly excited by the laser contribute to the absolute value of DVP of the hybrid reached up to ~47% under $\sigma^-$ excitation but decreased to 11% under $\sigma^+$ excitation near the A exciton of MoS$_2$ (675 nm). Compared with the pristine MoS$_2$, that with the antenna resulted in brighter emission with $\sigma^-$ polarization excitation and a higher proportion of $\sigma^-$ PL emission. For comparison, the valley-polarized PL spectra of the MoS$_2$ coupled with a single GNR are presented in fig. S2. The absolute values of DVP with $\sigma^+$/-$\sigma^-$ polarized excitation are similar (DVP, ~21%) because of the chirality-independent localized field of a single GNR. As mentioned above, the relative position of the intersection between the stacked GNR determines the chirality and optical response of the antenna. We constructed another representative antenna with opposite chirality [right-handed (RH)] as that of the LH antenna discussed above, as shown in fig. S7. The conclusions drawn from the LH antenna and associated mechanisms were equivalent to those for the RH antenna. This result indicates that we can obtain valleytronic devices with a certain helicity by controlling the configuration using the AFM manipulation method.

![Image](image-url)
DISCUSSION

It is well known that an antenna can increase the localized excitation field to enhance the PL intensity and radiative decay rate of excitons through the Purcell effect (33–35). We measured the PL intensity with the antenna (PL_{with}) and without the antenna (PL_{w/o}), as shown in Fig. 1C. Taking S_0 as the hot-spot averaged area for effective enhancement, the PL enhancement factor f_{PEF} was calculated using f_{PEF} \sim \frac{S_0}{S_0 \text{PL}_{with}} \text{PL}_{w/o} - 1 \) (details in section S1), where S_0 is the focus area. We obtained f_{PEF} of ~300 for σ+ polarized excitation and ~950 for σ− polarized excitation at 675 nm. In the simulations, the relative PL enhancement factor was estimated using g_{ex} = g_{em} = |E/E_0|^2 \times |\eta/\eta_0|, where g_{ex} = |E/E_0|^2 is the in-plane excitation field enhancement at an excitation wavelength of 633 nm and g_{em} = |\eta/\eta_0| is the enhancement of the quantum yield at an emission wavelength of 675 nm. Figure 4C shows the relative intensity of the in-plane electromagnetic field at the excitation wavelength with opposite polarization excitation at Z = 45 nm, where the equivalent MoS_2 layer was placed. E_{XY} showed higher enhancement and a broader distribution with σ− polarized excitation than with σ+ polarized excitation. Then, we placed 16 dipole pairs with a phase difference of ±90° in the nanogap with random phases and positions for chiral emission simulation. \( \eta_0(0) \) is related to \( \eta_0(0) \) (the original quantum efficiency of MoS_2), \( \eta_0(0) \) (the antenna quantum efficiency), and \( F(0) \) (the radiative decay rate enhancement, i.e., the Purcell factor) according to \( \eta_0(0) = \eta_0(0) / \left[ 1 - \frac{\eta_0(0)}{F(0)} \right] + \frac{\eta_0(0)}{\eta_0(0)} \), \( \eta_0(0) \) is the ratio of \( P_{rad} \) (the energy that reaches the far field) to \( P_{tot} \) (the total power dissipated by the emitter), and \( F \) is the ratio of \( P_{rad} \) to \( P_{rad}^0 \) (the energy that reaches the far field without the antenna). The corresponding \( \eta_0 \) as a function of wavelength is shown in Fig. 4E, revealing a higher quantum yield efficiency with σ− dipole pairs at a wavelength of 675 nm than that with σ+ dipole pairs. The assumed quantum efficiency of monolayer MoS_2 (\( \eta_0(0) \)) at \( \lambda = 675 \text{ nm} \) was ~1% in our simulation (36). After calculation, the antenna has a larger enhancement factor under σ− polarized excitation (~1620) than under σ+ polarized excitation (~630), which is roughly consistent with the experimental results.

The valley helicity is strongly affected by the competition between the intervalley scattering rate (Γ_{vl}) and the recombination rate (Γ_0) of e-h pair (3). The valley polarization of pure MoS_2 is depolarized mainly by intervalley scattering between K and K’ valley (DVP \propto \frac{1}{1 + 2 \Gamma_{vl}/\Gamma_0}). Generally, a faster radiative decay rate will lead to a higher polarization helicity. The exciton transition from excited states to ground states can be affected strongly by the antenna through the Purcell effect (37), which can contribute to the increasing of valley polarization. The chiral plasmonic antenna modulates the excitons in the K or K’ valley with different decay rates and antenna efficiencies. For example, as shown in Fig. 4D, for an LH antenna, the antenna quantum efficiency for σ− dipoles (\( \eta_0^- \sim 0.36 \)) is higher than that for σ+ dipoles (\( \eta_0^+ \sim 0.27 \)), leading to a higher quantum yield efficiency for excitons in the K’ valley than those in the K valley. To quantify the contributions of the Purcell effect, we modeled the behaviors of valley excitons within the hot spot with steady-state rate equations (details in section S3). For the exciton transition, the modulated DVP can be written as DVP = \frac{\eta_0^- n_+ - \eta_0^+ n_-}{\eta_0^- n_+ + \eta_0^+ n_-} \approx \frac{P_{Gamma}^- - P_{Gamma}^+}{P_{Gamma}^- + P_{Gamma}^+}, where \( P_z \) and \( \Gamma_{at} \) are the pumping rates and the radiative decay rates modulated by the antenna. Here, \( \Gamma_{at} \gg \Gamma_{vl} \gg \Gamma_0 \), and we assumed that \( \Gamma_{vl} \) and \( \Gamma_0 \) are not influenced by the antenna since they dominantly depend on the intrinsic properties of MoS_2, ambient temperature, excitation energy, etc. We found that the antenna-assisted valley transition increases the DVP greatly (more than 90%) through the Purcell effect after calculating (details in section S3). For the LH antenna, the DVP value (\( \rho_{PL} \)) by σ− excitation is higher than that (\( \rho_{PL} \)) under σ+ excitation owing to the chiral efficiency effect.

However, the antenna can also strongly modify the polarization states of the light, which depolarized the DVP of the detected far-field PL signal. We can divide this depolarization effect into two processes, i.e., the excitation and emission process involving the conversions between the near field and the far field. First, the excitons generate both in the K and K’ valley even under pure circularly polarized excitation. The generation rates in the K/K’ valley are governed by the \( E_k \) and \( E_l \) components in the near field within the hot spot, which can be obtained numerically through \( E_k = \frac{1}{2}(E_x + i E_y) \) and \( E_l = \frac{1}{2}(E_x - i E_y) \). Under excitation using σ− polarized light, more excitons generate in the K’ valley rather than K valley because of the stronger \( E_k \) than \( E_l \) for the LH antenna (shown in Fig. 4E). For comparison, schematic illustrations of the \( E_k \) and \( E_l \) distributions and the valley-polarized modulation under σ+ excitation are shown in fig. S8. Second, after the exciton transitions, the antenna would scatter the near-field light to the far field as the detected PL signal. This scattering process also changes the DVP (discussed in section S3). Considering all these factors, we obtained the final DVP_{σ−} \approx 50% and DVP_{σ+} \approx 10% for the LH antenna under σ− or σ+ polarized light excitations, respectively. The numerical results are in reasonable agreement with the experiment result (\( \rho_{PL} = 47% \) and \( \rho_{PL} = 11% \)). The deviations could be due to differences between the simulation and experiment such as the distribution and quantity of excitons or the antenna configuration such as different separation and orientation of the GNRs.

In addition, we calculated the optical chirality C as \( C(γ) = \frac{1}{2} \text{Im} [\tilde{E}(γ) \times \tilde{B}(γ)] \) in the nanogap, which has been applied to describe the enhancement performance of circular dichroism signals detected from chiral chemical and biological molecules (23–25). \( \tilde{E}, \tilde{B}, \eta_0 \), and \( Ω \) in this equation are the magnetic fields, the permittivity of vacuum, and angular frequency. As shown in fig. S10, \( C(C_0) \) of the chiral field is as large as 18 under σ− excitation, where \( C_0 \) is the value of optical chirality for a σ− polarized plane wave in a vacuum. \( C(C_0) \) has the opposite distribution under σ+ excitation with a maximum value of 8, indicating the intrinsic chirality for this antenna. In the experiment, the antenna can steer the valley-polarized PL of monolayer MoS_2 in a broader spectral range, with a DVP of 34 to 54% at wavelengths ranging from 640 to 800 nm. We believe that the nanogap volume would result in a strong coupling strength, which allows the chiral plasmonic effect to extend to the nonresonant range. Hence, tuning of the LSPR mode of the antenna to the exciton energy of MoS_2 was not necessary, which is beneficial for the nanofabrication of valleytronic devices based on the plasmon-TMDC hybrid system.

CONCLUSION

In summary, we demonstrated that an ultracompact chiral antenna formed by corner-stacked GNRs can efficiently tailor valley-polarized PL of monolayer MoS_2 at room temperature. With the aid of AFM, we could manipulate such plasmonic antennas with different configurations with the monolayer MoS_2 sandwiched in the nanogap. The nanogap (~5 nm) between the GNRs made it possible to steer...
In our experiments, the MoS$_2$ monolayer was prepared using the CVD method (32). The GNRs with a size of approximately 70 nm by 160 nm were synthesized using a seed-mediated wet chemical method (38). After the GNRs were sparsely deposited on a glass coverslip, the MoS$_2$ monolayer was transferred onto the glass coverslip using polydimethylsiloxane (39).

**Experimental setup**

We combined an inverted Olympus optical microscope with an atomic force microscope on the top side to scan and manipulate the GNRs. The CVD-grown MoS$_2$ monolayer boundary can be distinguished by the gray contrast of the AFM image in fig. S1, as indicated by the dotted line. To study the valley polarization PL of the MoS$_2$ monolayer, we used polarized excitation and circularly polarized PL measurements. A continuous wave laser at 633 nm was used to excite the samples passing through an oil immersion objective lens (1.4 numerical aperture, 60×; Olympus). We set a quarter-wave plate (633 nm) after the laser irradiation to obtain circularly polarized excitation. To resolve and measure the circularly polarized PL spectra, we placed a broadband quarter-wave plate (400 to 800 nm) and a Glan lens before the spectrometer. As shown in Fig. 1C, one characteristic PL peak of MoS$_2$ monolayer was observed in the spectrum, which was the direct bandgap transition (A exciton, 675 nm). Moreover, the far-field PL emission patterns were measured using the BFP imaging method. In addition, the scattering spectra of the same nanostructures were obtained in situ using the white-light total internal reflection dark-field method (40). All the measurements were performed at room temperature.

**FDTD simulations**

Three-dimensional (3D) FDTD methods were used to simulate the emission flux, emission patterns, and electric-field distributions of the assembled hybrid nanoantenna. The calculations of the far-field radiation pattern were based on the NFTFF transformation method. Although the NFTFF method is derived for a closed surface, here, we chose a large transformation plane (2 μm by 2 μm) placed 50 nm beneath the air/glass interface to collect most of the flux directed to the substrate to achieve a better approximation. In this 3D simulation, the mesh size is 0.5 nm to match the memory resources and computation time. The optical dielectric function of gold was modeled using the Drude-Lorentz dispersion model (29). The optical dielectric function of the equivalent MoS$_2$ layer was modeled using the experimental data in the references. In addition, the refractive indices of the media were set to 1.0 for air and 1.49 for silica (glass substrate). To form a chiral excitation, we used two plane waves polarized perpendicular to each other in the X-Y plane with a phase difference of ±90° for σ+/− polarized light. The chiral emission of monolayer MoS$_2$ was regarded as dipole pairs in the X-Y plane. Each dipole pair consisted of two orthogonal electrical dipoles. The phase difference of ±90° represented a σ+/− polarized exciton.

**Supplementary Materials**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/21/eaao0019/DC1

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