NANOPHOTONICS

Nanoscale chiral valley-photon interface through optical spin-orbit coupling
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The emergence of two-dimensional transition metal dichalcogenide materials has sparked intense activity in valleytronics, as their valley information can be encoded and detected with the spin angular momentum of light. We demonstrate the valley-dependent directional coupling of light using a plasmonic nanowire—tungsten disulfide (WS2) layers system. We show that the valley pseudospin in WS2 couples to transverse optical spin of the same handedness with a directional coupling efficiency of 90 ± 1%. Our results provide a platform for controlling, detecting, and processing valley and spin information with precise optical control at the nanoscale.

Valleytronics and nanophotonics provide powerful routes to address the heating problem in electronics by offering an alternative to information transport with the charge of electrons (1, 2). Valley pseudospin provides an additional degree of freedom to encode and process binary information in matter, analogous to the spin degree of freedom. The emergence of two-dimensional transition metal dichalcogenide (TMD) layers provides a versatile materials platform for both optoelectronics (3) and valleytronics (4–9). These materials have direct band gaps consisting of two (energy-degenerate) valleys at the corners of the Brillouin zone (labeled K and K′). The spin and valley information in TMD materials can be optically addressed and detected by using the spin angular momentum of light because of their valley-dependent optical selection rule (7, 8, 10–12). However, the relatively short lifetimes (<10 ps) of valley-polarized excitons limit local processes and the spatial transport of valley information (2). Although valley information is difficult to transport in realistic material systems, particularly at room temperature, photons are ideal information carriers owing to the normally small light–matter interaction. Nanoscale coupling of valley pseudospin to photonic degrees of freedom is desirable for on-chip integrated valley devices.

The spin angular momentum as a degree of freedom of light at the nanoscale offers the ability to influence and exploit light–matter interactions. In highly confined light fields, transverse optical spin angular momentum (t-OSAM) results from the spin-orbit interaction of light (13–25). This t-OSAM provides a robust one-to-one relation between the handedness of optical spin and the propagation direction of a photonic mode—so-called spin-momentum locking—because of their time reversal symmetry. The information of spin angular momentum can be directly transferred to the direction of light and vice versa. Recently, t-OSAM—dependent directional coupling of light with near-unity efficiency has been successfully demonstrated experimentally by using various photonic structures, e.g., optical fibers (13–15), metal surfaces (16–18), metasurfaces (26), semiconductor waveguides (20, 21), and microdisks (22).

We demonstrate a room-temperature chiral-coupling interface between the transverse optical spin of a plasmonic nanowire mode and the valley pseudospin of tungsten disulfide (WS2), which provides robust valley-polarized directional emission. The resulting coupling between the photonic path and valley-spin in TMD materials and the metallic nature of the waveguide should enable applications in valley-chiral networks, valley-gates, and quantum photonic devices.

Our configuration for a chiral valley-photon interface (Fig. 1A) consists of a combination of a few-layer TMD material and a single silver nanowire. The evanescent fields of the plasmonic guided modes possess t-OSAM perpendicular to the TMD layers. The population of excitons in the two different valleys K and K′ can be directly controlled by a circularly polarized excitation laser. The radiative decay of a valley-polarized exciton is associated with a circular transition dipole, which emits valley-dependent circularly polarized light. This circular transition dipole couples to the plasmonic eigenstate with the same handedness of its local transverse optical spin. As a result of this chiral coupling between the circular nature of the emission dipole of the valley-polarized exciton and the local direction-locked transverse optical spin, emission from the different valleys couples to plasmonic modes propagating in opposite directions (Fig. 1B) when the transverse optical spin is maximal. To obtain a high degree of valley polarization, we exploit the high spin-valley coupling strength and spin-layer locking effect of WS2 (27, 28). The degree of valley polarization is denoted by PR = (IK − IK′)/IK + IK′, where IK and IK′ represent the photoluminescence (PL) intensity from K and K′, respectively. The number of WS2 layers is identified by the optical contrast of the layers in a microscope image and their PL spectrum (fig. S1). The results presented here are based on a WS2 flake consisting of five layers. Its polarization-resolved spectrum, measured in the absence of the silver nanowire (Fig. 1C), reveals a measured PR = 0.7 at room temperature. Figure 1D depicts an optical microscopy image of one of the samples and a collected PL image dominated by emission from the indirect band gap. The two bright spots at each end of the waveguide demonstrate that emission is coupled to the guided modes of the plasmonic nanowire. Through the chiral coupling, the valley information is converted to the propagation direction of the mode to which the exciton coupled: The plasmonic mode itself has no overall optical spin (fig. S2). Under local excitation at the middle of the silver nanowire, directional emission from the TMD layers is investigated by measuring light scattered at the ends of the wire.

To quantify the magnitude of the t-OSAM near a silver nanowire, we performed numerical calculations with a finite-difference eigenmode solver (Fig. 2A). The guided plasmonic mode exhibits strong evanescent fields at the glass-nanowire interface, i.e., exactly at the position of WS2 layers. Due to the strong transverse confinement and the plasmonic nature of the modes, a large longitudinal (x direction) component of the electric field is present. The x and y components of the electric field have comparable amplitudes and are roughly ±90° out of phase: The modes exhibit a large t-OSAM. The local sign of the local t-OSAM has a one-to-one relation with the propagation direction of light and the position with respect to the mirror plane of the geometry (y direction). We calculated the density of the t-OSAM as a function of position in the x–y plane, which corresponds to the Stokes parameters S = −2Re(ExEy)/(|Ex|2 + |Ey|2) (29) (Fig. 2B). The plasmonic guided modes of infinite-length nanowires locally have a t-OSAM density near unity. The sign of the transverse optical spin on either side of the nanowire is opposite, as expected from symmetry considerations. Clearly, when the propagation direction of the mode is reversed, so are all the signs of the helicity everywhere. These results are qualitatively summarized in Fig. 2C, demonstrating that the handedness of optical spin is determined by a combination of y position and propagation direction.

We simulate the emission from one valley in WS2 near a finite-length plasmonic nanowire.
Fig. 1. Valley-controlled directional coupling of light. (A) A conceptual illustration of directional emission of a valley-polarized exciton in WS2. The valley pseudospin and photon path are coupled by means of spin-orbit coupling of light. (B) A schematic sketch of the band diagram of WS2 and its optical selection rules, which depend on the valley index. The two opposite-handed circularly polarized emissions from each valley would couple preferentially to modes propagating in opposite directions. LHC, left-handed circular; RHC, right-handed circular. (C) Polarization-resolved emission spectrum of WS2, measured without a silver nanowire, at room temperature excited by a left-handed circularly polarized laser beam at 594 nm. Red and blue spectra indicate left- and right-handed circularly polarized emission, respectively, a.u., arbitrary units. (D) Image of a fabricated WS2–silver nanowire coupled system (top). WS2 emission coupled to the plasmonic waveguide mode and scattered at the ends of the nanowire (bottom). Scale bar, 5 μm.

Fig. 2. Numerical modeling of the transverse spin angular momentum of light. (A) Cross-sectional electric field intensity (|E|^2) distribution of the plasmonic guided mode. (B) Density of transverse optical spin, i.e., Stokes parameter S_3, of the in-plane (x-y plane) electric field component. (C) Illustration of the distribution of handedness of elliptical polarization, optical spin, which is dependent on both the position and propagation direction. (D) Directionality of circularly polarized dipole emission, D_0(x,y), as a function of its position. (E) Normalized coupling strength of circularly polarized dipole source to the plasmonic guided modes, K_{tot}(x,y). (F) Chiral-coupling coefficient indicating how efficient circular dipole in one valley couples to one direction of the guided mode, K_{valley-path}(x,y) = D_0(x,y)K_{tot}(x,y).
with the three-dimensional finite-difference time-domain method. The polarized valley emission is described with a circular dipole. Figure 2D depicts the position-dependent directionality \(D_0\) of the emission of a left-handed circular dipole in the \(x\)-\(y\) plane. It is calculated from the light transmitted \((T_L(x,y))\) and the right \((T_R(x,y))\) ends of the wire, \(D_0(x,y) = (T_L(x,y) - T_R(x,y))/\left[ T_L(x,y) + T_R(x,y) \right]\). First, note that the circular dipole emission is preferentially coupled to a propagating mode with an appropriate sign of optical spin (compare Fig. 2, B and C). Second, note that a high degree of directionality is obtained, which is opposite in sign when exciting a circular dipole on either side of the nanowire. A standing wave-like pattern is visible along the wire, which is caused by a small reflection at the end of the finite-length nanowire (7-\(\mu m\)). Note that the high value of directionality of 0.91 is still observed despite the presence of the low-amplitude counterpropagating mode that tends to reduce the I-OSAM. The length of the silver nanowire is ~7 \(\mu m\). (C) Measured directional coupling efficiency, \((I_L - I_R)/(I_L + I_R)\), of the guided emission as a function of the position of the excitation laser with the (C) left- and (D) right-handed circular polarization. Gray lines represent fitting results obtained by using the calculated directional coupling efficiency. Purple dotted lines correspond to the total PL intensity measured from the ends of the nanowire as a function of the excitation position.

[Fig. 3. Experimental demonstration of valley-controlled directional emission. (A and B) Fluorescence images of the emission of valley-polarized excitons (log scale: top) and line cuts of the intensity profiles (linear scale: bottom) along the silver nanowire under (A) left- and (B) right-handed circularly polarized excitation. The intensities at the excitation regions were deliberately saturated for clarity of the images. The length of the silver nanowire is ~7 \(\mu m\).]
displacement of the excitation spot with respect to the middle of the nanowire in combination with propagation losses, was subtracted from the measured result (fig. S5). A more quantitative description and comparison with calculations will be presented below. The measurements have been reproduced for a number of plasmonic nanowires and WS₂ flakes with different layer thicknesses (fig. S6). In all cases, a strong directionality was observed.

Measurements performed for different excitation polarizations and emission wavelengths confirm that the directional emission is caused by valley-dependent chiral coupling to the plasmonic modes. The direction in the charge-coupled device camera image of the luminescence perpendicular to the length of the nanowire is dispersed in wavelength by using a grating. This configuration provides position-dependent PL spectra along the wire from which we can determine \( k_{\text{exp}} \) for different wavelengths (fig. 4). Figure 4C demonstrates that an equal mix of \( k \) and \( k' \) excitons, generated by using a linearly polarized excitation light, displays no directionality, whereas for the same nanowire, \( K \) or \( K' \) excitons separately (fig. 4, A and B) display valley-controlled directionality. Note that the maximum \( k_{\text{exp}} \) values in fig. 4, A and B, (−0.17) are lower than the value we observed in Fig. 3 of 0.35. This is caused by the decreased signal-to-noise ratio, resulting from the fact that the wavelength-dispersed image could, by its very nature, not resolve any spatial information along the \( y \) direction (fig. S7). Figure 4, D to F, shows that the indirect band gap has a nondirectionality for both handances of the circularly polarized laser (A and B), whereas the transition of the indirect band gap for \( k_{\text{exp}} \sim 0.7 \) for \( k \) or \( k' \) values in Fig. 4, C and D). With a reduction of directionality as \( k_{\text{exp}} \) is less than maximum in the calculations. This is caused, in part, by obvious experimental limitations, e.g., a finite excitation spot size and background noise: Both decrease the experimental maximum of the directionality, and the finite-excitation spot size also spatially smears out the pattern. However, the reduction in directionality is also caused by the finite \( P_{\kappa} \) of the WS₂ system itself. The valley-to-valley hopping (29) would actually flip the direction in which the plasmonic modes are launched. Therefore, the effective directionality with a finite \( P_{\kappa} \) would be a simple product of \( k_{\text{valley-path}} \) and \( P_{\kappa} \). These experimental matters are readily taken into account to properly compare our realized chiral valley-photon interface to the ideal simulated interface

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k_{\text{fit}}(y) = \left[ k_{\text{valley-path}} \cdot P_{\kappa} \right] \otimes \text{PSF} \quad \frac{1}{\left[ k_{\text{tot}} + 2p \right] \otimes \text{PSF}}
\]

where \( k_{\text{valley-path}} \) is the calculated chiral valley-path coupling coefficient for a perfect circularly polarized dipole, \( P_{\kappa} \) is the measured degree of valley polarization (0.7), and \( \rho \) is an unpolarized background noise relative to the maximum signal that represents a heuristic decrease of chiral coupling. To account for the effect of the finite size of the excitation region, we convolute the calculated signal with the point spread function estimated from the experimental data (figs. S8 and S9). We obtain excellent agreement between fit and data for \( \rho = 0.009 \pm 0.005 \) (gray lines in Fig. 3, C and D). With a reduction of directionality as \( D_{\text{PSF}}(1 + 2p/k_{\text{tot}}) \) the fitting procedure reveals that the experimental valley-to-path coupling efficiency, including background noise, is as high as 0.90 ± 0.01. The quality of the fit yields a number of important conclusions. First, because the experimentally determined point spread function is sufficient to explain the broadening of the directional coupling efficiency, exciton diffusion does not notably affect the experiment. Second, any effect of the plasmonic nanowire on the polarization of the excitation focal spot is negligible. Lastly, and most importantly, the chiral valley-to-path coupling is only limited by the magnitude of the transverse optical spin of the plasmonic nanowire modes. Thus, we have been able to realize a room-temperature interface between the valley pseudospin of WS₂ and the propagation direction of nanowire plasmonic modes with a fidelity as high as 0.9.

Our results show that the efficient coupling of valley-polarized excitons in WS₂ to the transverse optical spin of plasmonic nanowire modes
results in valley-photon direction locking with high fidelity. The high valley-spin coupling strength of WS₂ and high transverse optical spin density in plasmonic modes provide a chiral valley–photon interface at room temperature and without the necessity of external magnetic fields. It is important to note that because of the one-to-one relation between optical path and the local transverse optical spin, the propagation direction of the guided light can also be exploited to transfer the valley degree of freedom to other valley devices on a chip through suitable, localized breaking of mirror symmetry, again exploiting the transverse optical spin of the mode, but this time to excite. Realization of such an interface presents a platform for both fundamental studies and a wide range of exciting applications of chiral photonics and chiral quantum optics.

REFERENCES AND NOTES

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Occupation of different valleys within the band structure of some materials can be used to encode information. That information is typically encoded in terms of the chirality or polarization of emitted photons. Gong *et al.* combined a plasmonic silver nanowire with a flake of the transition metal dichalcogenide WS₂ to form a nanophotonic platform for the transfer of solid-state spin into optical information over mesoscopic distances. The direction of light emission from the nanowire was strongly dependent on the spin-orbit coupling of light and the WS₂ layer. Such a highly efficient interface should prove useful for developing valleytronics into a practical on-chip technology.

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