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Inducing Strong Light–Matter Coupling and Optical Anisotropy in Monolayer MoS$_2$ with High Refractive Index Nanowire

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ABSTRACT: Mixed-dimensional heterostructures combine the merits of materials of different dimensions; therefore, they represent an advantageous scenario for numerous technological advances. Such an approach can be exploited to tune the physical properties of two-dimensional (2D) layered materials to create unprecedented possibilities for anisotropic and high-performance photonic and optoelectronic devices. Here, we report a new strategy to engineer the light–matter interaction and symmetry of monolayer MoS$_2$ by integrating it with one-dimensional (1D) AlGaAs nanowire (NW). Our results show that the photoluminescence (PL) intensity of MoS$_2$ increases strongly in the mixed-dimensional structure because of electromagnetic field confinement in the 1D high refractive index semiconducting NW. Interestingly, the 1D NW breaks the 3-fold rotational symmetry of MoS$_2$, which leads to a strong optical anisotropy of up to ∼60%. Our mixed-dimensional heterostructure-based phototransistors benefit from this and exhibit an improved optoelectronic device performance with marked anisotropic photoresponse behavior. Compared with bare MoS$_2$ devices, our MoS$_2$/NW devices show ∼5 times enhanced detectivity and ∼3 times higher photoresponsivity. Our results of engineering light–matter interaction and symmetry breaking provide a simple route to induce enhanced and anisotropic functionalities in 2D materials.

KEYWORDS: MoS$_2$, AlGaAs, mixed-dimensional heterostructure, electromagnetic field confinement, rotational symmetry breaking, light–matter interactions

INTRODUCTION

Since the exfoliation of graphene, the unique electrical, optical, magnetic, and topological properties of two-dimensional (2D) van der Waals (vdW) materials have attracted significant interest and largely transformed the landscape of fundamental research and technological advances in physics, material sciences, and chemistry.1–4 Remarkably, the dangling-bond-free nature of 2D materials enables them to be integrated with non-2D materials (e.g., 0-, 1-, or 3-dimensional materials) through noncovalent interactions to form emerging mixed-dimensional vdW heterostructures.5–7 These heterostructures can combine the synergistic advantages of different dimensional materials, thus providing a more favorable platform than bare 2D materials for numerous advanced applications ranging from on-chip photodetectors to nanolasers.8

Usually, atomically thin 2D layered materials such as transition metal dichalcogenides (TMDCs) suffer from poor luminescence quantum yield because of defect-mediated nonradiative electron–hole recombination and a very short light–matter interaction length compared with the bulk crystals. These properties result in low-performance 2D TMDC-based optoelectronic devices.14,15 To date, a wide variety of approaches have been reported to enhance the light–matter interaction in 2D materials,16,17 e.g., by incorporating Fabry–Perot optical cavities,18 plasmonic structures,20 and meta-surfaces.21 Among these techniques, plasmonic nanostructures made of noble metals (e.g., Ag and Au NWs) are the most advanced in modulating light–matter interactions in 2D materials. However, the structures and their fabrication processes are typically complex, suffer from metal-induced optical losses, and are incompatible to be integrated with semiconductor fabrication processes.22,23

Semiconductor nanowires (NW) with high refractive indices can provide a solution to avoid these issues and provide new opportunities to functionalize 2D materials. Recently, III–V semiconducting one-dimensional (1D) NWs have emerged as promising candidates for various optoelectronic applications because of their direct band gap, simple and low-cost synthesis, high integration ability, and...
precise control in doping. As a III–V semiconducting material, AlGaAs NW possesses a high-refractive index and generates highly localized and strong optical fields within its 1D geometry. Furthermore, these NWs are suitable to be integrated with nanophotonic elements, optical circuits, or other dimensional materials because of their matured transfer and growth processes. Simple integration of 2D TMDCs with such 1D NWs can readily tailor the excitonic response of TMDCs by enabling strong light–matter interactions. Additionally, the hexagonal lattice structure of a monolayer TMDC

Figure 1. Our 2D MoS2/1D AlGaAs NW mixed-dimensional heterostructure. (a) Schematic of the crystal structure of monolayer 2H-MoS2. The blue color lines and red circle at the center of the triangle represent mirror reflection planes and the 3-fold rotational symmetry axis of the crystal, respectively. (b) Illustration of monolayer MoS2 transferred over a NW. (c) Optical image of a typical MoS2/NW sample. Scale bar: 5 μm. Inset shows an atomic force microscope (AFM) image of the heterostructure measured at the blue dashed rectangular area of the image. (d) Raman spectra of bare MoS2, a AlGaAs NW, and a MoS2/NW heterostructure.

Figure 2. Enhanced and broken-symmetry-induced anisotropic optical response of mixed-dimensional heterostructures. (a) Raman spectra of a bare MoS2 flake and a MoS2/NW heterostructure at room temperature. Inset shows the schematic of the crystal structure with in-plane and out-of-plane Raman modes adjacent to the corresponding peaks. (b) Comparison of room temperature PL spectra of a bare MoS2 flake and a MoS2/NW heterostructure. (c) Variation of the E1g and A1g mode normalized intensities in a MoS2/NW heterostructure at different polarization angles of the incident light. The polarization angle θ denotes the angle between the NW long axis and the polarization detection angle. (d) Anisotropy of PL in the MoS2/NW heterostructure and bare MoS2. Solid lines in (b) and (d) are fitted curves using a cos2θ function.
has 3-fold (C3) rotational symmetry and inherently broken inversion symmetry.28 Typically, C3 symmetry of these materials can be broken by bending, applying strain, and reduction in dimensionality. Consequently, the materials manifest strong anisotropic vibrational, optical, and electrical responses.29–32

In this work, we report a new approach to engineer the light–matter interaction and symmetry of monolayer MoS2 by integrating it with 1D AlGaAs NW. Because of the strong electromagnetic (EM) field confinement in the NW, MoS2 photoluminescence (PL) increases significantly in the mixed-dimensional heterostructure compared with that in a bare MoS2 flake. Further, the mixed-dimensional heterostructure breaks the 3-fold rotational symmetry of 2D MoS2 explicitly, which leads to strong anisotropic optical responses. Additionally, we fabricate mixed-dimensional heterostructure-based phototransistors and benchmark their performance against bare MoS2 phototransistors. Compared with bare MoS2 devices, 2D MoS2/1D AlGaAs NW phototransistors show improved detectivity and photoresponsivity with marked anisotropic photoresponse due to enhanced light–matter interaction and symmetry breaking. Our findings establish mixed-dimensional heterostructures as a promising approach to realize strong and polarization-sensitive light–matter interactions in 2D materials for various photonic and optoelectronic applications.

**RESULTS AND DISCUSSION**

The crystal structure of monolayer 2H-phase MoS2 is illustrated in Figure 1a, in which one layer of Mo atoms is sandwiched between two layers of S atoms to form a 2D hexagonal lattice structure. The unit cell of monolayer MoS2 is noncentrosymmetric and belongs to the nonpolar D3h point group. In the monolayer MoS2 crystal, mirror reflection and 3-fold rotational symmetries are respected along the armchair direction. A schematic of the 2D MoS2/1D AlGaAs NW mixed-dimensional heterostructure is shown in Figure 1b. The chemical vapor deposition (CVD)-grown, monolayer MoS2 flakes are transferred onto NWs by wet transfer method (see Materials and Methods for details), as shown in the optical microscope image in Figure 1c. The typical diameter of the NWs used in this study is 80–150 nm. When monolayer MoS2 is transferred over a NW, the MoS2 layer covers the NW with a small suspended area close to both lower facets of NW. Figure 1d presents the typical room-temperature Raman spectra of 2D MoS2, a 1D AlGaAs NW, and a MoS2/AlGaAs mixed-dimensional heterostructure under 532 nm (~2.33 eV) laser excitation. In the range of 250 cm\(^{-1}\) to 450 cm\(^{-1}\), two distinctive MoS2 Raman modes—in-plane E2g (~385, 50 cm\(^{-1}\)) and out-of-plane A1g (~405 cm\(^{-1}\))—and the prominent AlGaAs NW mode—LO (~260 cm\(^{-1}\))—are observed. These modes agree with the previous literature.33–35 The peak at ~521 cm\(^{-1}\) corresponds to the Raman mode of the Si substrate.

The enhanced light–matter interactions in the mixed-dimensional heterostructure were investigated by comparing the linear optical responses of the MoS2/NW heterostructure with bare MoS2 samples. In Figure 2a, both MoS2/NW heterostructure Raman modes match perfectly with those of monolayer MoS2, which indicates the intact quality of the transferred monolayer MoS2 onto NW. Interestingly, the Raman intensity from the MoS2/NW heterostructure increased by ~3 times compared with the bare MoS2. In comparison with the Raman spectrum of bare MoS2 flake (red line), we observe a small peak shift of around ~0.6 cm\(^{-1}\) from both E2g and A1g modes of the MoS2/NW structure (blue line). This indicates a small strain and doping of MoS2 flake in the heterostructures.36–39 The estimated uniaxial local strain on MoS2 induced by a ~100 nm diameter NW is ~0.3% (see Section S1). Previously reported results show that the E2g Raman mode of MoS2 shifts by ~1.7 cm\(^{-1}\) per percent of strain.40 The Raman peak shift due to strain in our samples is comparable. Therefore, the strain effect in our mixed-dimensional heterostructures is negligible.

Figure 2b shows the room temperature PL spectra of MoS2 with and without NW under 532 nm excitation. We observe a strong PL emission from bare MoS2 at 1.86 eV. The PL intensity from the MoS2/NW heterostructure increases remarkably by a factor of ~9. We notice a small ~25 meV blue shift of the MoS2 A-exciton peak from the heterostructure, which could be the reason for the reduced binding energy of the exciton due to an increased Coulomb screening of monolayer MoS2 induced by NW.41 It is pertinent to note that the uniaxial tensile strain usually causes redshift in PL.40,42 However, we observe a blue shift in the PL which implies that there is no influence of strain in enhancing the light–matter interaction in our mixed-dimensional heterostructures.

Furthermore, the polarization-dependent Raman intensity variations are shown in Figure 2c with a polar diagram, where the maximum and minimum intensity of the MoS2 Raman modes are recorded when the polarization of the incident linearly polarized pump light is parallel (θ = 0°) and perpendicular (θ = 90°) to the NW long axis, respectively. We define polarization angles as the difference between the polarizations of the linearly polarized pump light and the NW long axis. The degree of anisotropy [defined as, (I\(_{\text{max}}\) − I\(_{\text{min}}\))/ (I\(_{\text{max}}\) + I\(_{\text{min}}\))] is calculated as ~34% and 36% for modes E2g and A1g, respectively. A polar plot for comparing the polarization-dependent PL responses from bare MoS2 and MoS2/NW heterostructure samples is shown in Figure 2d. We do not observe any polarization angle dependency of PL emission from bare MoS2. In marked contrast, MoS2 shows a strong anisotropic PL response from the heterostructure. The PL intensity reaches its maximum (minimum) value when the excitation polarization is parallel (perpendicular) to the NW axis. The degree of MoS2 PL anisotropy from the heterostructure is calculated as ~60%. The effective dimension of MoS2 is reduced from 2D to 1D at the heterostructure region, which breaks the C3 rotational symmetry of MoS2 and leads to strong anisotropy in the PL response.40 The PL response from monolayer MoS2 is much stronger than the Raman signals. This could be the reason for such a difference in the enhancement factor and degree of anisotropy between the Raman and PL of MoS2/NW.

The Raman and PL enhancement factor (EF), defined as the intensity ratio of signals measured in MoS2/NW and MoS2, depends on the NW diameter. In this work, the diameter of NWs varies between 80 and 150 nm (see Figure S1a). In general, EF increases with an increasing NW diameter, which offers a larger interfacial area in the heterostructures. We observe the highest Raman and PL EF of ~3 and ~9, respectively, with NWs of ~150 nm diameter. The MoS2 Raman and PL EF as a function of NW diameters is shown in Figure S1b.

The enhanced PL emission from our heterostructures may arise from different reasons, e.g., charge transfer,41–43 and
Figure 3. PL responses of a hBN intercalated MoS$_2$/hBN/NW heterostructure. (a) Schematic of the heterostructure. (b) Optical image (left panel) of the sample, where the white dashed line indicates the area of monolayer MoS$_2$ partially covering the hBN layer and a NW. Scale bar: 2 μm. The PL intensity mapping (right panel) is taken from the area marked in an orange rectangular box in the optical image. Scale bar: 1 μm. (c) PL spectra from bare MoS$_2$, MoS$_2$/hBN, and MoS$_2$/hBN/NW regions.

Figure 4. Numerical simulations of EM field distribution in mixed-dimensional heterostructure under 532 nm excitation. (a) EM field confinement around MoS$_2$/NW heterostructure for excitation polarization parallel (left panel) and perpendicular (right panel) to the NW axis (in this case, y axis). (b) Variation of the EM field density at the top of the NW for different excitation polarization states.

optical field confinement in NW. On the basis of the Al composition of ∼30% (±5%) in AlGaAs NW in this study, the MoS$_2$ and NW energy diagrams suggest a type-II band alignment at the heterostructure interface (see Figure S8a). Therefore, quenching of the MoS$_2$ PL from the heterostructure is expected. In contrast, our results show enhancement in the MoS$_2$ PL response from the heterostructure. We fabricated a mixed-dimensional sample where a multilayer (~11 nm) hexagonal boron nitride (hBN) flake is inserted between the monolayer MoS$_2$ and AlGaAs NW to determine the reason behind the enhanced optical properties in the MoS$_2$/NW heterostructure and rule out the charge transfer mechanism. The insulating hBN would prevent any possible charge transfer in the heterostructure. Figure 3a,b shows the schematic and the optical image of the hBN intercalated mixed-dimensional heterostructure, respectively. A PL mapping taken from the highlighted area of the optical image shows maximum emission from the MoS$_2$/hBN/NW region. The PL responses at different locations are presented in Figure 3c. A blue shift of ∼50 meV in MoS$_2$ PL from both the MoS$_2$/hBN and MoS$_2$/hBN/NW regions compared with that in the bare MoS$_2$ (on SiO$_2$/Si) is attributed to the hBN substrate effect. Compared with SiO$_2$, hBN provides an atomically clean interface with 2D materials because of the low-density of charged impurities. Therefore, the enhancement in optical and electrical characteristics of 2D materials on hBN is expected. The PL intensity of MoS$_2$ on hBN increases by ~9 times compared with bare MoS$_2$. Surprisingly, the PL intensity from the MoS$_2$/hBN/NW heterostructure increases further by ~3 times compared with that in the MoS$_2$/hBN heterostructure. This confirms that the charge transfer phenomenon is not responsible for the enhanced optical properties from our mixed-dimensional heterostructures.

We perform numerical simulations employing finite element modeling to understand the origin of the enhanced optical response in our mixed-dimensional heterostructure. For the simulation, we consider a monolayer MoS$_2$ with a refractive index ($n$) of ~3 for a 532 nm laser excitation is placed on a single hexagonal AlGaAs NW ($n=3.5$) of 100 nm diameter. The heterostructure resides on a SiO$_2$/Si substrate with a 285 nm thick SiO$_2$ capping layer ($n=1.45$). The model structure is illuminated normally with a linearly polarized excitation at a wavelength of 532 nm. As shown in Figure 4a, our results reveal strong EM field confinement in the NW, because of its high aspect ratio, and a large refractive index contrast compared with the surrounding media. Interestingly, when the excitation polarization direction is parallel to the NW axis, the confinement occurs at the top and bottom surfaces of the NW. In contrast, for the perpendicular configuration, the confinement becomes localized only near the bottom sides of the NW. The strong field confinement at the top of the NW results in an enhanced light–matter interaction in MoS$_2$, which leads to a higher exciton excitation rate in the heterostructure region than in bare MoS$_2$. Figure 4b summarizes the strength of the EM field confined only at the top of NW under different excitation polarization. Because of the anisotropy of the confined field and the 3-fold rotational symmetry breaking in monolayer MoS$_2$, the strong anisotropy of the MoS$_2$ PL in the mixed-dimensional heterostructure is enabled.

After understanding the anisotropic and enhanced light–matter interactions in the MoS$_2$/NW heterostructures, we now fabricate MoS$_2$/NW-heterostructure-based photodetectors to...
benchmark them against the performance of bare MoS$_2$ devices. The inset of Figure 5a shows an optical image of the fabricated devices. All phototransistors show n-type behavior in the transfer curve (see Figure S5a). The output characteristics of the devices under dark and 532 nm laser illumination conditions at a constant gate voltage ($V_g$) of 20 V are shown in Figure 5a,b. The excitation polarization direction is always kept parallel to the NW axis during all the photocurrent measurements. Under laser illumination, the current density ($J_d$) in the devices increases for a corresponding gradual increase in incident optical power. The calculated photocurrent ($I_{ph}$) values, defined as $I_{light} - I_{dark}$, as a function of laser power in a range of 50–500 µW are presented in Figure 5c. Saturated $I_{ph}$ curves are observed in both types of phototransistors. In comparison with the bare MoS$_2$ photodetector, we observe ~5 and ~8 times enhancement in $I_{ph}$ from the MoS$_2$/NW device at a laser power of 50 µW and 500 µW, respectively. This can be attributed to the strong EM field confinement at the top of NW that reinforces the applied electric field in the MoS$_2$ channel to pass more photogenerated carriers toward the electrodes. For photodetectors, responsivity ($R$) and specific detectivity ($D^*$) are two important parameters that give a measure of conversion efficiency and ability to detect weak optical signals, respectively:

$$R = \frac{I_{ph}}{P_{in}}$$

(1)
\[ D^* = \frac{RA^{1/2}}{(2eI_{dark})^{1/2}} \]

where \( P_{inc}, A, \) and \( e \) are incident optical power, irradiation area, and the elementary charge of the electron, respectively. Figure 5d includes a comparison of calculated \( R \) and \( D^* \) values at different laser powers. In general, the values of both parameters decrease with increasing laser power. At lower power, the higher responsivity in all devices results from a reduced recombination of the electrons as the trap states in MoS\(_2\) capture photoexcited electrons, which leads to a prolonged carrier lifetime.\(^{52} \) At higher power, a dominating carrier recombination rate results in lower responsivity. In comparison with bare MoS\(_2\), at 100 \( \mu \)W laser power we observe an \( \sim 3 \) and \( \sim 5 \) times increase in \( R \) and \( D^* \), respectively, from the MoS\(_2/\)NW device.

We study the dependence of photocurrent with linearly polarized light aligned parallelly (\( 0^\circ \)) and perpendicularly (\( 90^\circ \)) to the NW long axis to further understand the orientation of the maximum photocurrent in the MoS\(_2/\)NW FET. As shown in Figure 6a, the maximum (\( \sim 36 \) nA) and minimum (\( \sim 27 \) nA) \( I_d \) are obtained with parallel and perpendicular polarization of the light, respectively, under the 532 nm laser with 100 \( \mu \)W power. The photocurrent mapping in Figure 6b further ensures the enhanced photocurrent along the NW, which varies with \( 0^\circ \) and \( 90^\circ \) polarization direction of the laser. This polarization-dependent photocurrent from MoS\(_2\) at the heterostructure is also attributed to the breaking of the rotational symmetry of monolayer MoS\(_2\) because of reduced dimensionality. Thus, an anisotropic photoresponse from isotropic 2D materials is achievable by virtue of 2D/1D mixed-dimensional heterostructures.

## CONCLUSIONS

We demonstrate enhancement and excitation polarization-sensitive optical and optoelectronic properties of monolayer MoS\(_2\) by integrating it with high refractive index AlGaAs NW. The Raman and PL intensity of MoS\(_2\) increases by \( \sim 3 \) and \( \sim 9 \) times in the MoS\(_2/\)NW heterostructures compared with the bare MoS\(_2\) samples. Further, we observe a strong anisotropic response in the PL with an \( \sim 60\% \) degree of anisotropy because of the breaking of rotational symmetry induced by the NW. Numerical simulations reveal that excitation polarization-dependent EM field confinement of NWs is the basis of such optical responses. The mixed-dimensional photodetectors offer improved device performance with polarization sensitivity. In comparison with bare MoS\(_2\) phototransistors, the responsivity and specific detectivity in MoS\(_2/\)NW devices increase by \( \sim 3 \) and \( \sim 5 \) times, respectively. Our results pave the way for 2D/1D mixed-dimensional heterostructure-based high-performance photonic and optoelectronic devices.

## MATERIALS AND METHODS

### Sample Preparation.

AlGaAs NWs are grown on a cleaned Si (111) substrate inside a horizontal flow atmospheric pressure metal–organic vapor phase epitaxy (MOVPE) system.\(^{27,43} \) NWs grown in this technique are predominantly zinc-blende phase. The cross-section of the NWs is assumed to be a hexagonal shape since most of the reported III–V semiconductor NWs grown along the [111] direction have a hexagonal crosssection.\(^{53,55} \) Afterward, as-grown NWs are transferred on top of the p-doped silicon substrate covered with a 285 nm thick SiO\(_2\) by nano combing technique.\(^{41} \) Monolayer and few-layer MoS\(_2\) crystals are grown on O\(_3\) plasma cleaned SiO\(_2\)/Si substrate using a salt assisted CVD method.\(^{56} \) After a wet transfer process, the as-grown monolayer MoS\(_2\) flakes are transferred onto the NW nanocombed substrate. For hBN intercalated mixed-dimensional samples, commercially available MoS\(_2\) and hBN crystals (2D Semiconductors, USA) are used for exfoliation. All the samples are annealed at 250 °C for 2 h in a vacuum to ensure a clean interface.\(^{57} \)

### Raman and PL Spectroscopy.

The room-temperature Raman and PL spectra are collected in backscattering geometry with a confocal micro-Raman system (WITec alpha300 RA+). Samples are excited using a 532 nm laser with a spot size of less than 1 mm (\( \times 100 \) objective, 0.9 NA). Low laser power (<500 \( \mu \)W) is used to avoid laser-induced damage in the samples.

### Device Fabrication and Optoelectronic Measurements.

After the material transfer, electron beam lithography (EBL Vistec, EPBG 5000) and metallization (MASA, IM-9912) are performed to realize Ti/Au (5/50 nm) electrodes. Optoelectronic measurements are carried out with a custom-built setup consisting of a confocal microscope (WITec Alpha 300 RA+) and two source meters (Keithley 2400 and 2401). During photomeasurements, samples are illuminated with \( \times 20 \) [numerical aperture (NA) = 0.4] and \( \times 100 \) (NA = 0.9) objective lenses to ensure proper illumination of the entire effective channel region.

### Theoretical Simulation.

Theoretical simulations were performed with the finite element modeling method using COMSOL Multiphysics. A hexagonal structure of 100 nm width with a 3.5 refractive index\(^{50} \) is placed on top of a 285 nm thick SiO\(_2\) layer, which is on top of a Si substrate. A linearly polarized plane wave is illuminated normally from the top to the bottom of the structure. The polar angle of this plane wave is varied from 0 to 360°; a polarization angle of 0 degrees corresponds to an EM field oscillating parallel to the NW, while a 90° angle corresponds to the perpendicular oscillation. Section S3 of the Supporting Information provides more information on simulations.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.2c07705.

AFM characterization and NW diameter-dependent PL responses of the mixed-dimensional heterostructure; differential reflectivity spectra of the heterostructure, and PL response of a single AlGaAs NW; details of the simulation, transfer curves, and output \( I_d−V_d \) curves of devices in dark condition; transfer curves of the devices under light illumination; photocurrent of the devices under different laser illuminations; and energy band alignment and charge transfer between MoS\(_2\) and AlGaAs NW (PDF)

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Notes

The authors declare no competing financial interest.

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