Highly Tunable Layered Exciton in Bilayer WS_2 : Linear Quantum Confined Stark Effect versus Electrostatic Doping

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ABSTRACT: In the 1H monolayer transition metal dichalcogenide, the inversion symmetry is broken, while the reflection symmetry is maintained. On the other hand, in the bilayer, the inversion symmetry is restored, but the reflection symmetry is broken. As a consequence of these contrasting symmetries, here we show that bilayer WS_2 exhibits a quantum confined Stark effect (QCSE) that is linear with the applied out-of-plane electric field, in contrast to a quadratic one for a monolayer. The interplay between the unique layer degree of freedom in the bilayer and the field driven partial interconversion between intralayer and interlayer excitons generates a giant tunability of the exciton oscillator strength. This makes bilayer WS_2 a promising candidate for an atomically thin, tunable electro-absorption modulator at the exciton resonance, particularly when stacked on top of a graphene layer that provides an ultrafast nonradiative relaxation channel. By tweaking the biasing configuration, we further show that the excitonic response can be largely tuned through electrostatic doping, by efficiently transferring the oscillator strength from neutral to charged exciton. The findings are prospective toward highly tunable, atomically thin, compact, and light, on chip, reconfigurable components for next generation optoelectronics.

KEYWORDS: bilayer WS_2 , excitonic reflection, absorption, quantum confined stark effect

The ultrafast radiative recombination rate^{1–3} of the strongly bound excitons^{4–6} in monolayer transition metal dichalcogenides (TMDCs) leads to efficient photo-emission.^{7,8} The fast reradiation of the absorbed photon by the exciton without ample opportunity to suffer from nonradiative processes results in near perfect excitonic reflectance.^{9–11} On the other hand, in a bilayer TMDC, a lower indirect energy bandgap appears while the direct energy gap at the $K(K')$ point of the Brillouin zone is maintained. This causes the momentum-direct excitons in bilayer to encounter strong nonradiative processes,¹² and thus a bilayer TMDC is weakly luminescent,¹³ while still exhibiting strong excitonic absorption.

To be able to tune the excitonic absorption in a dynamic manner is crucial to multiple applications such as programmable optical modulators, tunable photodetectors, optical computing and imaging, laser pulse shaping, and ultralight optomechanical mirrors. A bilayer distinguishes itself from the monolayers in a number of ways in terms of tunability of the exciton oscillator strength and its spectral control. First, the reflection symmetry along the c -axis is present in a monolayer TMDC, but it is broken in a bilayer. Thus, in the presence of a vertical electric field, one expects that a bilayer would show a linear Stark shift, in contrast to a quadratic one,^{14–17} in the

monolayer. In spite of theoretical predictions of linear quantum confined Stark effect (QCSE) in bilayer TMDC,^{18–25} there has not been any experimental demonstration of the same to date for the direct intralayer A exciton. Second, the presence of two layers in a bilayer, which are rotated at 180° with respect to each other, offers an additional layer degree of freedom.¹² With a vertical electric field, there is a possibility to convert the intralayer exciton at zero field to an interlayer exciton, resulting in a strong tunability of the oscillator strength. Third, it is easier to inject carriers in a bilayer from a metal contact due to reduced bandgap, increased valley degeneracy, and enhanced orbital coupling to metal.²⁶ This allows fast and efficient modulation of the excitonic oscillator strength by interconversion between a neutral exciton and a charge exciton (trion) through electrostatic doping.^{27–29}

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Figure 1. Intra- and interlayer direct exciton in bilayer WS₂. (a) Left panel: Lowest energy bands of bilayer WS₂ for a given spin-valley ($l_+ = s_z \tau_z = +1$) configuration around $\mathbf{K}$ ($\mathbf{K}'$) point. Right panel: The corresponding layer distribution of the electron and hole wave functions for $\mathbf{k} = \mathbf{K} + \Delta\mathbf{k}$ where $\Delta\mathbf{k} = \frac{2\pi}{a}[0.0033, 0.0033]$, a is the lattice constant. (b) Exciton energy band structure as a function of center of mass momentum ($\mathbf{Q}$) showing different 1s and 2s bands. (c) The $\mathbf{k}$ space distribution around $\mathbf{K}$ for both the interlayer [$A_{1s}^{(2)}$] and the intralayer [$A_{1s}^{(1)}$] exciton at $\mathbf{Q} = \mathbf{0}$ for the given layer index (l_+). (d) Schematic of the isolated bilayer (sample S1) WS₂ on Si/SiO₂ substrate. (e) Temperature dependent (4.2 to 295 K) differential reflectance of bilayer WS₂ governed by the intralayer exciton. The dashed lines show the fitting of the spectra obtained from the model. (f) Color plot of $\frac{d}{dE}\left(\frac{\Delta R}{R}\right)$ as a function of energy and temperature. (g) Normalized oscillator strength of the intralayer $A_{1s}^{(1)}$ exciton obtained from the fitting as a function of temperature.

In this work, we use two different device structures to segregate the effects of electric field induced QCSE and electrostatically induced doping on both the strength and the spectral tunability of the excitonic response in a bilayer WS₂. We use a few-layer graphene (FLG)/hBN/2L-WS₂/hBN/FLG stack to keep the bilayer WS₂ electrically floating to report the linear QCSE of the direct intralayer exciton. The corresponding excitonic oscillator strength is shown to exhibit a large tunability due to an electric field driven partial interconversion between intralayer and interlayer exciton. We further demonstrate that placing a graphene layer right under the bilayer WS₂ is promising for an atomically thin, gate tunable electro-absorption layer. To demonstrate the electrostatically induced doping effect, we use a similar stack, but with the bilayer WS₂ connected to a grounded metal contact. The efficient carrier injection from the contact leads to effective oscillator strength transfer from neutral exciton to positive or negative charged trions depending on the polarity of the gate voltage.

In the 2H form of bilayer WS₂, the individual layers are rotated by 180° with respect to each other, making the unit cell inversion symmetric. This requires an additional layer degree of freedom ($l_{\pm} = \pm 1$) to describe excitons in bilayer for a given spin ($s_z = \pm 1$) and valley ($\tau_z = \pm 1$). To describe the lowest energy excitonic states in a bilayer, we use a simple quasiparticle Hamiltonian using tungsten d orbitals as the basis functions^{12,30} and solve the Bethe–Salpeter (BS) equation,^{12,31} as described in the Supporting Information, Section S1. As an example, in Figure 1a, we schematically illustrate the lowest energy conduction and valence bands for one of the degenerate cases (l_+) along with the layer

distribution of the wave functions close to the $\mathbf{K}$ point. The energy dispersion of lowest energy excitonic states with center of mass momentum ($\mathbf{Q}$) is schematically depicted in Figure 1b. The calculated $\mathbf{k}$ -space distribution of the excitonic state with $\mathbf{Q} = \mathbf{0}$ is depicted in Figure 1c. Depending on the layer distribution of the single particle wave functions, we have both intralayer [$A_{1s}^{(2)}$] and interlayer [$A_{1s}^{(1)}$] 1s excitons, with the latter being the lowest energy one for bilayer WS₂. The layer index of the intralayer exciton is coupled with spin and valley such that $l_{\pm} = s_z \tau_z = \pm 1$,¹² which results in two degenerate configurations for each at zero external field. While radiative recombination is allowed for both these types of excitons by standard selection rules, the intralayer exciton exhibits more than an order of magnitude higher radiative decay rate compared with the interlayer exciton,¹² and hence, the optical response in bilayer discussed here is dominated by the intralayer $A_{1s}^{(2)}$ exciton.³² The interlayer $A_{1s}^{(1)}$ exciton noted above is momentum-direct with $\mathbf{Q} = \mathbf{0}$,^{12,33–36} which is distinct from the indirect interlayer exciton.^{37,38} Due to the difference in spin splitting of the bright and the dark states in Mo and W-based TMDCs, the interlayer exciton is energetically above the intralayer exciton in MoTe₂,³³ MoSe₂,³⁴ and MoS₂,^{35,36,39} while it is below the intralayer exciton in WS₂.¹²

To extract the spectral feature of the intralayer exciton in isolated bilayer WS₂ transferred on 285 nm SiO₂/Si substrate (sample S1 as schematically shown in Figure 1d, see Supporting Information, Section S2 for Raman characterization), we perform differential reflectance measurement (see Methods for measurement details). Figure 1e shows the temperature dependent differential reflectance $\left(\frac{\Delta R}{R} = \frac{R_{on} - R_{off}}{R_{off}}\right)$

Figure 2. Linear QCSE for bilayer WS₂. (a) Schematic diagram of the broken reflection symmetry in a bilayer WS₂ along the *c*-axis while it is maintained for monolayer. (b) Calculated electric field dependent linear shift in intralayer 1s exciton position obtained from the BS equation. Here the top axis shows the interlayer potential difference while the bottom axis denotes the corresponding electric field. (c) Schematic diagram of the device sample S2-F where bilayer WS₂ is sandwiched between hBN and graphite layers. The vertical field is applied between the top and the bottom graphene layers while the WS₂ layer is kept electrically floating. (d) Experimental linear Stark shift (in symbols) for the bilayer device. The solid line represents a linear fit to the Stark shift at low field regime. (e) Change in the binding energy ($\Delta\zeta_b$) of the exciton with electric field.

in the range of 4.2–295 K. Throughout the text, R_{on} denotes the reflected intensity from full stack and R_{off} denotes the same from the stack without the bilayer WS₂. For notational simplicity, we denote experimentally observed neutral exciton and charged trion peaks as X^0 and $X^\pm$, respectively. The optical response and the line shape are modified by the interference between WS₂ and substrate reflection, and the excitonic response of the bilayer is deconvoluted from the overall response. To reproduce the reflectance spectra obtained from the experiment, the exciton contribution to the dielectric response is represented as a Lorentzian oscillator:^{40–42}

$$\epsilon(E) = \epsilon_\infty + \sum_j \frac{\lambda_j}{E_j^2 + E^2 - i\gamma_j E} \quad (1)$$

where the ϵ_∞ is the background response, λ_j and E_j are the oscillator strength and the position of the *j*th oscillator respectively with a broadening parameter γ_j . R_{on} and R_{off} are then calculated using transfer matrix method (see Supporting Information, Section S3 for more details). The color plot of $\frac{d}{dE} \left(\frac{\Delta R}{R} \right)$ in Figure 1f shows a conspicuous spectral red shift of the neutral X^0 exciton with an increase in temperature, which results from a corresponding reduction in the bandgap. The corresponding temperature tunability of the exciton oscillator strength is shown in Figure 1g.

Due to the reflection symmetry in a monolayer WS₂ (Figure 2a), in the presence of an out-of-plane electric field, the resulting spatial symmetry of the wave function along the *c*-axis forces the first order perturbation term of the energy eigenvalue to be zero. This causes a zero linear Stark shift in

monolayer, and the second order perturbation provides a quadratic QCSE, as verified experimentally.^{14–17} However, the reflection symmetry is broken in a bilayer case (Figure 2a) and the resulting asymmetry in the layer distribution of the intralayer exciton is clearly visible in Figure 1a,b. This in turn results in a nonzero first order perturbation term and thus a linear QCSE. The electric field dependent linear shift in the intralayer 1s exciton position as obtained from the solution of the BS equation (see Supporting Information, Section S1 for calculation details) is shown in Figure 2b. We do not consider any additional screening of the electric field inside the bilayer.

To demonstrate the linear QCSE experimentally, and to segregate the electric field effect from the doping effect, we prepare a FLG/hBN/2L-WS₂/hBN/FLG (sample S2) stack as schematically shown in Figure 2c (see Methods for sample preparation). The FLG layers act as transparent gate electrodes in the back reflection geometry. The thickness of the top and the bottom hBN layers is 20 nm each. The high quality of the hBN layers allows us to apply high electric field without appreciable gate leakage (see Supporting Information, Section S4). In the field-configuration (denoted as S2-F), a voltage (V_g) is applied at the top gate electrode keeping the bottom gate grounded. The electrode directly contacting the bilayer WS₂ is kept in open circuit mode, avoiding carrier injection and thus the WS₂ is electrically floating. This allows us to study the electric field effect without any confounding electrostatic doping introduced by V_g . In this mode of operation, the lack of gate-induced or photoinduced doping¹⁴ in our sample is evidenced by the absence of any signature of trion in the entire V_g range. The top FLG layer partially covers the device and the

Figure 3. Modulation in oscillator strength with electric field due to intra- to interlayer conversion of excitons. (a) Extracted oscillator strength (normalized) for the sample S2 with the applied vertical electric field. The dashed line shows a guide to eye. (b) k space distribution of the contribution of electron ($|\psi_e(\mathbf{k})|^2$) and hole ($|\psi_h(\mathbf{k})|^2$) for a given layer index (l_+) at different interlayer potential (or electric field) values. The corresponding k space distribution of the exciton at $\mathbf{Q} = \mathbf{0}$ is presented in the right panel, indicating largest contribution from regions close to the $\mathbf{K}$ point. As the vertical field increases, the electron wave function moves away from the $\mathbf{K}$ point, and the interlayer character increases.

sample is illuminated in the FLG-covered portion (Figure 2c). $\frac{\Delta R}{R}$ and $\frac{d}{dE}\left(\frac{\Delta R}{R}\right)$ for the stack are provided in the Supporting Information, Section S5, at different electric fields, measured at 4.2 K. The extracted X^0 peak shift (ΔE_X^0) with respect to zero field is plotted in Figure 2d as a function of V_g (top axis) and the vertical electric field ξ (bottom axis). The electric field inside the bilayer WS_2 is estimated as

$$\xi = \frac{V_g}{d_w + d_h \frac{\epsilon_{\perp,0,w}}{\epsilon_{\perp,0,h}}} \quad (2)$$

where $d_{w(h)}$ is the thickness of the bilayer WS_2 (hBN), $\epsilon_{\perp,0,w}$ ($=6.3$) and $\epsilon_{\perp,0,h}$ ($=3.76$)⁴³ are the static out-of-plane dielectric constants of WS_2 and hBN, respectively. We find that ΔE_X^0 varies linearly with $|\xi|$ —a direct evidence of linear QCSE in bilayer. The average slope of the lines fitting the Stark shift in Figure 2d suggests an out of plane static dipole moment value of ~ 5.8 meV/MVcm⁻¹. The predicted electric field required to provide the same shift as in the experiment is less in the calculation, which indicates a strong screening⁴⁴ in the bilayer WS_2 , particularly in the presence of optical excitation.

At higher $|\xi|$, the experimental Stark shift deviates from linearity as the rate of shift reduces, pointing to strong excitonic effects⁴⁵ in the QCSE. The exciton binding energy (ζ_b) reduces at higher electric field, which results in a blue shift in the X^0 position, partially compensating the QCSE induced red shift. In Figure 2e, we plot the difference between the shift predicted from the fitted line and the experimental values (from Figure 2d), which is an estimate of $\Delta\zeta_b$ with an increase in $|\xi|$. The reduction in ζ_b is attributed to two many-body effects. First, an electric field in the out of plane direction forces the electron and hole to move away from each other,^{46,47} reducing the spatial overlap of the redistributed wave functions. The second effect arises due to field induced orientation of the excitons in the out of plane direction. The resulting proximity of the particles with similar polarity enhances exciton–exciton repulsion, causing an excitonic blue shift.^{48,49}

Figure 3a shows that the extracted oscillator strength (λ) of X^0 sharply reduces with an increase in $|\xi|$, which is promising

for atomically thin tunable electro-optic devices. Such a reduction in λ with increasing $|\xi|$ is in contrast with monolayer samples, where the photoluminescence intensity is usually a weak function of the vertical field. Since the reduction in binding energy ($\Delta\zeta_b$) discussed above is quite small compared with ζ_b , the exciton remains vertically confined in the bilayer^{46,47} and thus dissociation of the exciton under the applied field is an unlikely possibility. As we do not observe any trion peak at higher $|\xi|$, in congruence with the floating WS_2 channel, transfer of oscillator strength from neutral exciton to trion is also not plausible. We attribute this reduction of λ to a field driven conversion of intralayer exciton to interlayer exciton. The simple Hamiltonian described in the Supporting Information, Section S1, is again useful to get a qualitative understanding of the underlying physics. At the left and the middle panels of Figure 3b, we plot, in the two-dimensional k space centered around $\mathbf{k} = \mathbf{K}$, the fractional contribution of the individual layers to the electron ($|\psi_e(\mathbf{k})|^2$) and the hole ($|\psi_h(\mathbf{k})|^2$) constituting the l_+ exciton at $\mathbf{Q} = \mathbf{0}$ for different vertical fields. The k -space distribution of the exciton in the right most panel suggests that the maximum contribution of the $1s$ exciton is from the states close to $\mathbf{k} = \mathbf{K}$. At $\xi = 0$, the bright exciton arises from electron and hole both lying at the same layer (intralayer exciton at layer 1 in the top row of Figure 3b). However, with an increase in ξ , the contributing electron concentration around $\mathbf{k} = \mathbf{K}$ is pushed to the layer 2, while the hole distribution is maintained at layer 1 (bottom three rows). Since there is a large weight of the electronic states close to $\mathbf{k} = \mathbf{K}$ in the formation of the $\mathbf{Q} = \mathbf{0}$ exciton, this gives rise to an increasing interlayer character of the exciton. This, in turn, reduces the oscillator strength with an increase in the vertical electric field.

The intralayer $A_{1s}^{(2)}$ exciton in bilayer WS_2 has fast nonradiative scattering channels to lower lying interlayer $A_{1s}^{(1)}$ exciton at the $\mathbf{K}(\mathbf{K}')$ point¹² and to the indirect band edge states. The relaxation channel can be further aided if the bilayer WS_2 is stacked on a few layer graphene, due to ultrafast interlayer carrier transfer processes^{50–54} (top panel of Figure 4a). This limits the possibility of reradiation making the bilayer $\text{WS}_2/\text{graphene}$ stack an excellent atomically thin tunable

Figure 4. QCSE of bilayer excitons directly placed on few-layer graphene. (a) Top panel: Cartoon picture of the ultrafast interlayer transfer process from WS₂ to few-layer graphene. Bottom panel: Schematic diagram of the device (sample S3) where bilayer WS₂ is directly placed on graphene. Bias is applied between the two graphene layers. (b) 2D color plot of $\frac{d}{dE}\left(\frac{\Delta R}{R}\right)$ from sample S3 across the applied voltages. The exciton resonance is red-shifted at higher field and the corresponding oscillator strength decreases. (c, d) Stark shift and the normalized oscillator strength of WS₂ as a function of the applied vertical field (bottom axis). The top axis shows the applied voltage. The solid lines in part c represent a linear fit to the data, and the dashed lines in part d are guides to eye.

absorber when excited at the exciton resonance. The ultrafast charge transfer from WS₂ to graphene also results in a strong dedoping of the WS₂ layer. This is investigated in a stack of FLG/2L-WS₂/hBN/FLG (sample S3), as schematically illustrated in the bottom panel of Figure 4a. The temperature dependent $\frac{\Delta R}{R}$ and the corresponding oscillator strength at different temperatures are shown in Supporting Information, Section S6. Figure 4b depicts the $\frac{d}{dE}\left(\frac{\Delta R}{R}\right)$ in a color plot at different vertical electric fields. The extracted excitonic red shift and the reduction in the oscillator strength are shown separately in parts c and d of Figure 4. While the linearity in the Stark shift is maintained in this structure as well, we observe about 4-fold reduction in the slope (1.48 meV/MVcm⁻¹) compared with device S2-F, requiring a higher electric field to obtain a similar Stark shift. This can be correlated with the additional screening in the bilayer WS₂ due to the proximity of the graphene layer.

We now investigate the role of electrostatic doping induced by the gate in tuning the intralayer exciton. For this purpose, we again use sample S2, however, now in a doping configuration (denoted as S2-D). In this mode of operation, the top gate electrode is electrically open circuited, and the metal pad directly contacting the WS₂ is electrically grounded. The bottom FLG layer is used as the gate electrode, and a voltage is applied with respect to the grounded electrode, as

shown in Figure 5a. The metal contact can efficiently inject both electrons and holes into the bilayer WS₂, depending on the polarity of the gate voltage (V_g). Figure 5b shows a two-dimensional color plot of the first derivative of the differential reflectance as a function of V_g (individual spectra are presented in Supporting Information, Section S7). At low $|V_g|$, the optical response is dominated by X^0 , while at higher positive (negative) V_g , X^- (X^+) dominates the response, red shifting the resonance energy.

The small blue shift (≈ 2 – 3 meV) of the exciton with an increase in $|V_g|$ (Figure 5c) can be attributed to a combined effect of (a) renormalization of the excitonic gap due to the increased doping,^{55,56} (b) reduction in exciton binding energy due to separation of electron–hole wave function along the *c*-axis by the gate field,^{46,47} and (c) reduction in the exciton binding energy due to enhanced exciton–exciton repulsion due to oriented exciton dipoles.^{48,49} On the other hand, the trion position red shifts with an increase in $|V_g|$, leading to an increased separation between the exciton and trion resonance energies (Figure 5d). Similar observations are reported in monolayer as well,^{57,58} which is usually attributed to the enhancement in the energy required to move the additional electron (for X^-) or hole (for X^+) to respective bands due to doping induced Pauli blocking.^{52,59}

The extracted oscillator strength of X^0 and $X^\pm$ in Figure 5e shows a doping dependent transfer of oscillator strength from one excitonic species to another. At low V_g , when the bilayer Fermi level is deep inside the band gap, X^0 formation is favored. Efficient carrier injection in bilayer allows us to sweep the Fermi level almost the entire bandgap, providing strong n-type (p-type) doping at positive (negative) V_g , which favors the formation of X^- (X^+), suppressing the formation of X^0 . Such a near complete modulation of oscillator strength of different excitons is promising for electrically tunable ultrathin optoelectronic components.

We observe a sharp increase in the extracted γ_f for both X^0 and $X^\pm$ with an increase in $|V_g|$ (Figure 5f). For X^0 , this is attributed to an enhanced Coulomb scattering in the presence of large electron or hole density, and neutral exciton to trion conversion. The charged nature of the trion species makes the Coulomb scattering stronger resulting in a larger broadening. Similar enhancement in broadening has been observed previously in III–V quantum wells under vertical electric field,⁴⁶ and has been attributed to increased rate of dissociation of exciton in the presence of the electric field. However, in the present case, both the exciton and trion binding energy are significantly larger than their III–V quantum well counterpart. In addition, a stronger quantum confinement in the out of plane direction forces the change in the binding energy to be relatively small, as discussed previously. It thus appears that dissociation induced increase in broadening may play a negligible role in our experiment, particularly for the neutral exciton.

In summary, We propose bilayer WS₂ as an atomically thin, tunable electro-absorption layer when excited at the excitonic resonance. The spectral feature as well as the oscillator strength of the excitonic response is highly tunable by an external gate voltage through both electrostatic doping and quantum confined Stark effect. Contrary to the monolayer case, the quantum confined Stark effect in bilayer shows a linear variation with electric field owing to reflection symmetry breaking, coupled with a large modulation in the oscillator strength due to a partial interconversion between intralayer

Figure 5. Electrostatic doping induced tuning of excitons and trions in bilayer WS_2 . (a) Schematic of the device (S2-D) used for gating analysis. Here the bilayer WS_2 is sandwiched between hBN layers but directly connected to one of the electrode. Voltage is on the bottom graphene layer keeping the electrode connecting the WS_2 layer grounded. (b) 2D color plot of $\frac{d}{dE} \left(\frac{\Delta R}{R} \right)$ as a function of the gate voltage (V_g). The exciton (X^0) and trion ($X^\pm$) dependence with the V_g is also clearly observed. (c) V_g dependent X^0 and $X^\pm$ positions. (d) Increase in separation between the X^0 and the $X^\pm$ with increasing V_g . (e) Extracted oscillator strength (normalized) of the X^0 and $X^\pm$ as a function of V_g . (f) Extracted broadening parameter (γ) with V_g variation.

and interlayer exciton. The electrically tunable nature of the excitonic response has interesting prospects for ultrathin, light and compact, fast, reconfigurable optoelectronic applications like modulators, pulse shapers, photodetectors, digital mirrors and phased arrays, tuning opacity of surfaces, and possible elements for optical and quantum information processing.

METHODS

Device Fabrication. The heterojunctions are prepared using dry transfer of individual exfoliated layers to a Si substrate coated with 285 nm SiO_2 layer. The transfer of each layer is performed one at a time under a microscope with controlled translational and rotational stages for precise alignment. After the transfer of each layer, the sample is heated on a hot plate at 80 °C for 2 min for improved adhesion between layers. Optical images of samples S2 and S3 are shown in the Supporting Information, Section S8, after all the layers are transferred. Standard nanofabrication techniques are used to define the metal electrodes contacting the graphene layers. The substrate with the transferred layers is spin coated with PMMA 950C3 and baked on a hot plate at 180 °C for 2 min. This is followed by electron beam lithography with an acceleration voltage of 20 kV, an electron beam current of 210 pA, and an electron beam dose of 220 $\mu\text{C cm}^{-2}$. Patterns are developed using MIBK:IPA solution in the ratio 1:3. Later samples are washed in IPA and dried in N_2 blow. Electrodes are made with 10 nm Ni/50 nm Au films deposited by DC magnetron sputtering at 3×10^{-3} mbar. Metal lift-off is done by dipping the substrate in acetone for 20 min, followed by washing in IPA and N_2 drying.

Reflectance Measurement. For temperature-dependent microreflectance measurements, the samples are placed in a closed cycle He cryostat with an optical window placed above the sample. The samples are illuminated with a broadband

white LED source and response is collected through a $\times 50$ objective with a numerical aperture of 0.5 and analyzed using a spectrometer with a grating of 1800 lines/mm. The differential reflectance is then calculated via normalizing the signal obtained from stack with (R_{on}) and without bilayer WS_2 (R_{off}) layer as follows: $\frac{\Delta R}{R} = \frac{R_{\text{on}} - R_{\text{off}}}{R_{\text{off}}}$. To match the experimental data, apart from the A_{1s} exciton, we consider oscillators separated by 50 meV in the spectral range below the exciton while the higher energy side is fitted with oscillators placed at A_{2s} , A_{3s} , A_{4s} , A_{5s} , and B_{1s} energy positions.^{4,5}

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsphotonics.0c01159>.

Section S1, Stark shift calculation of the excitons in bilayer TMD; Section S2, Raman characterization of bilayer WS_2 ; Section S3, calculation of reflectance with transfer matrix method; Section S4, gate leakage current for sample S2; Section S5, field dependent differential reflectance for the device configuration S2-F; Section S6, temperature dependent reflectance spectra for sample S3; Section S7, gate dependent reflectance spectra for device configuration S2-D; and Section S8, optical images of the stacks (PDF)

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Notes

The authors declare no competing financial interest.

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