Understanding the Effects of Dielectric Property and Band Alignment on Interlayer Excitons in 2D van der Waals Heterostructures

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Abstract
Two dimensional (2D) van der Waals heterostructures from transition metal dichalcogenide (TMDC) semiconductors show a new class of spatially separate excitons with extraordinary properties. The interlayer excitons (X\textsubscript{I}) have been studied extensively, yet the mechanisms that modulate X\textsubscript{I} are still not well understood. Here, we introduce several organic-layer-embedded hybrid heterostructures, MoS\textsubscript{2}/organic/WSe\textsubscript{2}, to study the binding energy of X\textsubscript{I}. We discover that the dielectric screening of the quasi-particle is greatly enhanced with organic molecules having a large energy gap, resulting in a distinct blue shift in interlayer emission, and that the band alignment is also critical. When the organic layer provides a staggered energy state, interlayer charge transfer can transition from tunneling to band-assisted transport, further increasing X\textsubscript{I} emission energies due to a stronger dipolar interaction. The formation of X\textsubscript{I} may also be significantly suppressed using electron or hole trapping molecules. These findings should be useful in realizing X\textsubscript{I}-based optoelectronics.

Keywords: Interlayer exciton, van der Waals heterostructure, transition metal dichalcogenide, dielectric property, optoelectronics
Two-dimensional (2D) heterostructures built by stacking transition metal dichalcogenides (TMDCs) such as MoS\textsubscript{2} and WSe\textsubscript{2} have garnered much attention due to their extraordinary optoelectronic properties. The strong light-matter interaction in the heterostructures gives rise to interlayer excitons (X\textsubscript{I}) which are a bound state of spatially separate electrons and holes in opposite 2D layers.\textsuperscript{1} The enhanced Coulomb interaction in p-n hetero-stacks, including MoS\textsubscript{2}/WSe\textsubscript{2} and MoSe\textsubscript{2}/WSe\textsubscript{2},\textsuperscript{2,3} causes a large binding energy of X\textsubscript{I} around ~110 meV.\textsuperscript{4} The interlayer excitons have a long lifetime (~100 ns)\textsuperscript{5} and scalable diffusion-length (~a few μm)\textsuperscript{6}, which are suitable for light-emitting diodes,\textsuperscript{7} photodetectors,\textsuperscript{8} and photovoltaics\textsuperscript{9}. In addition, the separation of charges brings about permanent electric dipoles in the out-of-plane direction of the structure.\textsuperscript{10} It allows for electrical control of the interlayer excitons, which may not be possible with individual TMDC layers.\textsuperscript{11} The exceptional properties and controllability could be harnessed to realize emerging applications such as excitonic devices and valleytronics.\textsuperscript{12}

Tunneling of the photogenerated charges through the van der Waals gap between the stacked TMDC heterolayers is the mechanism of X\textsubscript{I} formation.\textsuperscript{13} Various mechanisms were investigated to modulate the band-penetrating transfer process, including doping\textsuperscript{13}, external electrical field\textsuperscript{14}, or mechanical strains\textsuperscript{15}. Despite the extensive studies, understanding the mechanisms that govern X\textsubscript{I} and the related emission still remains limited. For example, the dielectric property can change the binding energy of X\textsubscript{I} by disrupting the Coulombic interaction between the charges. Previous studies mostly focused on the dielectric property of the environment (e.g., various substrates),\textsuperscript{16-17} while few studies investigated that of the heterolayers.\textsuperscript{18-19} Modulating the dielectric properties of the heterostructures, for example, by inserting a layer between TMDC layers, may provide critical insights on X\textsubscript{I} and generate novel strategies for tuning the emission signatures. Further, such an approach could also help study additional mechanisms that can tailor the interlayer charge transfer process, thereby regulating the photoluminescence (PL). The new level of understanding would form the foundation for X\textsubscript{I}-based applications.

This work elucidates the dielectric property and the charge transfer mechanisms between the stacked heterolayers by embedding a uniform layer of organic molecules between them. The 2D MoS\textsubscript{2}/organic/WSe\textsubscript{2} hybrid heterostructures show distinct characteristics compared with MoS\textsubscript{2}/WSe\textsubscript{2}, and the overall properties depend strongly on the nature of the organic molecules. We discover that the dielectric screening of X\textsubscript{I} is enhanced with large-energy molecules such as 1,3-bis(3,5-dipyrid-3-ylphenyl)benzene (B3PyPB), leading to a strong blue shift in the interlayer emission. In addition to the screening, we find that the band alignment with the organic layer can also change the X\textsubscript{I} emission drastically. Eosin Y (EY) forms an energy state between MoS\textsubscript{2} and WSe\textsubscript{2}, thereby transitioning from tunneling to band-assisted transport. As a result, even higher energy emission is observed. Further, the interlayer emission may also be suppressed completely when electron or hole trapping layers are used including tetracyanoquinodimethane (TCNQ) and cobalt phthalocyanine (CoPc), respectively. These findings provide critical insights on the interlayer excitons and should be useful in designing optoelectronic devices from 2D TMDC materials.

To investigate the effects of distinct mechanisms on X\textsubscript{I}, we prepared the organic molecule embedded hybrid heterostructures with a sequential stacking method (see Materials and Methods for details).\textsuperscript{2,20} First, monolayer MoS\textsubscript{2} flakes were placed on a silicon (Si/SiO\textsubscript{2}) substrate with a mechanical exfoliation method.\textsuperscript{21} The MoS\textsubscript{2} samples were subsequently immersed in the dye-dissolved solutions to form organic layers on top of MoS\textsubscript{2} flakes. The organic molecules, B3PyPB, EY, TCNQ, and CoPc, form uniform layers with an almost identical thickness of ~0.9 nm (see Figure S1). Then, monolayer WSe\textsubscript{2} was transferred onto
the MoS$_2$/organic sample by polydimethylsiloxane (PDMS) stamping. MoS$_2$/WSe$_2$ heterolayers were also prepared as a control with the same method except for the solution-incubation step.

The irradiation from a HeNe laser at 633 nm generates interlayer excitons as well as excitons in individual MoS$_2$ and WSe$_2$ layers (Figure 1a). The staggered energy gap between MoS$_2$ and WSe$_2$ facilitates the dissociation of the excitons in each flake, and the photoexcited electrons in WSe$_2$ and holes in MoS$_2$ can transfer to the opposite layers via tunneling (Figure 1b). The recombination of the spatially separate electron-hole pair leads to the interlayer emission. The organic layer embedded between MoS$_2$/WSe$_2$ can affect the formation of the interlayer excitons and the relevant emission. For example, an organic layer will increase the distance between the TMDC layers and enhance the dielectric screening. As illustrated in Figure 1c, the tunneling will be dominant in MoS$_2$/B3PyPB/WSe$_2$ due to the large energy gap between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of B3PyPB. This energy gap (~3.7 eV) is substantially greater than those between conduction band minima (CBM) and valence band maxima (VBM) of both MoS$_2$ and WSe$_2$. Therefore, the dielectric screening will predominantly affect the interlayer excitons and the emission. In addition, the organic molecules may alter the band alignment, thereby causing different charge transfer pathways. EY has a LUMO situated between the CBM of MoS$_2$ and WSe$_2$. As a result, the EY layer will favor the band-assisted transport of photoexcited electrons, while the holes will transfer via tunneling (Figure 1d). Other mechanisms may also be explored such as trapping of the electrons from both TMDC layers by implementing molecules with a LUMO level lower than the MoS$_2$ CBM. TCNQ is such an exemplary molecule with its LUMO at approximately -2.8 eV. Similarly, one can design hole trapping by constructing a layer of molecules such as CoPc whose HOMO (~-6.5 eV) is higher than the VBM of both MoS$_2$ and WSe$_2$. It is worth noting that all four molecular layers have a uniform thickness, and thus the effect of the extended distance between MoS$_2$ and WSe$_2$ monolayers will be similar.

Figure 2 shows the effects of the enhanced dielectric screening and expanded gap in the B3PyPB-inserted MoS$_2$/WSe$_2$ heterostructure. The control MoS$_2$/WSe$_2$ sample exhibits two distinct peaks in the PL spectrum (Figure 2a). The spectral deconvolution with three Gaussian functions (see Figure S2) reveals the interlayer emission at ~801 nm as well as MoS$_2$ and WSe$_2$ PL at approximately 668 and 750 nm, respectively. It is notable that the interlayer emission is stronger than the intralayer PL, which may be attributed to the strong coupling of the spatially separate charges in the TMDC layers, as illustrated in Figure 2b. The sharp features at short wavelengths correspond to the Raman scattering of the heterostructure. In Figure 2c, MoS$_2$/B3PyPB/WSe$_2$ displays several distinct features as well as similar characteristics. Its intralayer PL peaks are measured at approximately 665 nm (MoS$_2$) and 746 nm (WSe$_2$) which are very close to those of the control sample. In contrast, the interlayer emission appears as a shoulder at ~776 nm which is significantly blue-shifted from the control by ~25 nm (or ~50 meV). The peak intensity of interlayer emission significantly drops, while that of WSe$_2$ PL increases by order of magnitude.

To understand the observed phenomena, we used a simple hydrogen-like particle model to define the energy state of $X_1$ (see the SI for details). The Hamiltonian ($H_{ex}$) of the quasiparticle may be expressed as:

$$H_{ex} = -\frac{\hbar}{2m_{ex}} \frac{1}{\rho} \frac{d}{d\rho} \left( \rho \frac{d}{d\rho} \right) - \frac{e^2}{\epsilon_{eff} \sqrt{\rho^2 + r^2}}$$

where $\rho$ is the relative coordinate, $e$ is an electrical charge, and $m_{ex}$ denotes the effective mass.
of $X_1$ as $m_{ex}^{-1} = m_e^{-1} + m_h^{-1}$, where $m_e$ and $m_h$ are the mass of electron and hole, respectively, and $h$ is Planck constant. $l$ is the separation length between the charges, and $\varepsilon_{eff}$ indicates the effective dielectric function of the hetero-layers. The eigenvalue of $H_{ex}$, that is, the binding energy $E_b$ of $X_1$, is closely related to the shift of the interlayer emission, as the inserted organic layer changes both $l$ and $\varepsilon_{eff}$. To estimate $\varepsilon_{eff}$, use a simple dielectric model where each layer is considered as the piecewise dielectric as shown in Figure 2e. Here, the organic layer and van der Waals gap are the intermediate layers of the hybrid structure and the control, respectively. The calculated dielectric function of MoS$_2$/organic/WSe$_2$ is $\varepsilon_{eff} = \sim 6.8$, which is greater than that of the control (~5.4). The dielectric function can yield the Bohr radius of $X_1$ ($a = \varepsilon_{eff} h/m_{ex} e^2$): approximately 1.47 nm for the hybrid and 1.17 nm for the control. The X$_1$ binding energy can then be expressed as a function of the separation distance and Bohr radius or the distance ratio ($x = l/a$), as shown in Figure 2f. With knowledge of the distance and Bohr radius in each sample, we estimate X$_1$ binding energies of the hybrid structure (~51 meV) and the control (~101 meV). The reduction of $E_b$ (~50 meV) in the MoS$_2$/B3PyPB/WSe$_2$ is in excellent agreement with the observed blue shift of X$_1$ emission (~50 meV). The results strongly suggest that the dielectric screening by the organic layer has a central role in the blue-shift of X$_1$ emission.

Next, we explored EY whose LUMO level is between CBM of WSe$_2$ and MoS$_2$. Figure 3a shows the PL spectrum of the MoS$_2$/EY/WSe$_2$ heterostructure. Three distinct PL signatures are observed. The intensity and peak positions of MoS$_2$ (~664 nm) and WSe$_2$ (~746 nm) are nearly identical with those from MoS$_2$/B3PyPB/WSe$_2$. The interlayer emission, however, is measured at around 764 nm, which is blue-shifted from MoS$_2$/B3PyPB/WSe$_2$ by ~12 nm (or ~25 meV). The emission intensity is also stronger by a factor of two. This observation suggests that besides the dielectric screening, other mechanisms may impact the energy state of X$_1$ in the EY-inserted heterostructure. Unlike B3PyPB, the EY layer may favor the band-assisted transfer of photoexcited electrons from WSe$_2$ to MoS$_2$ due to the staggered band alignment (Figure 3b). As a result, a greater amount of electrons will be transferred in the EY-embedded hybrid structure. Further, the energy barrier between MoS$_2$ VBM and EY HOMO is much smaller than that between MoS$_2$ VBM and B3PyPB HOMO, thus promoting more hole transfer. With the enhanced charge transfer, MoS$_2$/EY/WSe$_2$ may have a greater population of electrons in MoS$_2$ and holes in WSe$_2$. The EY-inserted heterostructure will thus have a stronger dipole-dipole interaction than the B3PyPB-embedded sample. We attribute this strong dipolar interaction to the origin of the additional blue-shift observed in the MoS$_2$/EY/WSe$_2$ structure.

The interaction energy ($U$) originating from the dipole-dipole interaction may be expressed as:

$$U = -\frac{e^2}{\varepsilon_{eff} \sqrt{\rho^2 + l^2}} + \frac{2e^2}{\varepsilon_{eff} \rho}. \quad (3)$$

where the first term represents the attractive interaction inside the X$_1$ and the second term stands for the repulsive interaction arising from the dipoles. From the mean-field approximation where the excitons follow the Boltzmann distribution, the average interaction energy of the dipoles ($E_{dip}$) may be evaluated as:

$$E_{dip} = n \int U(\rho) g(\rho) d^2 \rho \quad (4)$$

where $n$ is the dipole density. $g(\rho)$ represents the density distribution of the pair correlation: $g(\rho) = \exp[-U(\rho)/k_B T]$, where $T$ is the temperature and $k_B$ is the Boltzmann constant. The difference in the interaction energy ($\Delta E_{dip}$) responsible for the PL blue-shift may be written as:

$$\Delta E_{dip} = \int [U(\rho)_{EY} - U(\rho)_{control}] g(\rho) d^2 \rho,$$
\[ \Delta E_{\text{dip}} = \frac{4\pi e^2 l}{\varepsilon_{\text{eff}}} D \left( \frac{\varepsilon_{\text{eff}} l k_B T}{e^2} = z \right) \Delta n \] (5)

where \( D \) is a bias function with respect to a dimensionless parameter \( z \) which is a ratio between thermal and electrostatic energies of the charges (\( z \sim 0.13 \), in our system). For \( z < 1 \), \( D \) is reduced to a form \( D = \frac{\Gamma(4/3)}{2} z^{1/3} \), where \( \Gamma \) is a gamma function.\(^{37}\)

The model suggests that \( \Delta E_{\text{dip}} \) has a linear relationship with \( \Delta n \), from which we obtain \( \Delta n \sim 3.1 \times 10^{12} \text{ cm}^{-2} \). This is the density difference of interlayer excitons between EY- and B3PyPB-embedded heterostructures (filled red circle in Figure 3d). The \( X_1 \) energy shift due to the density difference is supported by previous studies. For example, electrical gating can change the \( X_1 \) population and demonstrate a similar blue-shift in the interlayer emission (~20 meV).\(^{14}\)

Laser irradiation also shows a similar correlation between \( X_1 \) population and emission energy in 2D heterostructures.\(^{39}\) The results strongly support that the interlayer emission can be tailored by not only dielectric screening, but also dipolar interaction due to the change in the \( X_1 \) population with the insertion of an organic layer in the TDMC heterolayers.

We also studied the trapping of photoexcited electrons and holes from the heterostructures, including TCNQ and CoPc layers. Figures 4a and 4b show the PL spectra of MoS\(_2\)/TCNQ/WSe\(_2\) and MoS\(_2\)/CoPc/WSe\(_2\). The two spectra appear nearly identical, displaying emission peaks at ~666 and ~746 nm corresponding to MoS\(_2\) and WSe\(_2\), respectively. These spectra are also very similar with those from other heterostructures in Figures 2 and 3. The intralayer excitons may recombine at the valleys of each TMDC layer, where the band energies of one layer are not significantly hybridized with the other layers. Therefore, we conclude that the intralayer emissions from the constituent layers are not significantly affected by the organic layers, which is consistent with our previous study.\(^2\) In contrast, the interlayer emission is not observed from either sample.

Figures 4c and 4d respectively present the energy band diagrams of the TCNQ- and CoPc-inserted heterostructures, illustrating the quenching mechanisms for interlayer emission. The TCNQ LUMO is located lower than both MoS\(_2\) and WSe\(_2\) CBM. In the same manner, its HOMO is placed below MoS\(_2\) and WSe\(_2\) VBM, thus forming a discontinuously staggered gap at the interfaces. Due to the alignment, the photoexcited electrons from MoS\(_2\) and WSe\(_2\) may be trapped in the TCNQ layer. The trapped charges in TCNQ may not be further transferred to other TMDC layers, because there is no energy state in MoS\(_2\) and WSe\(_2\) near the TCNQ LUMO. As a result, the interlayer exciton formation is suppressed. The CoPc-embedded heterostructure has an opposite band alignment, but displays the same PL characteristics. The LUMO and HOMO levels of CoPc are higher than CBM and VBM of both MoS\(_2\) and WSe\(_2\), respectively. The alignment now favors trapping of holes and prohibits additional hole transfer from CoPC to MoS\(_2\) or WSe\(_2\). Therefore, the interlayer excitons may be not formed in MoS\(_2\)/CoPc/WSe\(_2\), thus no related emission is observed.

Finally, it is worth noting that the embedded organic layers do not support interlayer excitons at the organic/TMDC interfaces, as the hybrid structures in this work do not show any relevant emission. This strongly suggests that the Coulomb interaction between the charges separately residing in TMDC and organic layers may not be as strong as that in TMDC/TMDC interfaces. Furthermore, the interlayer emission requires both energy and momentum conservation. The lack of PL emission led us to conclude that the organic layers may not compensate the momentum mismatch with TMDC layers due to the relatively weak coupling between organic and TMDC layers compared to the strong interaction at the MoS\(_2\)/WSe\(_2\) interface.\(^2\)
In closing, we have studied the effects of dielectric properties and band alignments on $X_I$ by inserting various organic layers in TMDC heterostructures. The organic layers reduce the $X_I$ binding energy, which leads to a strong blue shift of the interlayer emission. Depending on the types of the energy level alignment, $X_I$ emission can be further blue-shifted by the dipolar interaction, or completely quenched due to the trapping of charges. This approach may be further developed to provide additional controllability on interlayer excitons. For example, the inserted layer may be modified to have periodic patterns by using lithographic techniques. Such platforms may allow for a study of the localized Coulombic interaction in van der Waals heterostructures and the physics of exciton-polariton condensates. Furthermore, environmentally sensitive molecules could be used to tailor interlayer exciton properties by external stimuli, including electro-, chemo-, and photo-chromic molecules. The effort could lead to a new class of $X_I$-based devices with externally tunable characteristics.

**Materials and Methods**

**Sample fabrication.** Monolayer MoS$_2$ flakes were mechanically exfoliated from bulk crystals (SPI Supplies) and deposited on a p-doped Si/SiO$_2$ substrate. The substrate was ultrasonicated in various solvents, including acetone, methanol, and deionized water (DI) for 30 min each. Then, it was dried by blowing air and placed on a hot plate at 110 °C for 2 min. To fabricate the organic layers on top of the MoS$_2$ flakes, the samples were immersed in the solutions containing the dye molecules at room temperature for 8 h: B3PyPB (1 mg/ml in chloroform), EY (1 mg/ml in ethanol), TCNQ (0.6 mg/ml in chloroform), and CoPc (0.4 mg/ml in dimethylformamide). These molecules are purchased from Sigma-Aldrich in a powder form. The organic-layer/MoS$_2$ samples were subsequently washed with each solvent of the molecules and DI to remove the loosely bound particles. For example, B3PyPB deposited sample was rinsed with the excess amount of chloroform and then cleaned with DI water.

Top WSe$_2$ layer was stacked on the prepared organic/MoS$_2$ sample by a polymer-stamping method. WSe$_2$ flakes were first mechanically exfoliated on a dimethylpolysiloxane (PDMS, purchased from Krayden) stamp. The stamp was fabricated by mixing the base and curing agent of PDMS with 12:1 ratio and cured at 65 °C for 12 h. The stamp with a monolayer of WSe$_2$ was placed on a glass substrate which is connected to a micro-positioner (Signatone). Then, the WSe$_2$ flake was positioned above the MoS$_2$/organic layer and brought into contact. The elastomer maintained the position for 30 min at 50 °C and lifted slowly by manipulating the positioner. Finally, the samples were annealed under argon gas environment at 280 °C for 2 h for a better contact at the interface of the heterolayers.

**Sample characterization.** The optical and Raman measurements were performed using a Renishaw confocal microscope under ambient conditions. A 633-nm HeNe laser at ~0.1 mW was used to irradiate samples through a 100× objective lens. The optical signals of the samples were transferred by the same objective and collected by a charge-coupled device (CCD) camera. The layer numbers of TMDC flakes were first inspected by optical microscopy, and confirmed by the height measurement with atomic force microscopy (AFM). A Bruker Dimension Icon AFM was used with a SCANASIST-AIR probe under ambient conditions. To avoid any unwanted effects of the moisture, we baked the samples in the same manner described above before the measurement. The pixel window of 256 × 256 was maintained, and the actual size of the window was adjusted depending on the sizes of samples. The raster-scanning was conducted sequentially after each step of stacking to monitor the thickness of each layer in the heterostructure.

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Figure 1. (a) Schematic of a MoS$_2$/WSe$_2$ heterostructure. Laser irradiation creates $X_1$ as well as excitons in individual MoS$_2$ and WSe$_2$ layers. The Coulomb interaction between the spatially separate electron-hole pair forms a strong bound state as the interlayer exciton. (b) Mechanism for interlayer emission. The photoexcited electron in WSe$_2$ and hole in MoS$_2$ transfer to the opposite layers via tunneling facilitated by the staggered band alignment. Schematics of the organic layer embedded hybrid heterostructures: (c) MoS$_2$/B3PyPB/WSe$_2$ and (d) MoS$_2$/EY/WSe$_2$. The photoinduced charges in the B3PyPB-embedded heterostructure tunnel through the large-energy B3PyPB layer, while the electron transfer via EY is energetically favorable in MoS$_2$/EY/WSe$_2$. 
Figure 2. (a) PL spectrum of MoS$_2$/WSe$_2$ with a HeNe laser excitation at 633 nm. The deconvolution suggests interlayer emission at ~801 nm as well as PL peaks at ~668 nm (MoS$_2$) and ~750 nm (WSe$_2$). (b) Energy diagram of MoS$_2$/WSe$_2$ and X$_1$ formation in via tunneling of photoinduced charges. (c) PL spectrum of MoS$_2$/B3PyPB/WSe$_2$ showing the interlayer emission with much reduced intensity and a strong blue-shift. (d) Energy diagram and schematic of B3PyPB-embedded MoS$_2$/WSe$_2$. The organic layer increases the tunneling length and the dielectric screening, resulting in the 25-nm PL shift. (e) Dielectric model of TMDC heterostructure (left) and hybrid structure (right). Each structure consists of five layers with distinct dielectric constants. (f) Theoretical X$_1$ binding energies of MoS$_2$/B3PyPB/WSe$_2$ (blue line) and MoS$_2$/WSe$_2$ (black line) as a function of distance ratio $x$. The estimated binding energy difference between the samples (filled black and blue circles, ~50 meV) corresponds to the interlayer emission shift observed with the hybrid heterostructure.
Figure 3. (a) PL spectrum and (b) energy band diagram of MoS$_2$/EY/WSe$_2$ heterostructure. Compared to the B3PyPB-inserted sample, the interlayer emission is stronger and blue-shifted while intralayer PL signatures are nearly identical. The EY layer facilitates photoinduced electron transfer from WSe$_2$ to MoS$_2$ by providing an energy state between their CBM. The energetically favorable band-assisted transfer will thus lead to greater amounts of electrons and holes (i.e., out-of-plane dipoles) than the B3PyPB-embedded heterostructure. (c) A plot of dipolar interaction energy versus the dipole density. The interaction energy linearly increases with the dipole density.
Figure 4. PL spectra of hybrid heterostructures including a layer of (a) TCNQ and (b) CoPc between TMDCs. The intralayer PL spectra are nearly identical, whereas the interlayer emission is not observed from both hybrid heterostructures. Energy band diagrams of (c) MoS$_2$/TCNQ/WSe$_2$, favoring electron trapping from both TMDCs, and (d) MoS$_2$/CoPc/WSe$_2$, supporting the hole trapping. Both trapping processes induce the complete quenching of X$_1$ emission.
Supporting Information

Understanding the Effects of Dielectric Property and Band Alignment on Interlayer Excitons in 2D van der Waals Heterostructures

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1. AFM Imaging and Height Profiles

Figure S1. Raster-scanned AFM images and height profiles of (a) MoS$_2$/WSe$_2$ (control), (b) B3PyPB-, (c) EY-, (d) TCNQ-, and (e) CoPc-embedded hybrid-structures, where the organic layers are sandwiched by the TMDC flakes. The height profiles are measured along the arrow lines in the height map. The height of individual TMDC layer is about 0.8 nm. The thicknesses of all the organic layers are all nearly uniform at ~0.9 nm as determined by the height difference between TMDC and TMDC/organic.
2. Deconvolution of PL Spectra

Figure S2. PL spectra of (a) MoS\textsubscript{2}/WSe\textsubscript{2} (control), (b) B3PyPB-, (c) EY-, (d) TCNQ-, and (e) CoPc-embedded heterostructures. Three Gaussian profiles are used to deconvolute each PL spectrum: MoS\textsubscript{2} (cyan) and WSe\textsubscript{2} (red), and interlayer emission (dark cyan). The wavelengths of the deconvoluted emission peaks are noted as insets in the figure.
3. Solution of 2D Hydrogen-like Schrödinger Problem

An exciton is a quasi-particle formed as a bound state of photoexcited electron and hole, similar to the hydrogen atom with the electrostatic interaction between the constituent charges. From the Wannier equation, the energy of the particle can be expressed as a sum of the kinetic energy from the relative motion of the charges and the electrostatic interaction within the particle. The eigenvalue of the energy (i.e., exciton binding energy or $E_b$) can be obtained by solving a Schrödinger equation of the hydrogen-like particle:

$$\frac{-\hbar^2}{2m_{ex}} \frac{d^2 \phi(r)}{dr^2} - \frac{e^2}{\varepsilon_{ex} \rho^2 + l^2} \phi(r, l) = E_b \phi(r, l).$$

(1)

where $\hbar$ is the reduced Planck constant ($\hbar = h/2\pi$), $e$ is the electrical charge, $\rho$ is the relative coordinate, $m_{ex}$ denotes the effective mass of the exciton, and $l$ is the separation distance between photoexcited electron and hole residing in MoS$_2$ and WSe$_2$, respectively. $\varepsilon_{ex}$ is the dielectric constant of the exciton. $\phi$ and $E_b$ are the eigenfunction and eigenvalue, respectively. To simplify the equation, we define the radius of the interlayer exciton as $a = \varepsilon_{ex} \hbar / m_{ex} e^2$ and $E_{b0}$ as the binding energy of X$_I$ without the separation given by $l$: $E_{b0} = m_{ex} e^4 / 2 \varepsilon_{ex} h^2$. The charge separation may be determined by considering the thicknesses of TMDC layers and the intermediate layer. In the control sample, the intermediate layer is set to be a van der Waals gap (~0.35 nm), thus the separation distance is $l = ~1.15$ nm. The hybrid structure has the intermediate organic layer (~0.9 nm) and $l = ~2.05$ nm.

From the effective dielectric constant of each structure (see Section 4 below), we obtain $E_{b0} = ~72$ meV for the hybrid structure and $E_{b0} = ~114$ meV for the control (MoS$_2$/WSe$_2$), respectively. The $E_{b0}$ value of the control sample is in excellent agreement with previous reports. From the relationship, the equation (1) may be converted into a dimensionless form:

$$-\frac{1}{y^2} \frac{d}{dy} \left( y^2 \frac{d\psi}{dy} \right) - \frac{2}{\sqrt{y^2 + x^2}} \psi = w(x) \psi$$

(2)

where $y = \rho/a$, $x = l/a$, $w(x) = E_b/E_{b0}$, and $\psi = \phi/a$. The solution of the differential equation can be reduced to the function derived from the curve-fitting method reported by Leavitt et al.:

$$w(x) = \frac{4 + 12.97x + 0.718x^2}{1 + 9.65x + 0.24x^2 + 0.3706x^3}$$

(3)

Figure S3 displays the binding energy of the interlayer exciton as a function of the distance ratio $x$. $E_b$ is inversely proportional to $x$. As the ratio increases, the exciton binding energy monotonically drops. It implies that with a larger separation gap, the overlap between spatially separate electron and hole becomes weaker, thereby resulting in a smaller binding energy.

![Figure S3](image-url)

**Figure S3.** Theoretical prediction of the interlayer exciton binding energies in MoS$_2$/WSe$_2$ (black) and B3PyPB-inserted heterostructure (blue) as a function of distance ratio $x$. The X$_I$ binding energy decreases monotonically with increasing distance ratio.
4. Dielectric Model
The Coulomb interaction between the charges in X₁ highly depends on the dielectric screening of the quasi-particle. To account for the dielectric properties within the heterostructure, a dielectric model is used with a multi-layer Poisson’s equation. As illustrated in Figure 2c, five distinct layers of piecewise dielectrics are assumed, with εᵢ representing the relative dielectric constant of the iᵗʰ layer of the heterostructure. The bottom layer SiO₂ has a dielectric constant of ε₁ ~ 3.9, while the top layer which is air has ε₅ ~ 1. Both TMDC layers have similar dielectric properties (ε₂ ~ 15.1 for MoS₂ and ε₄ ~ 15.1 for WSe₂). The intermediate organic layer between TMDCs has a dielectric constant of ε₃ ~ 3.5, while the control has a small air gap with ε₃ ~ 1. For charges in Z and z’ planes, the electrostatic potential (Φ) with in-plane coordinate ρ can be expressed as:

\[-V(\rho, z, z') = \frac{1}{4\pi^2} \int \Phi(z, z', k)e^{ik\rho}d^2k, \tag{4}\]

where the potential V satisfies the Poisson’s equation and k is the inverse of the distance of the charges.

\[4\pi\delta(z - z') = [k^2\epsilon(z, k) - \frac{\partial}{\partial z}\epsilon(z, k)\frac{\partial}{\partial z}]\Phi(z, z', k). \tag{5}\]

As the charges of X₁ are confined in TMDC layers, the potential (Φₜₐₘ₅₈) from X₁₅ has a relationship:

\[\Phiₜₐₘ₅₈(z, z', k) = \epsilon_{eff}\Phi(z, z', k). \tag{6}\]

From the five-layer model, εeff may be derived as reported by Chen et al.:

\[\epsilon_{eff} = \epsilon₂\epsilon₀(1 + \epsilon'_1\epsilon'_2a^2 + \epsilon'_1\epsilon'_3\alpha^2\beta^2 + \epsilon'_1\epsilon'_4\alpha^4\beta^2 + \epsilon'_1\epsilon'_5\beta^2 + \epsilon'_2\epsilon'_4a^2\beta^2 + \epsilon'_2\epsilon'_5\beta^2 - \epsilon'_2\epsilon'_5\beta^2 - \epsilon'_2\epsilon'_5\beta^2 - \epsilon'_2\epsilon'_5\beta^2 - \epsilon'_2\epsilon'_5\beta^2 + \epsilon'_2\epsilon'_5\beta^2 + \epsilon'_2\epsilon'_5\beta^2), \tag{7}\]

where ε₀ stands for the electrical permittivity in vacuum. ε’ᵢ represents iᵗʰ layer of the heterostructure. α = exp(-tₐₘ₅₈/q) and β = exp(-tᵢₐₑᵢ/q), where q is the inverse of Bohr radius of the exciton. tₐₘ₅₈ (~0.8 nm) and tᵢₑᵢ represent the thicknesses of TMDC and intermediate layers, respectively. Note that the air gap of tᵢₑᵢ ~ ~0.35 nm is used the control sample (MoS₂/WSe₂), while the intermediate layer in the hybrid structure is the organic layer with a thickness of ~0.9 nm. Using these values, we estimate the overall effective dielectric constants of the hybrid structure (εeff = 6.8) and the control (εeff = 5.4). As a result, the organic layer embedded hybrid heterostructure has a greater extent of dielectric screening on X₁₅ than MoS₂/WSe₂.
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