Manipulating Interlayer Excitons for Ultra-pure Quantum Light Generation

Huan Zhao
Center for Integrated Nanotechnologies, Materials Physics and Application Division, Los Alamos National Laboratory https://orcid.org/0000-0002-4982-0865

Linghan Zhu
Department of Physics, Washington University in St. Louis

Xiangzhi Li
Center for Integrated Nanotechnologies, Materials Physics and Application Division, Los Alamos National Laboratory

Vigneshwaran Chandrasekaran
Center for Integrated Nanotechnologies, Materials Physics and Application Division, Los Alamos National Laboratory

John Baldwin
Los Alamos National Laboratory

Michael Pettes
Center for Integrated Nanotechnologies, Materials Physics and Application Division, Los Alamos National Laboratory

Andrei Piryatinski
Theoretical Division, Los Alamos National Laboratory

Li Yang
Washington University in St. Louis https://orcid.org/0000-0002-8611-6359

Han Htoon (htoon@lanl.gov)
Center for Integrated Nanotechnologies, Materials Physics and Application Division, Los Alamos National Laboratory https://orcid.org/0000-0003-3696-2896

Research Article

Keywords:

Posted Date: April 26th, 2022

DOI: https://doi.org/10.21203/rs.3.rs-1538546/v1

Abstract

Interlayer excitons (IXs) formed at the interface of two different atomically-thin semiconductors have been emerging as an exciting ground not only for exploring fascinating many-body phenomena such as exciton condensation,1-4 but also for realizing exciton-based information processing technologies.5, 6 In a parallel development, nanoscale strain engineering has emerged as an effective means for the localization of 2D intra-layer excitons and activation of defect states for quantum light generation.7-11 Exploring the intersection of these two exciting areas, where strain and defects are exploited for the manipulation of IX toward the emergence of new functionalities, is currently at a nascent stage.6 Here, using MoS2/WSe2 heterostructure as a model system, we demonstrate how strain, defects, and layering can be utilized as control knobs toward novel defect-bound IXs capable of bright, robust, and tunable quantum light emission in the technologically important near-infrared spectral range. More significantly, because the deep-level sulfur vacancy states isolate our quantum emitters (QEs) from any of the intra- and inter-layer excitons, we were able to achieve ultra-high single-photon purity with g(2)(0) = 0.01 meeting the critical milestone for quantum key distribution (QKD), logic gate and memory technologies.12 Our strategy of creating site-controlled QEs from the defect-bound IXs represents a paradigm shift in 2D quantum photonics research, from engineering intralayer exciton in monolayer structures towards IXs at the interface of 2D heterostructures.

Full Text

Heterobilayers (HBLs) of transition metal dichalcogenides (TMDCs) with staggered type–II band alignment are particularly fascinating as they allow the formation of charge-separated interlayer excitons (IXs), where electrons and holes have their energy extrema in opposite layers.[1-4, 13] Formation of such IXs gives rise to photoluminescence (PL) emissions at the lower energy side. In most hetero-bilayers such as $MoSe_2/WSe_2$, momentum-direct IXs locates at K valleys of constituent layers (K-K IX, Figure 1a) dominate the optical emission when the inter-layer twist angles are small.[13-16] Spatial charge separation in such IXs results in significantly reduced oscillator strength and an inefficient PL emission.[3] On the other hand, in certain heterobilayers such as MoS_2/WSe_2 and WS_2/MoS_2, strong interlayer hybridization at the G valley leads to G-K transition with enhanced oscillator strength (Figure 1b).[17-23] However, bright PL emission is still hindered by its momentum-indirect nature.

While both momentum-direct K-K IX (~ 1.0 eV)[24, 25] and momentum-indirect Γ-K IX (~ 1.57 eV)[17] have been investigated for MoS_2/WSe_2 HBLs, effects of strain field and in-gap defect states on complex IX transitions remain to be understood. Particularly, by band structure engineering, an electron residing in the sulfur-vacancy state ~200 meV below MoS_2 conduction band edge[26] could be utilized to form optically active excitonic transition with a hole in the hybridized G band (Figure 1c). This G-defect IX retains large oscillator strength of the hybridized Γ IX species while bypassing stringent momentum conservation limitations in traditional K-K IXs which require matched lattice constants and perfect rotational alignment among the two constituent layers. Owing to deep trapping potential, such defect-bound IXs could enable

high purity quantum light generation in technologically important near-infrared (NIR) spectral range and raise operating temperature out of cryogenic regime.

To explore this potential, we fabricate nanopillars made of dielectric material, with the diameter of 150 nm and height of 200 nm, directly on a gold substrate. We then prepared three categories of heterostructures on top of the nanopillars/gold substrate: (i) MoS_2 /WSe_2 HBLs (also called HBLs for short, illustrated in Figure 1d); (ii) MoS_2 /WSe_2 HBL on top of a 5 nm-hBN layer (HBL/hBN, illustrated in Figure 1e); (iii) bilayer-MoS_2/ monolayer-WSe_2 (also called hetero-trilayer, or HTL, illustrated in Figure 1f). Here nanopillars not only induce local strain within MoS_2/WSe_2 heterostructures but are also prone to create point defects via structural damage.[9] The hBN layer in sample category (ii) isolates the HBL from gold-induced fluorescence quenching and serves as a protective layer that shields the HBL from structural damages or substrate-induced adsorbates - hence defect generation - during the stamping process onto nanopillars. Since the G valley features an out-of-plane orbital character, its orbital hybridization is sensitive to interlayer interactions.[20, 27, 28] For this reason, we stacked MoS_2/WSe_2 heterostructures with near-zero or near-60^o twist angle with ±5^o error (unless otherwise specified) to achieve small layer separation[17] and strong interlayer interactions.[29, 30] Through Raman mapping, an average strain level of 0.27% was extracted from the Raman difference between the homogeneous HBL and the HBL on nanopillars (Supplementary Note 2). We developed a modified mechanical exfoliation method to efficiently create and transfer large (~ 100) heterostructures (Supplementary Note 1).

Under a widefield illumination at 750 nm wavelength, low-temperature PL images of the heterostructures were acquired by an InGaAs detector (Figure 1h & Supplementary Note 3) and revealed that NIR emitters are created at point-of-contact region in HBL/nanopillars. Note that we use a gold substrate to quench the PL emission from the homogeneous HBL region and thus confine the IX emission at nanopillar sites. When switching excitation wavelengths from 750 nm to 660 nm (Figure 1i), the HBL/hBN region (Figure 1e and white box in Figure 1g) becomes remarkably brighter while the HBL/nanopillars region (Figure 1d and brown box in Figure 1g) significantly dims (explained in detail later). Next, we spectrally resolve the HBL/nanopillar emitters and find that the 1.0 eV broad PL peak of K-K IX commonly reported in MoS_2/WSe_2 HBLs is replaced by narrow PL lines emerging mainly in the 1.35±0.08 eV range (Figure 1j, 1k), corresponding to a redshift distributed between 140 and 300 meV from the G-K IX emission energy at 1.57 eV.[17] Since this energy redshift is in agreement with that of the sulfur-vacancy-induced PL redshift observed in MoS_2 monolayers,[26, 31] we attribute the transition to defect-bound, G-band IX state (Γ-defect IX) formed between an electron trapped in the sulfur defect levels and a hole in the hybridized G band of the HBL (the case in Figure 1c). For HTL/ nanopillars (Figure 1f), the emission energy is further redshifted to 1.26±0.1 eV range without significant reduction in PL intensity, advocating that we can utilize layer engineering as a tuning knob to control the Γ-defect IX emission range. (See Supplementary Note 4 for more statistics and PL spectra).

Interestingly in HBL/hBN region, where the hBN serves as a shielding layer minimizing defect generation and gold-induced PL quenching, a complete suppression of the Γ-defect IX emission and recovery of the

delocalized K-K IX transition at 1.0 eV is observed. Due to nanopillar strain, this emission exhibits a redshift and spectral broadening compared to that observed within unstrained regions (Figure 1j). This is consistent with recent studies reporting narrow linewidth localized emissions which cannot appear solely due to strain effect.[9, 32] We also examined an HBL/nanopillar sample with 22° twist angle and found that the emission is significantly dimmer showing broader linewidth (Supplementary Note 5). These findings provide evidence that control of twist angle, layer structure, defect concentration, and excitation wavelength can be utilized along with the nanoscale strain engineering to achieve localization and activation of various IX transitions (see Supplementary Table 1 for a summary).

To demonstrate that the Γ-defect IX exciton states can act as quantum emitters (QEs), we performed time-tagged, time-correlated photon counting, and the Hanbury–Brown–Twiss (HBT) experiments at 10 K. Figure 2a shows the PL spectrum of a typical emitter, of which the spectrally filtered emission is guided into a two-channel single-photon detector to obtain time-resolved PL (TRPL) curve (Figure 2b), time-dependent PL counts (Figure 2c), and second-order photon correlation ($g^{(2)}$) trace (Figure 2d). Measured TRPL shows lifetime of 18.45±0.01 ns. Interestingly, the first few nanoseconds of the PL time-evolution features slow uprising from initial non-zero value. This is distinctly different from the TRPL curves of the K-K IX (Supplementary Note 7) showing monotonous decay, implying a delay between optical absorption and photocarrier recombination. The stability of the emission is furthered confirmed by time-dependent PL counting. The $g^{(2)}$ trace showing almost complete photon antibunching with $g^{(2)}(0) = 0.01$ (our experimental sensitivity limit) provides unequivocal confirmation of the single-photon nature of our Γ-defect IX. We note that the purity of this single-photon emission is significantly higher than that recently reported in moire-trapped excitons ($g^{(2)}(0) \sim 0.28$)[6] (see Supplementary Note 8 for a discussion of moire IX and the IX reported in our work). Such an ultra-high single photon purity now meets the strict requirement for implementation of QKD, logic gates, and memory technologies.[12] We attribute this dramatic improvement of single-photon purity to the fact that our QE is in complete spectral isolation from any of the intra- and inter-layer excitonic transitions and the gold substrate effectively quenches the emission of the HBL except directly on top of the dielectric nanopillars. HBT experiment performed on a different emitter at 77 K reveals that the single-photon emission can sustain up to liquid nitrogen temperature (Figure 2e). The temperature-dependent PL experiment shows that while the PL lines get broader at elevated temperatures, the reduction of the integrated PL intensity is essentially insignificant (Figure 2f).

Next, to confirm the G-defect IX states as physical origin of our ultra-pure QEs, we performed first-principles calculations on the electronic structures of the MoS_2/WSe_2 heterostructures. Compared with intrinsic HBL (Figure 3a), the band structure of strained HBL (Figure 3b) shows that strain brings the G band closer in energy to the WSe_2 K valence band, facilitating the WSe_2 hole transfer. When a sulfur vacancy is introduced, defect bands emerge below the conduction band minimum (CBM), in qualitative agreement with the red-shift of G-defect IXs (Figure 3c). The introduction of Se vacancy, on the other hand, results in defect levels above the CBM, leaving the low-energy optical transitions unchanged (Supplementary Note 9). While the G-band is highly hybridized and has a sizable amount of MoS_2

component, the VBM at K only has contribution from the WSe_2 layer. This fact, together with the confinement of electron in the S vacancy level, allows G-defect IX transition to dominate over K-defect IX transition. Once an hBN layer is inserted between the HBL and nanopillars, defect formation in MoS_2 layer is averted as evidenced by the clean PL spectrum of the hBN encapsulated MoS_2 (Figure 3e), and thus the K-K IX emission in the HBL is recovered. The room temperature PL measurements further confirm that the G-K emission in homogenous HBL is replaced by the G-defect emission in HBL samples on nanopillars (Figure 3f). Compared to HBL, the calculation for a HTL composed of bilayer MoS_2 and monolayer WSe_2 reveals a reduced band gap and an increased MoS_2 component at G band maximum (Figure 3d), leading to further redshifted NIR emission of defect G-defect IX (Figure 1k and Supplementary Figure 5b) and enhanced MoS_2-branch PLE features (Supplementary Note 10).

Finally, to understand strong influence of excitation wavelength on the competition between K-K and G-defect IX emissions, we conducted PL excitation (PLE) experiments on three types of samples: (i) HBL/hBN/nanopillar sample for probing K-K IX; (ii) HBL/nanopillar and (iii) HTL/nanopillar samples for probing G-defect IXs. For K-K IX emission from HBL/hBN on nanopillars, we observed enhanced PL intensity when the excitation is in resonance with delocalized 2D intralayer excitons (i.e., WSe_2 exciton at 1.75 eV and MoS_2 A & B excitons at 1.88 eV and 2.01 eV, respectively) (Figure 4a), evidencing that the K-K IX is primarily formed by nonlocal interband absorption in constituent layers followed by interlayer charge transfer (Figure 4c). We also observed an absorption peak at 1.66 eV, which corresponds to the visible spectral range PL emission of HBL on nanopillars stemming from the interplay of strain-induced bandgap narrowing and strain-activated dark exciton emission in the WSe_2 layer (Figure 4e). In stark contrast, the G-defect IX emitting at 1.34 eV from HBL/nanopillar shows a series of PLE peaks near the band edges of MoS_2 and WSe_2, predominantly in the 1.5 to 1.7 eV range (Figure 4b and Supplementary Figure 11a). Considering their resemblance to the emissions from WSe_2 localized excitons, we attributed the PLE features in 1.5 to 1.7 eV range to localized transitions that originate from the interband absorption of defect states in WSe_2 layer amid local strain (Figure 4d). Such absorption patterns dominated by localized optical transitions were also observed in HTL/nanopillar samples with increased MoS_2-branch contributions (Supplementary Figure 11b) but were not detected in HBL/hBN/nanopillar structures. A detailed discussion of the PLE features is provided in Supplementary Note 10.

The fact that G-defect IXs are mainly populated via photoexcitation in WSe_2 absorption bands explains the slow uprising feature observed in the first few nano-second of the TRPL curve (Figure 2b). When the repetition rate of the pump laser is increased, the uprising feature becomes more pronounced due to the acceleration of carrier injection (Supplementary Note 11). Such a slow rise suggests slow arrival of holes to the hybridized G band. While the electron transfer from the CBM of WSe_2 to the S-vacancy state is energetically downhill suggesting fast electron transfer, the hole transfer from the K point of WSe_2 to the hybridized G point occurs uphill and requires an activation energy at a low temperature. To get a better insight into the exciton photoexcited dynamics, a nonlinear rate equation model for coupled K/-K exciton state in WSe_2 and G-defect IX is introduced in Supplementary Note 11. The model successfully

reproduces the initial time uprising feature in Figure 2b and yields slow exciton transfer time ~5 ns to the defect state. Additionally, the model also explains the decay of the side peaks of the $g^{(2)}$ function in Fig. 2d as a result of a dark defect shelving state receiving some portion of population from the G-defect IX on the timescale of 100 ns and re-populating back on the timescale of 2 .

In summary, we show that strain, defect, layer engineering, as well as photon excitation energy, can be utilized together to manipulate the IX formation in MoS_2/WSe_2 heterostructures for robust quantum light generation in the 1.15 – 1.45 eV NIR spectral range. Our approach bypasses the momentum conservation limitation to form IX which otherwise would require lattice-matched component layers and perfect rotational alignment. In general, such an approach unlocks the freedom of combining arbitrary 2D semiconductors for building IXs with desired emission energy and extending this strategy for engineering interfacial Γ point transitions in other 2D heterostructures, for instance, in MoS_2/WS_2. Given monolayer TMDC components used in our study with the majority of emission yield in visible spectral range can be combined together to generate IXs that emit in the NIR, our findings pave the way for extending the operational wavelength of 2D quantum light sources into the technologically important NIR regime.

Methods

Sample Preparation. A detailed description of the sample preparation process can be found in Supplementary Note 1. All 2D flakes were exfoliated from bulk crystals purchased from HQ Graphene Inc.

Optical Characterization. A diagram of our optical measurement setup is presented in Supplementary Note 12. Micro-PL measurements were performed on a home-built confocal microscope with excitation from a supercontinuum pulsed laser with 77.6 MHz repetition rate unless otherwise specified. The excitation power was typically ~1 - 100 nW. Samples were mounted in a continuous flow cryostat and cooled to 10 K using liquid helium. The emitted light was collected through a 50× infrared objective lens (Olympus, 0.65 NA) and spectrally filtered before entering a 2D InGaAs array detector (NIRvana® LN, Princeton Instruments). We used 150 and 300 gr/mm gratings to resolve the spectra. For TRPL and HBT experiments, the emission signal was spectrally filtered before coupling into a 50:50 optical fiber beamsplitter, which equivalently split the signal into two beams and sent them into two channels of a superconducting nanowire single-photon detector (SNSPD, Quantum Opus). PL intensity time traces, PL decay curves, and $g^2(t)$ traces were obtained from photon detection events recorded by a PicoQuant HydraHarp 400 time-correlated single photon-counting module. For pulsed auto-correlation measurements, $g^2(0)$ was extracted by comparing the integrated photon coincidence counts at the zero-time delay peak with the averaged integrated photon coincidence counts at adjacent peaks.

First-principles Calculations. First principles calculations based on density functional theory (DFT) were performed within the general gradient approximate (GGA) using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional as implemented in the Vienna Ab initio Simulation Package (VASP). The plane-wave energy cutoff is set to be 450 eV, and spin-orbit coupling is considered in our calculations. The vdW interactions in the heterostructures are included via the semiempirical Grimme-D3 scheme. A

rigid shift of 0.92 eV is used to align with the experimental optical band gap as DFT is known to underestimate the band gap. For MoS_2/WSe_2 heterobilayer without defect, a k-grid of 15x15x1 is adopted. For defect systems, a single vacancy is included in a 4x4x1 supercell, where the k-grid sampling used is 3x3x1. The strain effect of the nanopillars is simulated by a biaxial tensile strain of 2%.

Data Availability

The data that support the findings of this study are available from the corresponding author upon request.

References

1. Jiang, Y., Chen, S., Zheng, W., Zheng, B. & Pan, A. Interlayer exciton formation, relaxation, and transport in TMD van der Waals heterostructures. *Light: Science & Applications* **10**, 1-29 (2021).

2. Rivera, P. et al. Interlayer valley excitons in heterobilayers of transition metal dichalcogenides. *Nature Nanotechnology* **13**, 1004-1015 (2018).

3. Wilson, N.P., Yao, W., Shan, J. & Xu, X. Excitons and emergent quantum phenomena in stacked 2D semiconductors. *Nature* **599**, 383-392 (2021).

4. Huang, D., Choi, J., Shih, C.-K. & Li, X. Excitons in semiconductor moiré superlattices. *Nature Nanotechnology*, 1-12 (2022).

5. Turunen, M. et al. Quantum photonics with layered 2D materials. *Nature Reviews Physics*, 1-18 (2022).

6. Baek, H. et al. Highly energy-tunable quantum light from moiré-trapped excitons. *Science advances* **6**, eaba8526 (2020).

7. Branny, A., Kumar, S., Proux, R. & Gerardot, B.D. Deterministic strain-induced arrays of quantum emitters in a two-dimensional semiconductor. *Nature Communications* **8**, 1-7 (2017).

8. Palacios-Berraquero, C. et al. Large-scale quantum-emitter arrays in atomically thin semiconductors. *Nature Communications* **8**, 1-6 (2017).

9. Parto, K., Azzam, S.I., Banerjee, K. & Moody, G. Defect and strain engineering of monolayer WSe2 enables site-controlled single-photon emission up to 150 K. *Nature Communications* **12**, 1-8 (2021).

10. Yu, L. et al. Site-controlled quantum emitters in monolayer MoSe2. *Nano Letters* **21**, 2376-2381 (2021).

11. Zhao, H., Pettes, M.T., Zheng, Y. & Htoon, H. Site-controlled telecom-wavelength single-photon emitters in atomically-thin MoTe2. *Nature Communications* **12**, 1-7 (2021).

12. Aharonovich, I., Englund, D. & Toth, M. Solid-state single-photon emitters. *Nature Photonics* **10**, 631-641 (2016).

13. Rivera, P. et al. Observation of long-lived interlayer excitons in monolayer MoSe2–WSe2 heterostructures. *Nature Communications* **6**, 1-6 (2015).

14. Förg, M. et al. Moiré excitons in MoSe2-WSe2 heterobilayers and heterotrilayers. *Nature Communications* **12**, 1-7 (2021).

15. Seyler, K.L. et al. Signatures of moiré-trapped valley excitons in MoSe2/WSe2 heterobilayers. *Nature* **567**, 66-70 (2019).

16. Tran, K. et al. Evidence for moiré excitons in van der Waals heterostructures. *Nature* **567**, 71-75 (2019).

17. Kunstmann, J. et al. Momentum-space indirect interlayer excitons in transition-metal dichalcogenide van der Waals heterostructures. *Nature Physics* **14**, 801-805 (2018).

18. Unuchek, D. et al. Room-temperature electrical control of exciton flux in a van der Waals heterostructure. *Nature* **560**, 340-344 (2018).

19. Cho, C. et al. Highly strain-tunable interlayer excitons in MoS2/WSe2 heterobilayers. *Nano Letters* **21**, 3956-3964 (2021).

20. Heo, H. et al. Interlayer orientation-dependent light absorption and emission in monolayer semiconductor stacks. *Nature Communications* **6**, 1-7 (2015).

21. Okada, M. et al. Direct and indirect interlayer excitons in a van der Waals heterostructure of hBN/WS2/MoS2/hBN. *ACS Nano* **12**, 2498-2505 (2018).

22. Tebyetekerwa, M. et al. Twist-driven wide freedom of indirect interlayer exciton emission in MoS2/WS2 heterobilayers. *Cell Reports Physical Science* **2**, 100509 (2021).

23. Ubrig, N. et al. Design of van der Waals interfaces for broad-spectrum optoelectronics. *Nature Materials* **19**, 299-304 (2020).

24. Karni, O. et al. Infrared interlayer exciton emission in MoS 2/WSe 2 heterostructures. *Physical Review Letters* **123**, 247402 (2019).

25. Tan, Q. et al. Layer-engineered interlayer excitons. *Science Advances* **7**, eabh0863 (2021).

26. Mitterreiter, E. et al. The role of chalcogen vacancies for atomic defect emission in MoS2. *Nature Communications* **12**, 1-8 (2021).

27. Komsa, H.-P. & Krasheninnikov, A.V. Electronic structures and optical properties of realistic transition metal dichalcogenide heterostructures from first principles. *Physical Review B* **88**, 085318 (2013).

28. Tongay, S. et al. Tuning interlayer coupling in large-area heterostructures with CVD-grown MoS2 and WS2 monolayers. *Nano Letters* **14**, 3185-3190 (2014).

29. Susarla, S. et al. Mapping Modified Electronic Levels in the Moiré Patterns in MoS2/WSe2 Using Low-Loss EELS. *Nano Letters* **21**, 4071-4077 (2021).

30. Alexeev, E.M. et al. Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures. *Nature* **567**, 81-86 (2019).

31. Klein, J. et al. Engineering the luminescence and generation of individual defect emitters in atomically thin MoS2. *ACS Photonics* **8**, 669-677 (2021).

32. Wang, W. & Ma, X. Strain-induced trapping of indirect excitons in MoSe2/WSe2 heterostructures. *ACS Photonics* **7**, 2460-2467 (2020).

Declarations

Acknowledgements

This work was performed at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Los Alamos National Laboratory (LANL), an affirmative action equal opportunity employer, is managed by Triad National Security, LLC for the U.S. Department of Energy's NNSA, under contract 89233218CNA000001. Deterministic quantum emitter creation capability was developed under the support of DOE BES, QIS Infrastructure Development Program BES LANL22. HH, XL, VC, and AP acknowledge partial support form Laboratory Directed Research and Development (LDRD) program 20200104DR. HZ and HH is also partially supported by Quantum Science Center. MTP is supported by LDRD 20210782ER and LDRD 20210640ECR. HZ also acknowledge a partial support from LANL Director's Postdoctoral Fellow Award. L.Z. is supported by the National Science Foundation (NSF) grant No. DMR-2124934, and L.Y. are supported by the Air Force Office of Scientific Research (AFOSR) grant No. FA9550-20-1-0255. This first-principles simulation uses the Extreme Science and Engineering Discovery Environment (XSEDE), which is supported by National Science Foundation (NSF) grant number ACI-1548562.

Author Contributions

HZ, under the supervision of HH, conceived, designed, and conducted the experiment, analyzed the data, and composed the initial draft of the paper. LZ and LY performed first principles calculations. MTP conducted Raman measurements. XL and VC assisted HZ in PL measurements. JKB assisted in sample preparation. AP performed the modeling of the TRPL and photon correlation function. HH and HZ wrote the manuscript with input from all the authors.

Competing Interests

The authors declare no competing interests.

Figures

Figure 1

Interlayer excitons (IXs) from MoS_2-WSe_2 heterostructures. a, b, c, type-II band alignment of K-K IX (**a**), G-K IX (**b**), and Γ-defect IX (**c**). The excitons are indicated by pink ovals. **d, e, f,** schematics of the heterostructures on nanopillar arrays (side view): HBL (**d**), HBL/hBN (**e**), and HTL (**f**). The green, blue, and purple layers are MoS_2, WSe_2, and hBN, respectively. The nanopillars residing on gold substrates are denoted by red color. **g** Optical image of an HBL flake on nanopillar arrays. Note that the top right corner is HBL/hBN. Brown dashed line: HBL with 2^o twist angle; white dashed line: HBL/hBN; black line: hBN flake; green line: 3L-MoS_2 / 1L-WSe_2; red line: 2L-MoS_2 / 1L-WSe_2 (HTL). **h** and **i**, the wide-field PL image of **g**, under excitation wavelength of 750 nm and 660 nm, respectively. **j**, Typical NIR PL spectra of HBL/nanopillar (blue), HTL/nanopillar, homogeneous HBL/hBN (brown), and HBL/hBN on nanopillar (red). **k**, Histograms of emission energies of HBL (violet) and HTL (brown) on nanopillars. Data were collected in six different samples. All measurements were conducted at 10 K temperature.

Figure 2

PL and photon correlation characterizations of localized HBL emitters on nanopillar arrays. a, PL spectrum of a localized HBL emitter on a nanopillar. The data in Figure 2b-2d are taken from this emitter with a band-pass filter that allows the shadowed spectral region to be detected. **b,** the PL decay curve (blue) and theoretical fit to the data (black) with an 18.45±0.01 ns extracted lifetime. Inset: The first 12 ns of the PL decay, which shows an uprising feature before the beginning of the single-exponential decay. **c,** PL intensity as a function of experiment time showing stability of the single-photon emission rate over

1000 s time. **d,** Second-order correlation measurement under 750 nm pulsed excitation with a 6.5 MHz repetition rate, from which a $g^{(2)}(0) = 0.01$ is extracted. **e,** Second-order correlation measurement of another HBL emitter on a nanopillar measured at 77 K, from which a $g^{(2)}(0)$ of 0.45±0.06 is extracted. **f,** Temperature-dependent PL of a localized HBL emitter measured from 20 K to 100 K. The inset shows the evolution of integrated PL counts with temperature increase. All data were obtained at 10 K temperature unless otherwise specified.

Figure 3

Understanding the roles of strain, defect, and layer-number engineering in MoS_2-WSe_2 IX emission. a-d, Band structures of (**a**) intrinsic MoS_2/WSe_2 HBL (**b**) MoS_2/WSe_2 HBL with 2% biaxial strain (**c**) MoS_2/WSe_2 HBL with a S vacancy and 2% biaxial strain (**d**) intrinsic 2L-MoS_2/1L-WSe_2 HTL. Projected wavefunction components of individual layers are represented by the color bar. The percentages of MoS_2 and WSe_2 wavefunction at G band edge are presented. **e,** PL spectra of a homogeneous BN/MoS_2/BN sandwich (brown line) and a bare MoS_2 on nanopillar (orange line) measured at 10 K temperature. The exciton (^0X_A), trion (^-X_A), and defect-band (X_L) emission bands of MoS_2 are labeled. **f,** Room-temperature PL spectra of a localized HBL emitter on nanopillar (green), a localized WSe_2 emitter on nanopillar (purple), and a homogeneous HBL sample (blue). The homogeneous HBL displays a G-K IX emission at 1.57 eV, which evinces a near-zero twist angle.

Figure 4

PLE study of MoS_2-WSe_2 IX emissions. a, b, The PLE intensity maps as a function of both excitation and emission photon energies. **a**, the PLE map of a K-K IX emission on a nanopillar; **b**, the PLE map of a G-defect IX emission on a nanopillar. The energy of the intralayer excitons of each material is marked by grey dashed lines. **c, d,** Schematic representations of the energy band alignment in HBL/hBN on nanopillars (**c**), and HBL on nanopillars with defect bands (**d**). The black arrows show interband absorptions. The dashed gray arrows show charge transfer processes leading to the IX formations. The configuration for the K-K and G-defect IXs are represented by the dashed brown oval and the dashed red oval, respectively. **e,** PL intensity at the K-K (purple line) and G-defect (blue line) IX peak as a function of excitation photon energy. The PL spectra of the same G-defect IX emitter collected at visible (green line) and near-infrared (brown line) spectral range are also shown. The spectral range of optical absorptions from WSe_2-branch and MoS_2-branch bound excitons are indicated by the blue and yellow shaded regions, respectively. All data were taken at 10 K temperature.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- ZhaodefectgammaIXSIsubmitfinal3b.docx