One-Dimensional Moiré Excitons in Transition-Metal Dichalcogenide Heterobilayers

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The formation of interfacial moiré patterns from angular and/or lattice mismatch has become a powerful approach to engineer a range of quantum phenomena in van der Waals heterostructures1,2,3,4. For long-lived and valley-polarized interlayer excitons in transition-metal dichalcogenide (TMDC) heterobilayers5, signatures of quantum confinement by the moiré landscape have been reported in recent experimental studies6–9. Such moiré confinement has offered the exciting possibility to tailor new excitonic systems, such as ordered arrays of zero-dimensional (0D) quantum emitters1 and their coupling into topological superlattices1,10. A remarkable nature of the moiré potential is its dramatic response to strain, where a small uniaxial strain can tune the array of quantum-dot-like 0D traps into parallel stripes of one-dimensional (1D) quantum wires4. Here, we present direct evidence for the 1D moiré potentials from real space imaging and the corresponding 1D moiré excitons from photoluminescence (PL) emission in MoSe2/WSe2 heterobilayers. Whereas the 0D moiré excitons display quantum emitter-like sharp PL peaks with circular polarization, the PL emission from 1D moiré excitons has linear polarization and two orders of magnitude higher intensity. The results presented here establish strain engineering as a powerful new method to tailor moiré potentials as well as their optical and electronic responses on demand.
We choose the MoSe$_2$/WSe$_2$ heterobilayer as model because signatures of moiré interlayer excitons trapped in 0D moiré potential have recently been reported$^{8,9}$. The heterobilayer is either sandwiched between thin hexagonal boron nitride (h-BN) flakes, schematically illustrated in Fig. 1a, or (wherever noted) without the top h-BN capping, obtained from the transfer stacking method$^{11,12}$. TMDC heterobilayers are intrinsically endowed with moiré landscapes, as illustrated by the hexagonal moiré pattern in Fig. 1b with period $b \approx a/\sqrt{\Delta \theta^2 + \delta^2}$, where $a$ is the monolayer lattice constant and $\delta$ is the lattice mismatch and $\Delta \theta$ is the twist angle$^{1,13}$. The latter can be determined by second harmonic generation (SHG)$^{14}$. This moiré pattern can be strongly modified by applying differential strain (i.e. inequivalent strains on the two constituting layers). Specifically, a modest uniaxial strain on the order of $\delta$ can transform the moiré pattern into a hierarchy of a primary structure consisting of elongated ovals (arrows) that are parallel to each other to form a secondary structure of pseudo 1D stripes (dashed lines), Fig. 1c. For small $\Delta \theta$ (from $0^\circ$ or $60^\circ$), the differential and uniaxial tensile strain merges the elongated ovals into 1D stripes, Fig. 1d$^4$. We directly image the moiré superlattice structure using piezoresponse force microscopy (PFM) on heterobilayer samples. Since TMDC monolayers and bilayers are known to exhibit strong piezoelectric response$^{15,16}$, the moiré landscape modulates the piezoelectric tensor elements, leading to lateral deformations of the TMDC heterostructure in the presence of a vertically applied AC electric field. This electromechanical coupling is the basis for sensitive real space imaging by PFM of the moiré patterns typically not visible in conventional atomic force microscopy (AFM)$^{17}$.

We observe two distinct moiré superlattice structures on multiple samples. Figure 1e shows the PFM image of type-I, i.e., normal hexagonal moiré pattern (illustrated in Fig. 1b), for a MoSe$_2$/WSe$_2$ heterobilayer without top h-BN capping on a polypropylene carbonate (PPC)/soft tape substrate. Fast Fourier transform (FFT, top right inset in Fig. 1e) identifies the pseudo 6-fold symmetry, with a superlattice constant of $b = 6.0\pm0.5$ nm, corresponding to $\Delta \theta = 57.0\pm0.5^\circ$, in good agreement with AB stacking of $\Delta \theta = 58.0\pm0.4^\circ$ determined in SHG (Fig. S1a). Similar type-I PFM image has also been obtained for heterobilayer with thin (2 nm) top BN capping, albeit at reduced resolution (Fig. S2). The second type is distorted moiré 1D superlattice experimentally observed as type-II PFM image in Fig. 1f. This is the same MoSe$_2$/WSe$_2$ heterobilayer sample imaged in Fig. 1e, but transferred from the tape to a harder SiO$_2$/Si substrate. The image features 1D-stripes with longitudinal periodicity of $10.0\pm2.0$ nm in the left and top regions in Fig. 1f, as
confirmed by the 2-fold symmetry in FFT (inset). The 1D structure may contain a hierarchy illustrated in Fig. 1c and partially visible as wavy stripes in the PFM image in Fig. 1f. In addition, the middle of the FFT reveals pseudo 6-fold symmetry. This is derived from pattern in the lower right, which is characterized by large rhombic unit cells (see yellow dashed box) with superlattice

![Image](image.jpg)

Fig. 1 | Piezo Force Microscopy (PFM) imaging reveals both hexagonal and quasi-1D moiré superlattices. (a) Schematic of the vertically stacked WSe₂/MoSe₂ heterobilayer with BN encapsulation on SiO₂/Si substrate. Schematic illustration of (b) intrinsic, (c-d) quasi-1D (strained) moiré superlattices with 8% differential and uniaxial strain (S) and with twist angles Δθ = 2° and 0.5°, respectively. The quasi-1D moiré in (c) is formed by an 8% uniaxial tensile strain on one (red) of the two monolayers. PFM image directly showing the intrinsic (e) and pseudo-1D strained (f) moiré patterns. The upper-right insert in each image shows 2D-FFT. Note the lower left side in (f) reveals regions of large superlattices with distorted hexagonal pattern, as illustrated by the yellow-dashed box. PFM imaging was carried out at 295 K. Panel (g) shows representative PL spectra from two WSe₂/MoSe₂ heterobilayer samples with h-BN encapsulation. Blue and red represent σ⁺ and σ⁻ circularly polarized emission under σ⁺ excitation. PL spectra were obtained with excitation density \( n_{\text{ex}} \approx 6.1 \times 10^9 \text{ cm}^2 \) at 4 K (\( h\nu_{\text{exc}} = 1.65 \text{ eV} \), repetition rate 76 MHz, pulse duration ~ 150 fs, same in all PL measurements).
constant varying between 20 and 50 nm, corresponds to varying local twist angles of $\Delta \theta = 59.2^\circ - 59.8^\circ$. The strain field causing the distortion of the moiré pattern varies from location to location.

The observation of two types of moiré patterns by PFM imaging is accompanied by two distinctive types of PL emission spectra at low excitation densities ($n_{\text{ex}} \leq 1 \times 10^{10}$ cm$^{-2}$). On ~20% of samples, we observe sharp PL emission peaks (full-width-at-half-maximum, FWHM $\leq 1$ meV), labeled type-I, with strong circular polarization under circularly polarized excitation (Fig. 1g). On the majority of samples (~80%), we observe a single broader PL emission peak (FWHM $\geq 8$ meV), labeled type-II, with little circular but strong linear polarization (see below). For either AA- or AB-stacked samples, the PL intensity of type-II is over two-orders of magnitude higher than that of type-I. These two distinct types of PL features originate from the two types of moiré patterns as we establish below.

The type-I PL is consistent with the spectral features reported by Seyler et al., which has been attributed to the 0D moiré excitons in the hexagonal moiré landscape (Fig. 1b). In such a moiré landscape, the excitonic potential traps in each super-lattice unit cell are located at high-symmetry points with C$_{3v}$ symmetry, a pre-requisite for preserving circular valley optical selection rule. At an excitation density of $n_{\text{ex}} \sim 1 \times 10^{10}$ cm$^{-2}$, we estimate that the total number of interlayer excitons, $N_{\text{ex}}$, in a diffraction-limited focal spot ($\sim 1 \times 10^{-8}$ cm$^2$) is about 100. This finite number of excitons can be loaded into a finite number of quantum-dot-like traps, with local variations due to heterogeneity in strain and electrostatics. As a result, one observes quantum dot like PL emission. We characterize in detail such type-I PL emission on a MoSe$_2$/WSe$_2$ heterobilayer sample with h-BN encapsulation (top and bottom), with a small twist angle $\Delta \theta = 1.6 \pm 0.4^\circ$ and sub-nm scale flatness (see Fig S3). The results are in excellent agreement with those reported by Seyler et al..

The type-II PL with a single emission peak, seen in ~80% of the samples with AA or AB stacking, originates from the type-II 1D stripe pattern as revealed by the linear polarization of the emitted light. We spatially correlate PFM imaging with PL polarization on two representative spots, labeled 1 and 2 on the AFM image in Fig. 2a, on the same MoSe$_2$/WSe$_2$ heterobilayer sample as in Fig. 1e. PL spectra from these two spots are shown in Fig. 2b. The vertical 1D stripe patterns in PFM images from both spots, Fig. 2c and Fig. 2d, are correlated with the linearly polarized PL emission, Fig. 2e and Fig. 2f. In each case, the direction of the peak linear polarization distribution is at an acute angle from the main direction of the stripes in PFM image and can be correlated with the primary structure (elongated ovals in Fig. 1c) in the hierarchical 1D moiré patterns. Linearly
polarized PL emission is well known from semiconductor nanowires and carbon nanotubes\textsuperscript{20,21}. Thus, we take the linear PL emission and its spatial correlation with the real-space 1D moiré morphology as evidence for the 1D moiré potential.

We now analyze the type-II PL emission in detail using MoSe\textsubscript{2}/WSe\textsubscript{2} heterobilayer samples completely encapsulated by h-BN, which is known to reduce electrostatic heterogeneity, decrease PL peak width, and increase PL intensity\textsuperscript{22}. Fig. 3 shows key characteristics of PL emission for

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Direct correlation between 1D moiré potential and linearly polarized PL emission. (a) AFM characterizations of WSe\textsubscript{2}/MoSe\textsubscript{2} heterostructure without top BN encapsulation on a SiO\textsubscript{2}/Si substrate (twist angle $\Delta \theta = 58.4 \pm 0.4^\circ$). The squares, “(1)” and “(2)”, mark the spatial locations within the heterobilayer (HB) where subsequent PFM and optical linear polarization experiments are performed. (b) PL spectra acquired for regions (1) and (2). (c) and (d) PFM images showing quasi-1D strained moiré patterns corresponding to region (1) and (2), respectively, in the AFM image (a). (e) and (f) Angular dependence of PL emission polarization (blue dots), with fits as blue curves. $\theta$ denotes the phase of the emission polarization, while $p$ denotes the degree of linear polarization, with $p = (I_{\text{max}} - I_{\text{min}}) / (I_{\text{max}} + I_{\text{min}})$, where $I_{\text{max}}$ ($I_{\text{min}}$) is the maximum (minimum) PL emission intensity. Excitation density $n_{\text{ex}} = 2.5 \times 10^{11}$ cm\textsuperscript{-2} (otherwise same experimental conditions as in Fig. 1g).}
\end{figure}
an AA-stacked MoSe$_2$/WSe$_2$ heterobilayer with $\Delta \theta = 3.3 \pm 0.4^\circ$ (see Fig. 3a for an optical image, Fig. S1c for SHG, and Fig. S5 for AFM image). Confocal PL imaging (intensity map in Fig. 3b) demonstrates that most of the sampling spots are characterized by single or predominantly single PL peaks. Four representative PL spectra corresponding to spots 1-4 in Fig. 3b are shown in Fig. 3c. The upper left side of the heterobilayer sample, as represented by spectra 1 and 2, is characterized by nearly pure single PL peaks, with no detectable circular polarizations. In contrast, spectra 3 and 4, representing the lower-right of the sample, are characterized by a main single PL peak on a background of multiple but poorly resolved peaks. Each main PL peak shows no circular polarization while the multiple background peaks show partially circular polarization; the latter is reminiscent of PL emission from type-I (Fig. S3). Figs. 3e-h show polar plots of PL polarization.
(blue dots) as a function of azimuthal angle ($\theta$) in the surface plane from four representative locations, at $n_{\text{ex}} = 8.6 \times 10^{10}$ cm$^{-2}$. In each case, the solid blue curve is fit to $I = A*\cos^2(\theta - \theta_0) + C$: where the first term describes linear polarization in the direction $\theta_0$ and the second term is an isotropic distribution. For regions exhibiting pure type-II PL emission (Fig. 3e, f), the polarization is also purely linear ($C$ negligible). In regions (Fig. 3g, h) characterized by mainly type-I and partially type-II-like PL emission, the anisotropic distribution of polarization can be approximated by $A$ and $C$ of similar amplitudes.

Fig. 3d summarize fitting results to the PL emission from four spots on the PL image map (5% contours). Importantly, the direction of linear polarization, which we correlate with the direction of the primary structure in 1D moiré strips, evolves from spot to spot ($\theta_0 = 95^\circ, 141^\circ, 37^\circ, \text{and } 31^\circ$ for e, f, g, h, respectively), suggesting the spatial evolution of strain fields in the sample. On the other hand, the PL intensity is independent of the polarization of excitation light, at the intralayer exciton energy, as shown by the isotropic dependence (orange dots in Fig. 3e). Thus, we infer that the 1D moiré pattern has negligible effect on the intralayer excitons, where moiré potential is expected to be shallower than that for interlayer excitons$^1$.

The hetero-strain field can vary locally depending on details of fabrications and sample condition, such as the presence of bubbles in the heterobilayer or between the heterobilayer and BN encapsulation layers. Both 0D and 1D moiré traps can co-exist at different regions on the same sample. We have confirmed this, as shown in Fig. S8 where both type-I and type-II PL emissions are observed at different locations on a single MoSe$_2$/WSe$_2$ heterobilayer sample with $\Delta\theta = 2.6 \pm 0.5^\circ$.

The formation of 1D moiré excitons on a strained moiré landscape explains key properties of type-II PL emission. The loss of valley-contrast properties, i.e. the absence of circular polarized emission, is attributed to the breaking of $C_3$ rotational symmetry by the 1D moiré trap. Thus, the electron-hole exchange can hybridize the two valley configurations of excitons and lead to a linearly polarized ground state$^{23}$. Moreover, the remarkable enhancement in type-II PL emission intensity, as compared to the type-I emission from 0D moiré excitons, also supports its origin from 1D moiré potential traps. This PL enhancement can be attributed to the much larger oscillator strength, which is proportional to the area of the trap, as well as the quasi-continuous density of states associated with the 1D potential wells. The latter results in faster relaxation of excess energy.
to reach the bottom of the potential well where the radiative recombination rate is highest and larger phase space for energy-momentum conservation in radiative recombination.

Since both 0D and 1D moiré excitons result from relatively shallow local potential traps on a 2D landscape, we expect interlayer excitons to also form outside these traps at sufficiently high excitation densities due to mutual repulsion among these excitons with permanent electric dipoles. These are confirmed for the 1D moiré excitons in Fig. 4. The first evidence comes from the reduction of linear polarization. As shown in Fig. 4a, when the excitation density increases from \( n_{\text{ex}} = 6.1 \times 10^9 \) to \( 7.3 \times 10^{12} \, \text{cm}^{-2} \), the purity of linear polarization, \( p = (I_{\text{max}} - I_{\text{min}}) / (I_{\text{max}} + I_{\text{min}}) \), decreases from 0.89 to 0.63. The second evidence comes from the evolution in peak shapes, Fig. 4b. With increasing \( n_{\text{ex}} \), the single PL peak attributed to the 1D moiré exciton evolves into a multi-peak shape, with growth of shoulder features on both lower and higher energy sides. This evolution is particularly evident above the Mott density \( (n_{\text{Mott}} \sim 3 \times 10^{12}) \).

![Fig. 4](image_url)

**Fig. 4 | Evolution of PL peak shape with exciton density for the 1D moiré excitons.** The data are obtained on the same sample as used in Fig. 2. (a) Linear-polarization distributions of PL emission at two excitation densities; the degree of polarization, \( p = 0.89 \) and 0.63, at \( n_{\text{ex}} = 6.1 \times 10^9 \, \text{cm}^{-2} \) and \( 7.3 \times 10^{12} \, \text{cm}^{-2} \), respectively. (b) Intensity normalized PL spectra (dots) acquired with varying excitation densities \( n_{\text{ex}} = 1.1 \times 10^{11} \, \text{cm}^{-2} - 3.4 \times 10^{13} \, \text{cm}^{-2} \). The colored curves are deconvolutions detailed in the text. The dashed lines mark the peak positions (1-4) of the four deconvoluted peaks. (c) Integrated intensities of the deconvoluted PL peaks shown in (b) as a function of excitation density. Sample temperature = 4 K.

The solid curves in Fig. 4b show approximate deconvolutions of the spectra into the main asymmetric PL peak (1) and three Gaussian peaks, with positions shown by the dashed lines. Peaks (2) and (3) are on the lower energy side of the main peak, and peak (4) is on the higher energy side. Similarly, we observe the evolution of the initially quantum-emitter like type-I moiré excitons...
into broader multiple PL peak-shapes as excitation density is increased above \( n_{\text{Mott}} \), as shown in Figs. S6 and S7. These results suggest the radiative recombination from increased population of interlayer excitons outside the 0D or 1D moiré traps at sufficiently high excitation density.

The exact origin of multiple PL peak shapes for interlayer excitons in the MoSe\(_2\)/WSe\(_2\) heterobilayer at high excitation densities is not well understood at the present. A recent report by Tran et al.\(^9\) attributed similar multiple peaks in the WSe\(_2\)/MoSe\(_2\) heterobilayer to quantized levels within the 0D moiré trap. Here we observe the multiple PL peaks in most WSe\(_2\)/MoSe\(_2\) heterobilayer samples only at high excitation densities, regardless of the initial nature of the moiré excitons (type-I or type-II); see Figs. S6, S7, and S9. This suggests that the four-peak structure originates from radiative recombination of free interlayer excitons outside the moiré traps.

In both MoSe\(_2\)/WSe\(_2\) and MoS\(_2\)/WS\(_2\) heterobilayers, density functional theory (DFT) calculations suggest that the conduction band minima are at the \( Q \) point, not the \( K \) point\(^{24,25}\). Thus, the two emission peaks on the lower energy side of the main PL peak may be attributed to the spin-orbital split conduction bands at \( Q \) involving the momentum indirect excitons \( Q_C - K_V \), where the subscript denote the conduction (\( c \)) and valence (\( v \)) bands, respectively. Previous pump-probe reflectance and time-resolved PL measurements on WSe\(_2\)/MoSe\(_2\) heterobilayers have revealed efficient scattering from \( K \) to \( Q \) valleys in the Brillouin zone\(^{12}\). In the related system of WS\(_2\)/MoS\(_2\), a time- and angle-resolved photoemission spectroscopy (TR-ARPES) study provided direct evidence for photo-excited electrons in the \( Q \) valleys that are of strongly mixed character of both TMDC monolayers\(^{26}\). Similarly, the emission peak on the high energy side may come from the momentum indirect \( K^* C - K_V' \) involving the upper spin-orbital split conduction band (\( * \)). Radiative emission of these momentum-indirect excitons can be facilitated at high density through the pair-annihilation, with momentum conservation satisfied, as detailed in Fig. S10. Similar to the mechanism proposed by Rivera et al.\(^{27}\) (see Fig. S12D in ref. \(^{27}\)), a pair of inter-valley dark excitons, \( K^* C - K_V \) (blue arrow) and \( K^* C' - K_V \) (blue dashed arrow) can virtually swap the electron-hole pairing and thus emit two photons through the intermediate state of two bright excitons (peak 4). Similarly, for a pair of dark \( Q_C - K_V \) and \( Q_C' - K_V' \) excitons, the pairwise \( Q \) to \( K \) and \( Q' \) to \( K' \) scattering of their electrons can result in two virtual \( K_C - K_V \) and \( K_C' - K_V' \) bright excitons (peaks 2 and 3). Radiative recombination from the pair annihilation of dark excitons is expected at higher exciton density, in comparison with the normal PL emission from the bright \( K_C - K_V \) exciton. Supporting this, we note that the multiple peaks (2-4) attributed to momentum-indirect excitons
are vanishingly small at low excitation densities (≤1x10^{11} \text{ cm}^{-2}) but grow much faster with \( n_{\text{exc}} \) than peak (1), Fig. 4c.

The discovery of the 1D moiré potential landscape from uniaxial strain and the corresponding 1D moiré excitons in TMDC heterobilayers has two implications in the growing field of moiré physics. On the one hand, given the susceptibility of 2D crystals to strain within samples consisting of multilayers created by mechanical transfer stacking, one must consider the presence of strain fields in understanding the moiré potential and its heterogeneity in determining physical properties. On the other hand, this very sensitivity to strain field provides a new method to control the moiré potential landscape in heterobilayers, leading to optical and electronic responses on-demand. The 1D moiré potential is particularly attractive for strongly correlated and anisotropic charge transport\(^4,28\). In comparison to the large strain required to bring in significant changes in monolayers of graphene\(^{29-31} \) and TMDCs\(^{32-34} \), moiré strain engineering can be achieved with a modest heterostrain in the order of the lattice mismatch\(^4 \). Combined with the twist angle, active or passive control of strain fields thus open the door to greater opportunities of artificially generating band structures\(^{28} \) and topological mosaics\(^4 \) at 2D material interfaces.

**MATERIALS and METHODS**

**TMDC monolayers.** Monolayers of WSe\(_2\) and MoSe\(_2\) were mechanically exfoliated from bulk crystals grown by the self-flux method. These monolayers possessed low defect densities (≤10^{11} \text{ cm}^{-2})\(^35 \). Flakes of h-BN with thickness 5 – 35 nm and with atomically flat surfaces were obtained by mechanical exfoliation. The flakes (WSe\(_2\), MoSe\(_2\), and h-BN) were characterized by atomic force microscopy (AFM) and Raman spectroscopy.

**Determination of TMD zigzag crystal orientation via SHG.** The crystal orientations of WSe\(_2\) and MoSe\(_2\) monolayers were determined by second harmonic generation (SHG) measurement on an inverted optical microscope (Olympus IX73). Linearly polarized femtosecond laser light (Spectrum Physics Tsunami, 80 MHz, 800 nm, 80 fs) was focused onto a monolayer with a 100x, NA 0.80 objective (Olympus LMPLFLN100X). The reflected SHG signal at 400 nm was collected by the same objective, filtered by a short-pass dichroic mirror, short-pass and band-pass filters, and a Glan-Taylor linear polarizer, detected by a photomultiplier tube (Hamamatsu R4220P), and recorded by a photon counter (BK PRECISION 1823A 2.4GHz Universal Frequency Counter). We obtain the azimuthal angular (θ) distribution of SHG signal by rotating the laser polarization.
and the SHG signal (via a half waveplate) with fixed sample orientation. Due to the D$_{3h}$ symmetry, the non-vanishing tensor elements of the second order susceptibility of WSe$_2$ and MoSe$_2$ monolayers are $\chi^{(2)}_{yy} = -\chi^{(2)}_{yx} = -\chi^{(2)}_{xy} = -\chi^{(2)}_{xx}$ where the $x$ axis is defined as the zigzag direction ($I$). When we simultaneously rotated the fundamental and SHG signals, the SHG intensity showed six-fold symmetry: $I_\perp \propto \cos^2(3\theta)$ and $I_\parallel \propto \sin^2(3\theta)$, where $\theta$ is the angle between the laser polarization and the zigzag direction. We use triangular flakes of monolayer WS$_2$ (6Carbon) or MoS$_2$ (2DLayer), where zigzag directions are the same as crystal edges, both grown from chemical vapor deposition (CVD), to calibrate the SHG setup.

**Preparation of 2D WSe$_2$/MoSe$_2$ heterostructure samples.** The 2D WSe$_2$/MoSe$_2$ heterobilayer samples were prepared from the polymer-free van der Waals assembly technique$^{11}$. A transparent polydimethylsiloxane (PDMS) stamp coated with a thin layer of polypropylene carbonate (PPC) was used to pick up a thin layer of exfoliated h-BN. This h-BN was then used to pick up the first TMDC monolayer. The second TMDC monolayer was aligned to and picked up by the first monolayer on a rotation stage. After picking up a second BN flake as the bottom encapsulation layer, we transferred the entire structure on to a clean silicon wafer (with 285 nm thermal oxide layer for enhanced optical contrast) at elevated temperatures (90-130 °C). The residual PPC was washed away by acetone to give a clean h-BN/MoSe$_2$/WSe$_2$/h-BN heterobilayer on the Si/SiO$_2$ substrate. The samples were then thermal annealed in an ultrahigh vacuum chamber ($10^{-8}$-$10^{-9}$ Torr): the sample temperature was raised from room temperature to 523K slowly over two hours, and kept at this temperature (523K) for an additional three hours; then the sample was cooled down to 173K over 30 min, after which the sample was eventually raise up to room temperature over 12 hours.

**Polarization-resolved confocal microscopic measurements.** PL imaging was performed on a home-built scanning confocal microscope system (Fig. S11) based on a liquid-helium recirculating optical cryostat (Montana Instruments Fusion/X-Plane) with a 100x, NA 0.75 objective (Zeiss LD EC Epiplan-Neofluar 100x/0.75 HD DIC M27). The temperature of the sample stage could be varied between 3.8 K and 350 K. In all experiments presented in this study, the TMDC heterobilayer and monolayers samples were at 4 K in a vacuum (<$10^{-6}$ torr) environment, unless otherwise noted. A Galvo-Galvo scanner (Thorlabs, GVS012/M) was used for mapping the PL signal emitted from the sample plane. Polarizers, $\lambda/2$ waveplate, and $\lambda/4$ waveplate were used for circular/linear polarization-resolved experiments (see Fig. S11 for the detailed optical setup). The incident laser beam (Coherent Rega, 750 nm, ~ 150 fs, 76 MHz) was focused by the objective to
a diffraction limited spot on the sample. The excitation power was measured by a calibrated power meter (Ophir StarLite) with broad dynamic range. The PL light was collected by the same objective, spatially and spectrally filtered, dispersed by a grating, and detected by an InGaAs photodiode array (Princeton Instruments PyLoN-IR). The wavelength was calibrated by neon-argon and mercury atomic emission sources (Princeton Instruments IntelliCal).

**Piezoresponse force microscopy measurements.** All PFM imaging experiments were performed on a commercially available Bruker Dimension Icon AFM with a Nanoscope V controller. We used ASYELEC-01 Ti/Ir coated silicon probes with a force constant of ~3 N/m from Oxford Instruments Asylum Research. The amplitudes of AC bias were <1 V and the single frequency excitation was at the lateral cantilever-sample resonance in the range 750-850 kHz. Most PFM imaging experiments were carried out on WSe₂/MoSe₂ heterobilayer samples without the top BN encapsulation layer, with the sample stack either on PPC/PDMS or transferred onto SiO₂/Si. When the sample was on the relatively soft PPC/PDMS substrate, we find a high probability of observing the intrinsic moiré pattern with hexagonal symmetry, as exemplified by Fig. 1e. For samples transferred onto the hard SiO₂/Si substrate, the presence of strain results in predominantly the 1D stripe patterns (Fig. 1f). Similar PFM images have been obtained for WSe₂/MoSe₂ heterobilayer samples with thin BN capping layers (Fig. S2), albeit at reduced resolution as compared to the exposed WSe₂/MoSe₂ heterobilayer.

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Supplementary Information

**Fig. S1** Determination of MoSe\(_2\)/WSe\(_2\) twist angles by azimuthal angle-resolved SHG. (a), (b) and (c) The six-fold symmetry in the angular distribution SHG intensity (dots) and fits (solid lines) for the MoSe\(_2\) and WSe\(_2\) monolayers, and their heterobilayer (HS). Panel (a) corresponds to the sample for PFM imaging in Fig. 1e and 1f, and the PFM-PL correlation measurements in Fig. 2. Panel (b) is the sample corresponding to type-I PL emission in **Fig. S3**; Panel (c) is the sample with linearly polarized PL as shown in **Fig. 3**. In (a), SHG signal from the heterobilayer is weaker than those from both the MoSe\(_2\) and the WSe\(_2\) monolayer (destructive interference), suggesting the stack is AB type; while in (b) and (c), SHG signal from the heterobilayer is stronger than either the MoSe\(_2\) and the WSe\(_2\) monolayer (constructive interference), suggesting the stack is AA type.

**Fig. S2** | Piezo-response force microscopy (PFM) image of a MoSe\(_2\)/WSe\(_2\) heterobilayer with thin (~2 nm) BN encapsulation. The 2D-FFT in the inset indicates a pseudo-6-fold symmetry of the moiré superlattice, with a constant of ~5.6 nm, corresponding to a twist angle of \(\Delta \theta \sim 3.4^\circ\).
Fig. S3 | Surface morphology of the WSe$_2$/MoSe$_2$ sample and polarization-resolved PL of type-I moiré excitons in the WSe$_2$/MoSe$_2$ heterobilayer. (a) Optical image of aligned MoSe$_2$/WSe$_2$ heterobilayer encapsulated in BN with small twist angles (determined by SHG in Fig. S2b); (b) Atomic force microscopy (AFM) characterization of the sample. The white solid lines (marked as “1”, “2” and “3”) trace the topography of the sample surface. Note that, the height fluctuations across 2 μm is within a few angstroms (the scale bar is 5μm). (c) Confocal photoluminescence microscope image of the BN-encapsulated WSe$_2$/MoSe$_2$ heterobilayer. The image is acquired with linearly-polarized excitation. (d) Circularly-polarized PL emission spectra ($\sigma^+$ blue, $\sigma^-$ red) from the WSe$_2$/MoSe$_2$ heterobilayer with $\sigma^+$ excitation. The numbers in parenthesis (“1”, “2”, “3” and “4”) describes the spatial locations of PL emission on the PL image map in (c). Experimental conditions: $h\nu_{\text{exc}} = 1.65$ eV, repetition rate = 76 MHz, pulse duration ~ 150 fs, $n_{\text{e-h}} \sim 6 \times 10^9$ cm$^{-2}$, temperature = 4 K.

Polarization-resolved confocal PL imaging (Figs. S3c&d) shows multiple sharp emission peaks (FWHM < 1 meV) with varying degree of circular polarization. The latter is quantified as $P = (I^- - I^+)/I^+ = 15$-40%, where $I^+$ ($I^-$) is the intensity of $\sigma^+$ ($\sigma^-$) PL emission under $\sigma^+$ excitation. We also determined the intensity distribution from the type-I PL emission while rotating a linear polarizer in the surface plane; the resulting angular distribution deviates from the completely isotropic distribution expected from 100% circular polarization and can be attributed to heterogeneity in strain (Fig. S4). We emphasize that the optical excitation density ($n_{\text{ex}} \sim 6 \times 10^9$ cm$^{-2}$) in these measurements is more than 2-3 orders of magnitude below the Mott density ($n_{\text{Mott}} \sim 3 \times 10^{12}$ cm$^{-2}$). These results are in excellent agreement with those reported by Seyler et al.$^2$. 

\[ \text{Experimental conditions: } h\nu_{\text{exc}} = 1.65 \text{ eV, repetition rate = 76 MHz, pulse duration ~ 150 fs, } n_{\text{e-h}} \sim 6 \times 10^9 \text{ cm}^{-2}, \text{ temperature = 4 K.} \]
**Fig. S4 | Polarization-resolved PL spectra for intrinsic (0D) moiré excitons.** (a) and (b) Emission and excitation polarization dependence of the interlayer excitons trapped in intrinsic (0D) moirés. (c) Polar plot of the integrated PL intensity based on the data shown in (a) and (b). Experimental conditions: \( h\nu_{\text{ex}} = 1.65 \, \text{eV}, \) repetition rate = 76 MHz, pulse duration \( \sim 150 \, \text{fs}, \) \( n_{\text{eh}} \sim 6 \times 10^9 \, \text{cm}^2, \) temperature = 4 K.

**Fig. S5 | Surface morphology of the WSe\(_2\)/MoSe\(_2\) sample for type-II PL emission.** AFM image of the WSe\(_2\)/MoSe\(_2\) heterostructure with BN encapsulation (scalebar is 4 \( \mu \text{m} \)), corresponding to SHG characterization in Fig. S1c. The heterobilayer region is highlighted by white dash line; the arrows point to bubbles in this heterostructure. The largest bubble likely exists at the WSe\(_2\)/MoSe\(_2\) interface, as its location corresponds to the dark area in PL imaging in Fig. 3b.
Fig. S6 | Excitation density-dependent PL spectra for intrinsic moiré excitons. (a) Circularly polarized PL emission from the same four spots (1-4) as in Fig. S3d, but at a higher excitation density of $n_{ex} \sim 7.3 \times 10^{12}$ cm$^{-2}$, for the same WSe$_2$/MoSe$_2$ heterobilayer exhibiting type-I PL emission. The degree of circular polarization ($P_s$) is much reduced for each spot in (a), as compared to Fig. S3d. Experimental condition: $h\nu_{ex} = 1.65$ eV, repetition rate = 76 MHz, pulse duration ~ 150 fs, temperature ~ 4K. (b) and (c) Intensity normalized PL spectra acquired with varying excitation densities from the same sample. The sharp peaks in the initial type-I PL emission broadens to multiple peaks as excitation density increases above $\sim 10^{11}$ cm$^{-2}$). Experimental condition: $h\nu_{exc} = 2.33$ eV, CW, temperature ~ 4K.

Fig. S7 | Spectral fitting of intrinsic moiré exciton emission at high excitation density. Gaussian peak fitting of the PL spectrum derived from the spectrum in Fig. S6c at an excitation density of $8.6 \times 10^{13}$ cm$^{-2}$. 
Fig. S8 | Coexistence of type-I and type-II PL emission spectra on the same sample. (a) Optical image of an aligned MoSe$_2$/WSe$_2$ heterobilayer encapsulated in BN with small twist angle; the scalebar is 5μm. (b) AFM image of the sample (scalebar is 5 μm); the white arrows point to bubbles in this sample. (c) The six-fold symmetry in the angular distribution of SHG (dots) along with fit (solid lines) for MoSe$_2$ and WSe$_2$ monolayers, and their stacked heterobilayer. SHG signal from the heterobilayer is stronger than that from either MoSe$_2$ or WSe$_2$ monolayer, suggesting the stack is AA type. (d) Confocal microscope PL mapping from the WSe$_2$/MoSe$_2$ heterobilayer (spectrally integrated photoluminescence intensity). The PL map was acquired with linearly-polarized excitation. (e) and (f) Selected spots from (d) showing two distinct types of PL features, i.e. type-II (e) and the type-I (f). Experimental condition: $h\nu_{exc} = 2.33$ eV, CW, temperature ~ 4K, excitation density ~ $1.05 \times 10^{10}$ cm$^{-2}$. 
**Fig. S9** | **Broadening of both type-I and type-II moiré excitons.** Excitation density dependent PL spectra for (a) type-I, circularly polarized emission attributed to normal moiré landscape; and (b) type-II, linearly polarized emission attributed to 1D moiré landscape. Panel (a) and panel (b) correspond to sampling spot s(8) and (3) shown in **Fig. S8**. Note that, the initially distinct PL shapes at low excitation densities ($n_{e-h} < 10^{12}$ cm$^{-2}$) become similar to each other at high excitation densities ($n_{e-h} > 10^{12}$ cm$^{-2}$).

**Fig. S10** | **Schematic illustration of direct and indirect interlayer excitons and radiative annihilation of the latter.** The band structure shows the conduction band minima at $Q$ and $Q'$ valleys of strongly mixed MoSe$_2$ and WSe$_2$ characters. The green solid and dashed arrows represent direct interlayer exciton emission at the $K$ and $K'$ valley to give peak 1 in PL emission.
at low excitation densities. The blue solid and dashed arrows represent indirect excitons involving $K$-$K'$ valleys. At high excitation densities, radiative annihilation can lead to the emission of two photons assigned to peak 4 in PL spectra. Similarly, the red arrows represent indirect excitons at $Q$-$K$ or $Q'$-$K'$ valleys. Radiative annihilation can yield peaks 2 and 3 in PL emission spectra. See text for details.

Fig. S11 | Optical setup for polarization-resolved confocal PL mapping. Polarization-resolved PL mapping measurements was performed on a home-built reflection-geometry microscope system based on a helium-recirculation optical cryostat (Montana, 3 K ~ 350 K). NDF: neutral density filter; Pol: Glan-Taylor polarizer; $\lambda/2$ WP: half waveplate; $\lambda/4$ WP: quarter waveplate; LPF: long-pass filter; DMLP805: 805 nm long-pass dichroic mirror; SL: telecentric scan lens; TL: tube lens. The black arrow (double-sided or circular) schematically illustrates the polarization nature of light. Note that, the above diagram is demonstrating the optical setup for circular polarization resolved experiments. For emission linear polarization resolved experiments, the $\lambda/4$ WP will be removed, while for the detection side, the $\lambda/2$ WP and Pol should be switched, and each time after $\theta$ degree rotation of Pol, the $\lambda/2$ WP should be rotated in the same direct but with $\theta/2$ degree, so that the dependence of the spectrometer detection sensitivity upon the polarization is eliminated.

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