Interlayer exciton laser of extended spatial coherence in atomically thin heterostructures

https://doi.org/10.1038/s41586-019-1779-x

Received: 31 December 2018
Accepted: 28 August 2019
Published online: 25 November 2019

Eunice Y. Paik1,4, Long Zhang1,4, G. William Burg2, Rahul Gogna3, Emanuel Tutuc2 & Hui Deng1*

Two-dimensional semiconductors have emerged as a new class of materials for nanophotonics owing to their strong exciton–photon interaction1,2 and their ability to be engineered and integrated into devices3. Here we take advantage of these properties to engineer an efficient lasing medium based on direct-bandgap interlayer excitons in rotationally aligned atomically thin heterostructures4. Lasing is measured from a transition-metal dichalcogenide heterobilayer (WSe2–MoSe2) integrated in a silicon nitride grating resonator. An abrupt increase in the spatial coherence of the emission is observed across the lasing threshold. The work establishes interlayer excitons in two-dimensional heterostructures as a gain medium with spatially coherent lasing emission and potential for heterogeneous integration. With electrically tunable exciton–photon interaction strengths5 and long-range dipolar interactions, these interlayer excitons are promising for application as low-power, ultrafast lasers and modulators and for the study of many-body quantum phenomena6.

Semiconductor lasers are ubiquitous in today’s technology because they are compact, cover a wide range of wavelengths and allow efficient electrical pumping and fast electrical modulation. They are predominantly based on traditional III–V quantum wells. To achieve lower power consumption, more compact size and a higher degree of integration with silicon, there has been tremendous effort to develop alternative gain materials and structures, such as nanowire lasers7, spasers8 and photonic crystal lasers9. However, tunability, electrical pumping and heterogeneous integration remain as common challenges.

Recently, monolayer transition-metal dichalcogenide crystals (TMDCs) have emerged as a new class of material for semiconductor lasers, as they are atomically thin and feature strong exciton emission1,2. Whereas lattice mismatch limits the choice of substrates for three-dimensional (3D) semiconductors, two-dimensional (2D) TMDCs do not have dangling bonds, and can be directly integrated with different substrates1. Previous studies have used two criteria to assess lasing in monolayer TMDCs: nonlinear intensity dependence, and linewidth reduction as a function of pump power10–12. However, the photon flux appears to be below the stimulated emission threshold of Spatial coherence—an important property for characterizing lasers—has not been studied. Hence it is difficult to exclude localized excitons, such as point defects, as the source of the observed nonlinear power dependence. Moreover, with only a monolayer as the gain medium, tunability is limited and vertical p–n junctions are not possible without contacting with other doped semiconductors.

In contrast, heterostructures open the door to the engineering of band structures and exciton states. Spatially indirect excitons in heterostructures have been intensively studied13, for they feature an electrically tunable static dipole with long-range dipole interactions, promising rich many-body quantum phenomena14. However, the reduced oscillator strength of spatially indirect excitons typically renders them dark and hard to access.

Here we show that in rotationally aligned 2D WSe2–MoSe2 heterobilayers integrated on a silicon nitride (SiN) cavity (Fig. 1), interlayer excitons form an efficient gain medium, supporting lasing with extended spatial coherence at a low population inversion density. As illustrated in Fig. 1b, by forming a direct bandgap between the two monolayers4 that are less than one nanometre apart, the interlayer excitons retain a sufficiently large oscillator strength. With type-II band alignment, the heterobilayer forms a three-level system that allows efficient pumping through the intralayer exciton resonances followed by rapid electron transfer to a lower-energy empty conduction band15–19 (Fig. 1c). As a result, population inversion is readily achieved at the reduced bandgap while avoiding fast intralayer radiative loss of the carriers. Moreover, unlike some of the cavities used for monolayer exciton lasers, the cavity mode in our device fully covers the heterobilayer, allowing gain over the full area of the bilayer, and supporting extended spatial coherence (Fig. 1a). We observe lasing accompanied by an abrupt increase in the spatial coherence length as the photon occupancy exceeds unity. The emission intensity increases nonlinearly more than 100-fold across the threshold, and then continues to increase linearly with pump power (without saturation) up to the highest power used. Our results establish interlayer excitons in engineered TMDC heterobilayers as an efficient lasing medium, which, compared to excitons in monolayer TMDCs, feature electrically tunable long-range dipole interaction and oscillator strength14, robust valley polarization19, and a type-II band alignment.
ill-suited for electrical injection via an atomically thin bilayer p–n junction.

The laser device comprises a rotationally aligned WSe2–MoSe2 heterobilayer placed on a Si grating resonator, as illustrated in Fig. 1a. To form bright interlayer excitons, we accurately align the crystal axes of the WSe2 and MoSe2 monolayers to within 1° of relative rotation, as verified by second-harmonic generation (SHG) measurements (Extended Data Fig. 1). Consequently, the band extrema at the K valleys of the two monolayers align in momentum space to form a direct bandgap (Fig. 1b). With type-II band alignment, carriers can be injected into the heterobilayer efficiently via the intralayer exciton resonance, followed by rapid electron transfer to the empty conduction band of MoSe2 on a timescale of 10–100 fs (Fig. 1c) as a result, band inversion can be established at the smaller, interlayer bandgap. Once separated into the two monolayers, radiative recombination is reduced, rendering long interlayer exciton lifetimes of the order of 1 ns (Extended Data Fig. 2). Photoluminescence (PL) measurements of the heterobilayer show that interlayer exciton emission is much stronger than intralayer emission (Fig. 2b), confirming efficient charge transfer and sufficient buildup of the interlayer-exciton population. Spatially resolved PL shows uniform emission from the interlayer (intralayer) excitons in the bilayer (monolayer) regions (Extended Data Fig. 3).

The grating cavity provides optical feedback when photons are coupled to its resonances. The cavity modes are sensitive to the propagation and the polarization directions of the electric field. We define the propagation (polarization) direction along the grating bar as x (TE) and across the bar as y (TM), as illustrated in Fig. 1a. We tune the grating period, thickness, and fill factor to obtain a high quality factor (Q-factor) for the TE mode and match it to the exciton resonance at zero in-plane wavenumber, \( k_x = 0 \). The heterobilayer lies directly on the grating where the evanescent field remains strong. The TM cavity modes are far blue-detuned from the excitons; therefore, the TM exciton modes are not affected by the cavity (Extended Data Fig. 4).

We confirm the TE-cavity modes by measuring the empty-cavity dispersion with angle-resolved reflectance spectroscopy; the results agree well with the simulation by rigorous coupled wave analysis (RCWA), as shown in Fig. 2c. The TE mode Q-factor from the simulation is around 2,000. However, the actual cavity Q-factor is presumably lower, owing to fabrication imperfections. From the reflectance spectra of the empty cavity, we estimate a Q-factor of between 500 and 680 (Fig. 2c inset), but the exact value is difficult to determine owing to low contrast and white-light noise. The PL spectral linewidth from the device corresponds to a Q-factor of around 630 (Fig. 2d).

The heterobilayer allows efficient optical pumping through the interlayer exciton resonances, which are far above the resonances of the interlayer excitons or the cavity. With the pump laser at 1.7 eV, the PL from the cavity mode at \( k_x = 0 \) and energy \( E = 1.35 \) eV brightens rapidly as the pump power increases, as seen in the along-bar angle-resolved PL (Fig. 3a).

Integrating over \( k_x \) = ±0.7 \( \mu \)m\(^{-1}\) and \( E = 1.352 \) to 1.359 eV, we obtain the photon occupancy \( I_p(k_x = 0) \) after accounting for the independently measured collection efficiency of the optical path (see Methods). As \( I_p(k_x = 0) \) approaches one, \( I_p(k_x = 0) \) shows a superlinear increase

**Fig. 2 | Properties of the heterobilayer and grating cavity.** a. An optical microscope image of the WSe2/MoSe2 heterobilayer integrated on a grating cavity. The red square outlines the grating region, and the red circle indicates the laser spot size. Inset, direction of the grating bars. b. PL spectrum from the heterobilayer. The sample was pumped with a 633-nm laser at power of 20 μW. The shaded boxes highlight the spectral range of interlayer (\( I_p \)), MoSe2, and WSe2 exciton emission. c. TE-polarized along-bar, angle-resolved, simulated (left and overlaid crosses in the right) and measured (right) reflectance spectrum. Inset, line-cut of the normalized reflectance spectrum around \( k_x \) = 1.7 \( \mu \)m\(^{-1}\) (blue trace); the red line is a fit to the cavity mode. The star symbol marks the peak of the fitted cavity mode. d. PL spectrum (blue dots) near \( k_x = 0 \). The pump was on resonance with WSe2, at pump power of 0.1 μW. The red line is a Lorentzian fit, with a fitted linewidth of 2.4 meV.
with pump power, consistent with the onset of stimulated emission into the cavity mode (Fig. 3b). The power-dependent PL measurement is reproducible, as shown by a measurement performed on a different day (Extended Data Fig. 5).

The pump power at the threshold of \( I_\text{th} = 1 \) is \( P_\text{th} = 0.18 \) μW. Considering the typical absorption efficiency (20%) of monolayer WSe\(_2\), we obtain the threshold carrier density \( n_\text{th} = 5.7 \times 10^{10} \) cm\(^{-2}\), in good agreement with the density required for the transparency condition \( n_e = 8 \times 10^{10} \) cm\(^{-2}\) (see Methods for calculations of \( n_e \)). Far above threshold, the output intensity becomes linear with pump power and does not saturate up to \( P = 28P_\text{th} \), the highest power used for TE measurements. The nonlinear increase of the intensity is reproduced by a simplified rate equation model, as described in Methods (Extended Data Fig. 6).

Accompanying the superlinear increase in the emission intensity at threshold, the linewidth of the emission drops sharply, as shown in Fig. 3b, signifying the increase of temporal coherence. The linewidth at excitation powers below –0.05 μW may be broader, but our detectors are not sufficiently sensitive to detect the emission. The saturation and slight increase of linewidth with increasing power above threshold may be due to interactions among the carriers and spatial mode competition. The sharp lasing emission decreases in intensity as we increase temperature, but persists up to 70 K, suggesting that lasing may survive at 70 K or higher (Extended Data Fig. 7).

In stark contrast with the TE emission, TM-polarized emission is not coupled to the cavity mode and does not show threshold behaviour. The emission becomes detectable only at high pump powers. An example is shown in Fig. 3c for \( P = 10 \) μW. The emission spreads uniformly in \( k \) over the numerical aperture of our collection optics and over a broad energy range of about 70 meV. With increasing pump power, the total integrated emission intensity increases sublinearly with pump power (Fig. 3d) and is a few times weaker than that of the TE intensity.

When integrated over the same small ranges of \( k \) and \( E \) near the lasing mode, the TE and TM output intensities differ by several orders of magnitude (filled diamonds in Fig. 3b, d). In other words, while the TM emission is suppressed and remains broadly distributed in \( k \) and \( E \), the TE emission is concentrated in ranges of energy and \( k \) that are one to two orders of magnitude smaller, as a result of stimulated emission.

To confirm the extended coherence expected of a laser with a 2D gain medium, we study the first-order spatial coherence function \( g^{(1)}(\mathbf{r}, \mathbf{r}) \) defined as follows:

\[
g^{(1)}(\mathbf{r}, \mathbf{r}) = \frac{\langle \rho(\mathbf{r}) \rho(\mathbf{r}) \rangle}{\langle \rho(\mathbf{r}) \rangle^2}
\]

where \( \rho \) is the density matrix operator, and \( E^{(+)} \) and \( E^{(-)} \) are field creation and annihilation operators, respectively.

Although spatial coherence properties have been extensively studied in semiconductor photon lasers, exciton–polariton lasers and plasmon lasers, coherence of TMDC lasers has not been studied thus far, making it difficult to rule out localized excitons as a source of lasing. However, the large spatial area of the grating resonator, and the large photon flux above threshold, allow us to investigate the spatial coherence of the interlayer exciton emission. First-order spatial coherence measurements were performed using a continuous wave excitation laser and a retro-reflector Michelson interferometer setup, where an image of the sample interferes with a centro-symmetrically inverted version of itself at the output with an intensity distribution \( I^\text{out}(\mathbf{r}) \) (Fig. 4a). Because of a small angle difference between the two beams, interference fringes are formed (Fig. 4b) that correspond to slightly varying path length differences \( \Delta z = z_0^\text{in} - z_0^\text{out} \) at different positions \( \mathbf{r} \) across the images; \( z_0 \) is the initial position of the retro-reflector.

Varying the path length of the interferometer, \( I^\text{out}(\mathbf{r}) \) of each \( \mathbf{r} \) oscillates, with the contrast of the oscillation proportional to the first-order spatial coherence, \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \) (Fig. 4c). We thus obtain spatial maps of \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \) (see Methods for details). Below threshold, the emission is too weak for \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \) measurements. Near threshold, the map is rather noisy without a clear pattern of \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \) versus \( \mathbf{r} \) (Fig. 4d, top panel). Above threshold, a clear pattern emerges, showing a high \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \) near \( \mathbf{r} = 0 \) that decays with increasing \( \mathbf{r} \) and extends above the background fluctuations to about twice the laser spot size (Fig. 4e, top panel).

To study the functional dependence of \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \), we average over \( y = \pm 0.2 \) μm and obtain \( g^{(1)}(\mathbf{x}, -\mathbf{x}) \). As shown in the bottom panels of Fig. 4d, e, the decay of \( g^{(1)}(\mathbf{x}, -\mathbf{x}) \) is clearly slower above threshold than below threshold. The plot of \( g^{(1)}(\mathbf{r}, -\mathbf{r}) \) versus \( \mathbf{r} \) is fitted well by a Gaussian function with the standard deviation \( \sigma \) as a fitting parameter. From the fits, we obtain the coherence length \( \lambda_c = \sqrt{2\pi\sigma} \). As seen in Fig. 4f, \( \lambda_c \) increases abruptly across the threshold, from 2.38 μm near threshold to about 3 μm above threshold, confirming the formation of extended spatial coherence in the laser. Above threshold, the \( \lambda_c \) value remains largely unchanged, possibly limited by the laser spot size and carrier diffusion length. The \( \lambda_c \) value decreases slightly at the highest powers, possibly because of competition of multiple spatial modes in the absence of lateral confinement potentials. We note that the measured \( g^{(1)}(\mathbf{x}, -\mathbf{x}) \) is much lower than the actual value, owing to difficulty in achieving good alignment.

In conclusion, we have demonstrated a 2D WSe\(_2\)–MoSe\(_2\) heterobilayer laser on a grating cavity. The injected carrier density at threshold is within an order of magnitude of the estimated transparency condition, suggesting band inversion between the MoSe\(_2\) conduction band and the WSe\(_2\) valence band as the gain mechanism. The type-II band alignment, resulting in charge separation and longer exciton lifetimes, may have facilitated the establishment of a population inversion. In addition,
for heterobilayers, moiré lattices are expected. By analogy to quantum dot lasers, localization of the interlayer excitons in a moiré lattice may lead to increased phase space density in the lasing mode for the same carrier density, as well as reduced non-radiative loss of the trapped interlayer excitons, enhancing the performance of heterobilayer lasers.

Future studies may clarify the role of moiré lattices in heterobilayer lasers. The present heterobilayer laser could be improved by reducing the inhomogeneous broadening of the gain medium via encapsulation with hexagonal boron nitride, improving the cavity $Q$, and reducing the mode competition with lateral confinement of the cavity modes. Also, different combinations of van der Waals materials in the heterobilayer could be used to create interlayer exciton lasers of different wavelengths. Using cavities with lateral rotational invariance would allow a valley-polarized interlayer exciton laser to be realized. Finally, electrical tuning of the oscillator strength might allow fast modulation of the laser, and electrical injection could be implemented via atomically thin, bilayer p–n junctions. Adiabatic electrical tuning might be used to explore coherent indirect exciton gases.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-019-1779-x.

1. Mak, K. F., Lee, C., Hone, J., Shan, J. & Heinz, T. F. Atomically thin MoS$_2$: a new direct-gap semiconductor. Phys. Rev. Lett. 105, 136805 (2010).
2. Splendiani, A. et al. Emerging photoluminescence in monolayer MoS$_2$. Nano Lett. 10, 1271–1275 (2010).
3. Geim, A. K. & Grigorieva, I. V. Van der Waals heterostructures. Nature 499, 419–425 (2013).
4. Zhang, C. et al. Interlayer couplings, moiré patterns, and 2D electronic superlattices in MoS$_2$/WSe$_2$ heterobilayers. Sci. Adv. 3, e1601459 (2017).
5. Fang, H. et al. Strong interlayer coupling in van der Waals heterostructures built from single-layer chalcogenides. Proc. Natl Acad. Sci. USA 111, 6198–6202 (2014).
6. Fogler, M. M., Butov, L. V. & Novoselov, K. S. High-temperature superfluidity with indirect excitons in van der Waals heterostructures. Nat. Commun. 5, 4555 (2014).
7. Eaton, S. W., Fu, A., Wong, A. B., Ning, C. Z. & Yang, P. Semiconductor nanowire lasers. Nat. Rev. Mater. 1, 16028 (2016).
8. Noginov, M. A. et al. Demonstration of a spaser-based nanolaser. Nature 460, 1110–1112 (2009).
9. Noda, S. Seeking the ultimate nanolaser. Science 314, 260–261 (2006).
10. Wu, S. et al. Monolayer semiconductor nanocavity lasers with ultralow thresholds. Nature 520, 69–72 (2015).
11. Ye, Y. et al. Monolayer excitonic laser. Nat. Photon. 7, 733–737 (2015).
12. Salehzadeh, O., Dajdov, M., Tran, N. H., Shih, J. I. & Mi, Z. Optically pumped two-dimensional Mo$_x$S$_{1-x}$ lasers operating at room-temperature. Nano Lett. 15, 5302–5306 (2015).
13. Li, Y. et al. Room-temperature continuous-wave lasing from monolayer molybdenum ditelluride integrated with a silicon nanobeam cavity. Nat. Nanotechnol. 12, 987–992 (2017).
14. Shang, J. et al. Room-temperature 2D semiconductor activated vertical-cavity surface-emitting lasers. Nat. Commun. 8, 543 (2017).
15. Zhao, L. et al. High-temperature continuous-wave pumped lasing from large-area monolayer semiconductors grown by chemical vapor deposition. ACS Nano 12, 9390–9396 (2018).
16. Reeves, L., Wang, Y. & Krauss, T. F. 2D material microcavity light emitters: to lase or not to laser? Adv. Opt. Mater. 6, 1800272 (2018).
17. Butov, L. V., Zrenner, A., Abstreiter, G., Böhm, G. & Weimann, G. Condensation of indirect excitons in coupled AlAs/GaAs quantum wells. Phys. Rev. Lett. 73, 304–307 (1994).
18. Rigosi, A. F., Hill, H. M., Li, Y., Chernikov, A. & Heinz, T. F. Probing interlayer interactions in transition metal dichalcogenide heterostructures by optical spectroscopy: MoS$_2$/WSe$_2$ and MoS$_2$/WSe$_2$. Nat. Nanotechnol. 15, 5033–5038 (2015).
19. Zhang, L. et al. Highly valley-polarized singlet and triplet interlayer excitons in van der Waals heterostructure. Phys. Rev. B 100, 041402 (2019).
20. Lee, C. H. et al. Atomically thin p–n junctions with van der Waals heterointerfaces. Nat. Nanotechnol. 9, 676–681 (2014).
21. Ross, J. S. et al. Interlayer exciton optoelectronics in a 2D heterostructure p–n junction. Nano Lett. 17, 638–643 (2017).
22. Zhang, L., Oogna, R., Burg, W., Tutuc, E. & Deng, H. Photonic-crystal exciton-polaritons in monolayer semiconductors. Nat. Commun. 9, 713 (2018).
23. Spivak, B. & Luryi, S. in Future Trends in Microelectronics (eds Spivak, B., Luryi, S. & Zaslavsky, A.) 68–76 (Wiley-Blackwell, 2007).
24. Deng, H., Solomon, G. S., Hey, R., Ploog, K. H. & Yamamoto, Y. Spatial coherence of a polariton condensate. *Phys. Rev. Lett.* **99**, 126403 (2007).
25. Hoang, T. B., Akselrod, G. M., Yang, A., Odom, T. W. & Mikkelsen, M. H. Millimeter-scale spatial coherence from a plasmon laser. *Nano Lett.* **17**, 6690–6695 (2017).
26. Daskalakis, K. S., Maier, S. A. & Kéna-Cohen, S. Spatial coherence and stability in a disordered organic polariton condensate. *Phys. Rev. Lett.* **115**, 035301 (2015).
27. Yu, H., Liu, G.-B., Tang, J., Xu, X. & Yao, W. Moiré excitons: from programmable quantum emitter arrays to spin-orbit–coupled artificial lattices. *Sci. Adv.* **3**, e1701696 (2017).
28. Wu, F., Lovorn, T. & MacDonald, A. H. Theory of optical absorption by interlayer excitons in transition metal dichalcogenide heterobilayers. *Phys. Rev. B* **97**, 035306 (2018).
29. Tran, K. et al. Evidence for moiré excitons in van der Waals heterostructures. *Nature* **567**, 71–75 (2019).
30. Seyler, K. L. et al. Signatures of moiré-trapped valley excitons in MoSe2/WSe2 heterobilayers. *Nature* **567**, 66–70 (2019).
31. Jin, C. et al. Observation of moiré excitons in WSe2/WS2 heterostructure superlattices. *Nature* **567**, 76–80 (2019); correction **569**, E7 (2019).
32. Kirstaedter, N. et al. Low threshold, large T, injection laser emission from (InGa)As quantum dots. *Electron. Lett.* **30**, 1416-1417 (1994).
33. Shahnazaryan, V., Kyrienko, O. & Shelykh, I. A. Adiabatic preparation of a cold exciton condensate. *Phys. Rev. B* **91**, 085302 (2015).

*Publisher's note* Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

© The Author(s), under exclusive licence to Springer Nature Limited 2019
Spatial coherence measurement was performed using a retro-reflector Michelson Interferometer setup as shown in Extended Data Fig. 8. Emission rid of scattered pump laser light was sent to a 50:50 beam splitter which divided the light into two paths, the mirror path and the retro-reflector path. In order to change the time difference \( \tau \) between the two paths, the retro-reflector is mounted on a stepper motor which can have a step size as small as 50 nm (about 0.167 fs). The interference pattern was collected by a Princeton Instruments eXcelon charge-coupled camera, with intensities described by:

\[
i^{\text{mm}}(\tau) = I(\tau) + I(-\tau) + 2\sqrt{I(\tau)I(-\tau)}g^{(0)}(\tau, -\tau)\sin\left(\frac{2\pi}{\Lambda}(z - z_0)\right)
\]

Here \( I(\tau) \) and \( I(-\tau) \) are intensities from the mirror and retro-reflector arms, respectively, and are measured by blocking one of the arms of the interferometer. \( z \) is the position of the retro-reflector. \( g^{(0)}(\tau, -\tau) \) is the first-order spatial coherence for two positions separated by \( 2\tau \), and is proportional to the visibility of the interference fringe. To obtain the visibility or \( g^{(0)}(\tau, -\tau) \), we can position the retro-reflector and record the sinusoidal oscillation of \( I^{\text{mm}}(\tau) \) versus \( \tau \) at each \( r \), as shown in Fig. 4c.

Photon number

Photon occupancy per pulse \( \langle N_p(k = 0) \rangle \) was estimated from the total count rate on the detector, \( n_\text{T} \). The total integration time for angle-resolved spectra was 90 s. The two values are related by: \( I_\text{p} = \eta \rho \eta \eta n_\text{T} \) (Extended Data Fig. 9).

Here \( n_\text{T} = 10^{-7} \) is the total detection efficiency of the setup, which is independently calibrated by replacing the sample with a laser-coupled single-mode fibre, \( p = 1 \) is the number of \( k \)-space modes within the integrated region, and \( f = 80 \text{ MHz} \) is the repetition rate of the pump laser.

### Methods

#### Sample fabrication

To fabricate the grating cavity, we first grew a SiN film with a SiO2 buffer on an Si substrate using low-pressure chemical vapour deposition, then patterned it using electron beam lithography and created the grating bars by plasma dry etching. The grating parameters indicated in Fig. 1a are as follows: \( d = 1.475 \text{ nm}, t = 11.3 \text{ nm}, h = 100 \text{ nm}, A = 615 \text{ nm} \) and \( g = 50 \text{ nm} \). The individual WSe2 and MoSe2 monolayers were mechanically exfoliated onto a SiO2 substrate using PDMS (polydimethylsiloxane) polymer. The exfoliated monolayers were stacked into a heterostructure using a high-accuracy rotational alignment method. First, the MoSe2 was picked up with a PDMS/PPC (polypropylene carbonate) stamp under an optical microscope. Second, the crystal axes of MoSe2 and WSe2 were rotationally aligned to be 0° or 60° before stacking. Third, the stacked heterostructure was dropped down onto the grating cavity. Last, the polymer residue was dissolved, and the sample was annealed at 350 °C for a total of 7 h.

#### Heterobilayer twist angle

We can verify the twist angle of the heterobilayer aligned under the optical microscope by angle-dependent second-harmonic generation (SHG) measurements. Extended Data Fig. 1 shows an optical microscope image of two different samples and the corresponding angle-dependent SHG measurements. By fitting the SHG pattern with a \( \cos^2(3\theta) \) function, where \( \theta \) is the angle between the armchair direction of the monolayer and the polarization direction of the beam, we can obtain the twist angle. We did not measure the SHG for the heterobilayer before putting it on the grating, but from experience, we have found that the straight edges of exfoliated monolayers reliably correspond to the armchair axis of the crystal. Therefore, we aligned the two straight flake edges under the optical microscope.

#### Time-resolved PL

To measure the decay time of the TM emission, we used a Hamamatsu streak camera system. The emission was polarization-selected for the TM direction and sent to the streak camera. As shown in Extended Data Fig. 2, a line-cut of the streak camera spectrum was fitted with a bi-exponential function to determine the lifetime. The fitted lifetime is around 2 ns.

#### PL mapping of the heterobilayer device

The spatially resolved PL mapping of the heterobilayer device is shown in Extended Data Fig. 3. For this measurement, we use a 633-nm continuous wave (CW) laser and selected TE polarization for excitation. The sample was mounted on an Attocube ANC 300 piezo stage and scanned as needed. Spectral band-pass filters are used to select the emission from bilayer, WSe2, and MoSe2 regions.

#### Measurements of lasing characteristics

Extended Data Fig. 8 shows the schematic of the optical setup used for the angle-resolved PL reflection and the coherence measurements of the heterobilayer laser device. The sample was cooled to 5 K using a Montana Instruments Fusion 2 cryostat. Fourier-space imaging was used to measure angle-resolved reflection and micro-PL of the device. For reflection, a tungsten halogen lamp was used. For micro-PL, a pulsed Ti:sapphire laser (80 MHz repetition rate, 150 fs pulse width) near-resonant with the WSe2 A-exciton (1.7 eV) was used to excite the sample. The emission was collected using a 0.42 NA objective lens, passed through a long-pass filter to filter out the excitation laser and a linear polarizer to selectively measure TE and TM modes, and sent to a Princeton Instruments spectrometer with a measured spectral resolution of 0.3 nm. The entrance slit of the spectrometer is aligned along the y direction. The slit width of 100 μm corresponds to a range \( |k_y| < 0.13 \text{ μm}^{-1} \). The NA of the collection optics corresponds to a range of \( |k_x| < 2 \text{ μm}^{-1} \).
\( n_n = 8 \times 10^{10} \text{ cm}^{-2} \), which is in good agreement with the threshold carrier density \( n_{th} = 5.7 \times 10^{10} \text{ cm}^{-2} \).

**Simplified rate equation model of the laser**

We use the following rate equations to describe the time evolution of interlayer exciton density \( N \) and the photon density \( S \) in the lasing mode:

\[
\frac{dN}{dt} = \frac{\eta P}{\hbar \omega V_s \tau_p} - \frac{F \beta_0 N}{\tau_{sp}} - \frac{\alpha_{vg} (N - N_c) S}{\tau_{av}} \tag{7}
\]

\[
\frac{dS}{dt} = \frac{F \beta_0 N}{\tau_{sp}} + \frac{\alpha_{vg} (N - N_c) S}{\tau_{av}} - \frac{S}{\tau_p} \tag{8}
\]

Parameters in the equation are listed in Extended Data Table 1. In the heterobilayer system, electron and hole transfer takes place on the subpicosecond timescale, much shorter than the interlayer exciton lifetime. Therefore, we consider a pulsed pump \( P \) that creates an initial carrier population of \( N(t=0) \propto P \). The photon number \( I_p \) is proportional to the photon density, and \( I_p = 1 \) at threshold. The simulated curve of \( I_p \) versus pump power matched the experiment well, as shown in Extended Data Fig. 6.
Extended Data Fig. 1 | Heterobilayer twist angle.  

**a, b.** Optical image (a) and angle-resolved SHG measurements (b; open circles) of WSe$_2$/MoSe$_2$ heterobilayers. The field of view of the optical images is around 60 μm. Solid lines in b, d, are fits by a cos$^2$(3θ) function, which give relative twist angles of 0.22° ± 1.78° for b, and 0.34° ± 1.5° for d.
Extended Data Fig. 2 | Interlayer exciton lifetime. **a**, Time-resolved PL spectrum for TM emission. **b**, Line-cut of **a** near 1.38 eV. Red line is a bi-exponential fit to the data, with a fitted lifetime of 2 ns.
Extended Data Fig. 3 | Spatial mapping of PL. The normalized intensity of PL from the device is shown as a function of position. Spectral filters centred around their respective exciton peak energies were applied for each image. The white contours mark the regions of heterobilayer (a), MoSe₂ (b) and WSe₂ (c).
Extended Data Fig. 4 | Electric field profiles. Shown are simulated normalized electric field profiles as a function of position near the centre of a grating cavity with lateral dimensions of 100 μm × 100 μm. a, TE-polarized light at the cavity resonance at $k = 0$, showing strong field enhancement in the grating layer including at its surface where the heterobilayer is placed. b, TM-polarized light at the same wavelength as a, showing negligible cavity effects. White lines outline different layers of the grating cavity. The corresponding enhancement of the exciton radiative decay rate, or the Purcell factor, is calculated to be around 2.4.
Extended Data Fig. 5 | Power-dependence reproducibility. The photon occupancy (red) and linewidth (blue) of TE emission from the heterobilayer versus input pump power, similar to that shown in Fig. 3b of the main text but measured on a different day to show the reproducibility of the device. The error bars on the photon occupancy data include the shot noise and detector read noise. The error bars on the linewidth data correspond to the 95% confidence interval of the Lorentzian fit.
Extended Data Fig. 6 | Rate equation fitting. The log-log plot of photon occupancy versus pump power. The diamonds represent measured data shown in Fig. 3b of the main text, and the solid line is a rate-equation fitting. Details of the rate equation simulation are described in Methods.
Extended Data Fig. 7 | Temperature dependence. Temperature-dependent real space PL spectra of the sample studied in the main text.
Extended Data Fig. 8 | Experimental setup. Schematic diagram of the optical experimental setup as described in Methods. BS, beam splitter; LP, long-pass filter; BP, band-pass filter; CCD, charge-coupled device.
Extended Data Fig. 9 | Temporal coherence. Shown are temporal coherence interference fringes at $g^{(1)}(\tau=0)$ measured using a two-retro-reflector Michelson interferometer under CW excitation above threshold.

**a.** Interferogram image at $\tau=0$. **b.** Horizontal line-cut of a around $y=1.5\,\mu m$. The red line is a fit to the Gaussian pump beam profile modulated by a cosine function. Here $g^{(1)}(\tau=0) = 0.78$. 
# Extended Data Table 1 | Rate equation fitting parameters

| Parameter     | Definition                                     | Value          |
|---------------|------------------------------------------------|----------------|
| $F$           | Purocell factor                                | 2.35           |
| $\Gamma$      | Confinement factor                             | 0.0208         |
| $V_s$         | Carrier injection volume                       | $1.4 \times 10^3 \mu m^3$ |
| $\tau_{sp}$   | Spontaneous emission lifetime                  | 2 ns           |
| $\tau_p$      | Photon lifetime                                | 0.3 ps         |
| $\eta$        | Absorption efficiency                          | 20 %           |
| $N_{tr}$      | Transparency density                           | $8 \times 10^{10} \text{cm}^{-2}$ |
| $\beta_0$     | Spontaneous emission factor                    | 0.046          |
| $\alpha$      | Absorption cross section                       | $1.9 \times 10^{-14} \text{cm}^2$ |
| $v_g$         | Group velocity                                 | $1.5 \times 10^6 \text{m/s}$ |

Definitions and values of the parameters used in the rate equation simulation. The values of $F$, $\Gamma$ and $V_s$ are estimates for our structure; $N_{tr}$ is estimated in Methods; measured values are used for $\tau_{sp}$, $\tau_p$ and $\eta$; $\beta_0$ and $\alpha$ are fitting parameters. See Methods for definition of the rate equation using these parameters.