Evidence of high-temperature exciton condensation in two-dimensional atomic double layers

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A Bose–Einstein condensate is the ground state of a dilute gas of bosons, such as atoms cooled to temperatures close to absolute zero1. With much smaller mass, excitons (bound electron–hole pairs) are expected to condense at considerably higher temperatures2–7. Two-dimensional van der Waals semiconductors with very strong exciton binding are ideal systems for the study of high-temperature exciton condensation. Here we study electrically generated interlayer excitons in MoSe2–WSe2 atomic double layers with a density of up to 10^{12} excitons per square centimetre. The interlayer tunnelling current depends only on the exciton density, which is indicative of correlated electron–hole pair tunnelling8. Strong electroluminescence arises when a hole tunnels from WSe2 to recombine with an electron in MoSe2. We observe a critical threshold dependence of the electroluminescence intensity on exciton density, accompanied by super-Poissonian photon statistics near the threshold, and a large electroluminescence enhancement with a narrow peak at equal electron and hole densities. The phenomenon persists above 100 kelvin, which is consistent with the predicted critical condensation temperature9–12. Our study provides evidence for interlayer exciton condensation in two-dimensional atomic double layers and opens up opportunities for exploring condensate-based optoelectronics and exciton-mediated high-temperature superconductivity13.

Exciton condensation is a macroscopic quantum phenomenon that has attracted tremendous theoretical and experimental interest. Condensed phases of excitons have been generated by optical pumping or via quantum Hall states under a magnetic field in coupled semiconductor quantum wells and graphene15–16. The weak exciton binding in these systems, however, limits the condensation temperature to about 1 K. Although a high-temperature exciton condensate has been reported in octahedral titanium diselenide17 (1T-TiSe2), systems based on a three-dimensional semimetal have limited configurability and applicability to future devices.

Two-dimensional (2D) transition metal dichalcogenide (TMD) semiconductors (MX2, M = Mo, W; X = S, Se) with large exciton binding energy (about 0.5 eV)18 and flexibility in forming van der Waals heterostructures provide an exciting platform for exploring high-temperature exciton condensation and condensate-based applications9–12. The maximum condensation temperature in TMD double layers, limited by exciton ionization in the high-density regime, has been predicted to be a fraction (about 10%) of the exciton binding energy9–12—that is, comparable to room temperature. Condensation of intralayer excitons in TMDs is, however, hindered by the short exciton lifetimes18 and the formation of competing exciton complexes at high densities, such as biexcitons18 and electron–hole droplets19. These difficulties can be overcome

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**Fig. 1** | Electrical generation of high-density exciton gases. a, b, Schematic (a) and optical micrograph (b) of a MoSe2–WSe2 electron–hole double-layer device. G, few-layer graphene. c, d, The lower panels show contour plots of the reflection contrast 6R/ΔR as a function of photon energy E and gate voltage V_{gate} at bias voltages of 0 V (c) and 5.5 V (d) for device 1. A gap is opened for the MoSe2 and WSe2 neutral exciton resonances under finite bias (between the white dashed lines). The circled bright spot in d is electroluminescence from the tunnel junction. The results are consistent with a type-II band alignment of the double layer (upper panels).
by separating the electrons and holes into two closely spaced layers in a double-layer structure. Long lifetimes and substantial binding energies (>0.1 eV) have been demonstrated for interlayer excitons. Further, they act like oriented electric dipoles, with repulsive interactions that prevent the formation of competing exciton complexes at high densities. Nevertheless, exciton condensation remains elusive.

Here we present experimental evidence of high-temperature interlayer exciton condensation in MoSe2–WSe2 double layers. The device (Fig. 1a) consists of two angle-aligned TMD monolayer crystals.

![Fig. 2](image-url) | Tunnelling characteristics and correlated pair tunnelling. a, b, Tunnelling current $I$ of the double layer as a function of gate voltage for varying bias voltage (a) and as a function of bias voltage for varying gate voltage (b) for device 2. c, d, The corresponding tunnelling characteristics, simulated based on $I = \alpha N^\beta$, where $N$ is the exciton density and $\alpha$ and $\beta$ are the free fitting parameters for the entire family of dependences. The best-fit result for exponent $\beta$ is 2.9.

![Fig. 3](image-url) | Threshold dependence of electroluminescence on exciton density at 3.5 K. a, Electroluminescence spectra at varying exciton densities (at charge balance) for device 2. b, Electroluminescence intensity correlation function at the same exciton densities as in a. Dashed lines denote $g^{(2)}(r) = 1$. The vertical bar denotes the y-axis scale. The curves in a, b are displaced vertically for clarity. c–e, Exciton density dependence of the integrated electroluminescence intensity and tunnelling current (c), the electroluminescence intensity correlation at zero time delay (d) and the electroluminescence and tunnelling density width (e). The density widths are extracted from the data in Fig. 4 as described in the text.
region (between the two horizontal dashed lines in Fig. 1d) opens up, electron doping at large positive gate. This is consistent with a type-II band alignment in which MoSe₂ is electron-doped (with electron density $n > 0$) and WSe₂ is hole-doped (with hole density $p > 0$). The bias voltage creates an interlayer electrochemical potential difference and an electron–hole double layer. The problem is well described by an electrostatic model (see Methods and Extended Data Fig. 6) that assumes the total charge density, $n + p$, and the charge density imbalance, $n - p$, to be given by $V_{\text{bias}}$ and by $2V_{\text{gate}} - V_{\text{bias}}$, respectively. The electron–hole pair density $N$ (the smaller of $n$ and $p$) is thus given by $V_{\text{bias}} - V_{\text{gate}}$ (for $n > p$) or $V_{\text{gate}}$ (for $n < p$). Very high pair densities of up to $10^{12}$ cm⁻² have been achieved.

Radiative recombination from interlayer excitons is completely suppressed in devices with a hBN barrier thicker than one layer owing to negligible electron–hole wave function overlap⁹ (Extended Data Fig. 7). We employ two other probes to study the electron–hole double layer. The large $V_{\text{bias}}$ creates a microamperes-level tunnelling current $I$ between the layers, which makes the exciton gas a steady-state non-equilibrium system (see Methods and ref. ¹⁰). Enhanced tunnelling is expected if the electrons and holes are bound⁹ (that is, form interlayer excitons). However, the large bias that is needed to open a p–n region excludes the possibility of observing any zero-bias Josephson-like effects in the exciton condensate⁵. Electroluminescence is also observed near 1.6 eV (the bright spot in Fig. 1d), which matches the energy of a charged exciton in MoSe₂ (see electroluminescence image in Extended Data Fig. 1). The electroluminescence arises as a hole tunnelling from WSe₂ to MoSe₂ and recombines radiatively with an electron in MoSe₂ (see Methods). The maximum electroluminescence quantum efficiency is of the order of $10^{-4}$. Electroluminescence resulting from the recombination of an electron tunnelled from MoSe₂ with a hole in WSe₂ is also observed; it is typically much weaker, presumably owing to the presence of lower-energy dark exciton states in WSe₂, and it is not monitored. The measured electroluminescence intensity is directly proportional to the radiative decay rate of a hole in n-doped MoSe₂, which—unlike the tunnelling current under large bias—is sensitive to the emergence of a condensate. A large enhancement in electroluminescence has indeed been reported in a similar situation when a hole recombines radiatively with an electron in a superconductor upon cooling below the critical temperature¹²,¹³.

Unless otherwise specified, all measurements presented here were performed at 3.5 K with two devices. Data for device 1 are shown in Figs. 1, 5 and Extended Data Figs. 1, 2, 4–6, and the rest of the figures show results for device 2.

Figure 2 illustrates the tunnelling characteristics of the double layer. For a fixed $V_{\text{bias}}$ (that is, constant density $n + p$), the gate dependence of $I$ shows a cusp-shaped peak centred at charge balance ($n = p$) and the current on the cusp increases with $V_{\text{bias}}$ (Fig. 2a). Similarly, for a fixed $V_{\text{gate}}$ (Fig. 2b), $I$ increases with increasing $V_{\text{bias}}$ and approaches a cusp at charge balance. Beyond this point, $I$ becomes a constant, the value of which increases with increasing $V_{\text{gate}}$. We note that in this region ($n < p$), the electron–hole pair density is also a constant, hinting that $I$ depends only on $N$. Indeed, the simulated tunnelling characteristics obtained using $I \propto N^{2.9}$ (Fig. 2c, d) are in excellent agreement with the experiment (Fig. 2a, b). Negligible temperature dependences for $I$ were observed for the entire temperature range studied in this work (3.5–180 K) (Fig. 5a).

The above observations for tunnelling are not consistent with the picture of non-interacting electrons and holes, in which $I$ is determined by the number of states available for tunnelling, which is proportional to $n + p$ (Fig. 1d). The gate dependence of $I$ for a given $V_{\text{bias}}$ would be flat inside the p–n region instead of exhibiting a cusp-shaped peak (Fig. 2a).

Tunnelling here is consistent with correlated electron–hole pair tunnelling⁶ involving the creation and annihilation of bound electron–hole pairs. The tunnelling current is thus only dependent on pair density⁸.

The absence of temperature dependence for $I$ is also consistent with the large exciton binding energy (>0.1 eV, equivalent to 1,200 K) predicted for interlayer excitons with hBN separation of about 1 nm (refs ⁹,²⁵).

Next we turn to the electroluminescence measurements. Figure 3a displays the electroluminescence spectra for different exciton densities separated by a two- to three-layer hexagonal boron nitride (hBN) tunnel barrier. The barrier suppresses interlayer electron–hole recombination to achieve high exciton density while maintaining strong binding⁹–¹². The double layer is gated on both sides with symmetric gates that are made of few-layer graphene gate electrodes and 20–30-nm hBN gate dielectrics. Figure 1b shows an optical image of a typical device. That are made of few-layer graphene gate electrodes and 20–30-nm hBN gate dielectrics. Figure 1b shows an optical image of a typical device. The lateral size is on the micrometre scale. By applying equal voltages ($V_{\text{gate}}$) to the two gates and a bias voltage ($V_{\text{bias}}$) to the WSe₂ layer, one can tune the carrier density in each TMD layer independently, in contrast to optical pumping. In addition, this method can create interlayer excitons at a temperature nearly identical to that of the lattice, which favours condensation.

Figure 1c (lower panel) shows a contour plot of the optical reflection contrast spectra of the double layer as a function of $V_{\text{gate}}$ for $V_{\text{bias}} = 0$ V. The two prominent features correspond to the neutral exciton resonance of monolayer MoSe₂ and WSe₂, which lose their oscillator strengths rapidly upon doping¹⁴. The sharpness of the full edge provides an estimate of the disorder density in each monolayer to be about $10^{13}$ cm⁻², which is consistent with the highest reported quality of monolayer TMD crystals²¹. For a wide range of $V_{\text{gate}}$ values, both neutral exciton resonances are present; the feature disappears in WSe₂ with hole doping at large negative $V_{\text{gate}}$ and in MoSe₂ with electron doping at large positive $V_{\text{gate}}$. This is consistent with a type-II band alignment of the heterostructure²⁶. By contrast, for $V_{\text{bias}} = 5.5$ V, a p–n region (between the two horizontal dashed lines in Fig. 1d) opens up,
Temperature dependence of electroluminescence. a, Dependance of the electroluminescence intensity (left axis) and tunnelling current (right axis) on charge imbalance at various temperatures for a fixed exciton density above threshold ($0.74 \times 10^{12}$ cm$^{-2}$) for device 1. The electroluminescence enhancement at charge balance disappears above 100 K. b, Temperature dependence of the relative electroluminescence enhancement at charge balance above the baseline for $N = 0.74 \times 10^{12}$ cm$^{-2}$ (top), $0.84 \times 10^{12}$ cm$^{-2}$ (middle) and $0.90 \times 10^{12}$ cm$^{-2}$ (bottom). Typical errors for the entire temperature range for each density were estimated from the standard deviations of the electroluminescence intensities. The transition temperature was estimated from the value at which the enhancement disappears (dashed lines).

The threshold behaviour is accompanied by a change in photon statistics revealed by an intensity correlation measurement conducted using a Hanbury Brown–Twiss-type setup (see Methods). The threshold dependence of the electroluminescence intensity is consistent with a critical threshold dependence on $N$, with the threshold at $N_{th} \approx 0.26 \times 10^{12}$ cm$^{-2}$ (Fig. 3c). In a narrow range around $N_{th}$, the electroluminescence intensity increases by two orders of magnitude, whereas the tunnelling current changes by a factor of only about 2.

The threshold behaviour is accompanied by a change in photon statistics revealed by an intensity correlation measurement conducted using a Hanbury Brown–Twiss-type setup (see Methods). The time resolution of the setup is about 40 ps, which exceeds by far the electroluminescence coherence time (about 100 fs) estimated from the spectral width. Figure 3b displays the electroluminescence intensity correlation function $g^{(2)}(\tau)$ for different $N$ values (at charge balance), where $\tau$ is the difference in arrival time between pairs of electroluminescence photons. Photon bunching ($g^{(2)}(0) > 1$) is observed near the threshold with a decay time of about 1 ns. Above the threshold, photon bunching is absent ($g^{(2)}(\tau) = 1$) and the electroluminescence statistics is Poissonian.

The electroluminescence enhancement is very sensitive to charge imbalance in the double layer. Figure 4 shows the electroluminescence intensity as a function of $n - p$ at several fixed values of $n + p$ ($n + p = 2N$ at charge balance). The tunnelling current is included as a reference. For $N \geq N_{th}$, similarly to the current, the electroluminescence shows a cusp-shaped peak centred at charge balance, but its enhancement occurs in a much narrower range than $I$. For $N < N_{th}$, the electroluminescence intensity does not follow the current strictly and does not have a cusp. Near the threshold, the electroluminescence is particularly sensitive to charge imbalance both in terms of intensity and photon statistics (Extended Data Fig. 3). In Fig. 3e, we use the normalized 'density width' (the full-width at half-maximum of the peak, $\delta n$, divided by the total density), $\delta n/(2N)$, to quantify the sensitivity of the two processes to charge imbalance. Whereas the current density width is nearly independent of $N$, the electroluminescence density width decreases sharply at the threshold and remains substantially smaller than the current density width. Similar electroluminescence characteristics have been observed in other devices. Extended Data Figs. 4, 5 show results from device 1 with a higher $N_{th}$. The observed threshold behaviour of electroluminescence as a function of exciton density, the photon bunching at the threshold and absence of bunching above it, and the high sensitivity of electroluminescence to charge imbalance are not compatible with the behaviour of conventional light-emitting diodes, which involves tunnelling and recombination of independent charge particles. In such a picture, the electroluminescence intensity is proportional to the current before reaching saturation and none of these observations can be explained. The threshold behaviour of electroluminescence is analogous to lasing and polariton condensation, which are known non-equilibrium continuous phase transitions.$^{6,26}$ In these processes, the cavity photons or the polaritons condense into a single electromagnetic mode above the threshold. Photon bunching arises near the threshold owing to critical electromagnetic fluctuations, which disappear above the threshold in the condensed phase. However, in the absence of an optical cavity that provides feedback, electroluminescence in our devices is not lasing or polariton condensation.

Instead, the threshold behaviour of electroluminescence is consistent with a continuous phase transition at the critical density $N_{th}$ from an exciton gas to an exciton condensate, whose wave function consists of spatially coherent electron–hole pairs.$^{3,10}$ When a ‘normal’ hole recombines radiatively with an exciton in MoSe$_2$, that is a part of a condensate, the recombination rate increases with the number of bosons in the condensate,$^{23}$ that is, the recombination becomes a superradiant process.$^{4,5}$ (see the electroluminescence rate analysis in Methods). This
conclusion is further supported by the absence of electroluminescence threshold behaviour in angle-misaligned double layers (Extended Data Fig. 8), in which interlayer excitons with large momentum are formed. The large exciton momentum does not favour condensation. The observed photon bunching with a correlation time much longer than the coherence time near the threshold corresponds to critical fluctuations and slowing down at the critical point26–27. The absence of photon bunching above the threshold corresponds to suppressed noise in the condensed phase. The observed strong sensitivity of the electroluminescence enhancement to charge imbalance above the threshold is also consistent with exciton condensation, which requires nearly perfect electron–hole Fermi surface nesting28–30. In particular, a non-analytic cusp at charge balance as that in Fig. 4 was predicted for exciton coherence as a function of charge imbalance28–30. Such sensitivity to charge imbalance disappears below the threshold as expected.

Finally, we estimate the transition temperature for exciton condensation. Figure 5a displays the electroluminescence intensity and tunneling current as a function of charge imbalance at various temperatures for a fixed pair density above the threshold (0.74 × 10^{12} \text{ cm}^{-2}). The tunneling current has a negligible dependence on charge imbalance or temperature T, as discussed above (Fig. 2b). By contrast, both the electroluminescence intensity and electroluminescence enhancement at charge balance decrease with increasing temperature. To exclude any potential trivial thermal effects, we use the relative electroluminescence enhancement at charge balance above the baseline to estimate the transition temperature. The top panel in Fig. 5b corresponds to the result for N = 0.74 × 10^{12} \text{ cm}^{-2}. For T > 100 K, the enhancement disappears (that is, the electroluminescence behaviour of the device returns to that of a normal light-emmitting diode). This value is very close to the predicted degeneracy temperature (onset of the macroscopic occupation of the ground state) for interlayer excitons in TMD double layers separated by two-layer hBN2. Figure 5b also shows similar results for two other exciton densities. No clear N dependence of the transition temperature can be concluded for the small range of densities investigated in this study. Although a systematic investigation of such N dependence is needed to test different theories (linear2–1 or more complicated dependence12), it is beyond the scope of this study. We also note that further improvement of the devices (in terms of electrical contact, as well as sample size and quality) is required to directly probe exciton superfluidity (for example, using a counterflow setup13) and long-range spatial coherence13–33.

In conclusion, we have electrically created a high-density electron–hole double layer based on 2D van der Waals heterostructures under zero magnetic field. By combining tunnelling and electroluminescence measurements, we have observed that the electroluminescence exhibits a threshold dependence on exciton density, a sensitive dependence on charge imbalance, and critical fluctuations, which are consistent with exciton condensation. These observations persist up to about 100 K. Our results open up opportunities to explore exciton condensates at high-energy scales9–12 and exciton-mediated high-temperature superconductivity13–33.

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METHODS

Device fabrication. Dual-gated WSe₂/hBN/MoSe₂ tunnel junctions were fabricated using the dry-transfer method developed by Wang et al. Each constituent layer of the junction and the gates was exfoliated from bulk crystals onto Si substrates with a 300-nm oxide layer. High-quality WSe₂ and MoSe₂ bulk crystals were synthesized with the flux growth technique. Thin flakes of appropriate thickness and size were identified optically. In particular, the dark-field imaging mode was used to enhance the optical contrast for few-layer hBN. The hBN tunnel barrier was about 2–3 layers thick (0.6–1.0 nm), as confirmed by atomic force microscopy. The identified flakes were picked up layer by layer by a stamp made of a thin layer of polycarbonate on polydimethylsiloxane supported by a glass slide. The finished stack was thermally released onto a Si substrate with pre-patterned electrodes for the source, drain and top and bottom gates. Finally the polycarbonate film was removed using chloroform. The schematic and the optical image of a finished device are shown in Fig. 1a, b.

Reflection contrast and electroluminescence measurements. Reflection contrast measurements were conducted to calibrate the doping density of the WSe₂ and MoSe₂ monolayers. The output of a broadband super-continuum light source was focused by a high-numerical-aperture (0.8) objective to a spot of about 1 μm² on the device. The reflected light was collected by the same objective, dispersed by a grating spectrometer and detected by a charge-coupled-device camera. The reflection contrast R/R₀ was obtained by normalizing the difference between the light intensity reflected from the tunnel junction and the substrate (R₀) to that reflected from the substrate (R). Electroluminescence from the tunnel junction was detected by the same optics. The typical integration time for electroluminescence was 0.1 s. A Hanbury Brown–Twiss setup was used to measure the electroluminescence intensity correlation function g²(τ). The collected electroluminescence from the tunnel junction was split by a 50:50 beam splitter and focused onto two identical single-photon detectors. The detector outputs were fed to a time-correlated single-photon counter (Picocarpp 300) to record the arrival time difference between a pair of electroluminescence photons. A histogram of the number of photons versus the arrival difference time and the intensity correlation function can then be obtained. The instrument response time was about 40 ps.

Electrostatics of the TMD double layer structure. The equivalent circuit of the TMD tunnel junction is a tunnel resistor connecting the bottom and top gates. The TMD bilayers are coupled and the junction capacitance can be ignored (compared with the gate capacitance, Cgate) under large bias beyond the breakdown voltage. The energy band alignment is shown in Fig. 1. For symmetric gating, as in our experiment, we express the electron–hole density imbalance (n − p) and the total density (n + p) for the coupled bilayer as

\[
(n - p) = C_{\text{gate}}(2V_{\text{gate}} - V_{\text{bias}} - V_{\text{off}} + V_{W})
\]

\[
(n + p) = C_{\text{gate}}(V_{\text{bias}} - V_{\text{off}} - V_{W})
\]

Here n > 0 is the electron density in the MoSe₂ layer and p > 0 is the hole density in the WSe₂ layer, e is the elementary charge and Voff = (Voff₁ + Voff₂)/2 \text{(V)}}\text{ is the potential required to move the in-gap Fermi level to the conduction band minimum of MoSe₂ (the valence band maximum of WSe₂). The gate capacitance Cgate = \text{(0.89–1.33)} \times 10^{-2} \text{Fcm}^{-2} \text{ is determined by the thickness of the hBN dielectric (20–30 nm) and its dielectric constant}^{2,35} \text{ (about 3). The density imbalance is controlled by } 2V_{\text{gate}} - V_{\text{bias}} \text{ and the total density by } V_{\text{bias}}. \text{The exciton density } N_{\text{e}} \text{ can be obtained as}

\[
N_{\text{e}} = \begin{cases} 
C_{\text{gate}}(V_{\text{bias}} - V_{\text{off}} - V_{W}) & \text{for } n > p \\
C_{\text{gate}}(V_{\text{off}} - V_{W}) & \text{for } n < p
\end{cases}
\]

We compare the electrostatic model with the experiment in Extended Data Fig. 6 for three special cases: n = p, n = 0 and p = 0

\[
V_{\text{gate}} = \frac{1}{2}(V_{\text{bias}} + V_{\text{off}} - V_{W}) \text{ for } n = p
\]

\[
V_{\text{gate}} = V_{\text{off}} \text{ for } n = 0
\]

\[
V_{\text{gate}} = V_{\text{bias}} - V_{\text{off}} \text{ for } p = 0
\]

The gate voltage for n = p can be unambiguously determined in the experiment from the cusps of the tunnelling current. The gate voltages for n = 0 and p = 0 can be determined from the reflection contrast of MoSe₂ and WSe₂. We assume that when the electrochemical potential touches the band edge, the reflection contrast of the corresponding neutral exciton resonance drops to 90% of its maximum value. We obtain V_{\text{off}}₁ ≈ 2.3 V and V_{\text{off}}₂ ≈ 2.0 V from the best fit in Extended Data Fig. 6b.

Electroluminescence rate equation and spectral width. The rate equation for the minority hole density p₀ in the n-doped MoSe₂ layer can be expressed as

\[
\frac{dp}{dt} = \frac{I}{2eC} - \frac{p}{\tau_p}
\]

The first term is the hole pumping rate with I denoting the tunnelling current and A the tunnel junction area. Because tunnelling here is a correlated pairwise process (Fig. 2), the hole pumping rate is determined by half of the tunnelling current, I/2. The second term is the total hole decay rate in MoSe₂ with γ₀ = γₚ + γₜ₀ + γₜ, where γₚ and γₜ represent contributions from radiative and non-radiative recombination in MoSe₂ and tunnelling out to the electrode, respectively. In the steady state dp/dt = 0, and equation (6) yields p₀ = I/(2eCγ₀p). The integrated electroluminescence power normalized by the tunnelling current is thus determined by the ratio of the radiative rate to the total decay rate

\[
\frac{\gamma_{\text{APE}}}{\gamma_0 \gamma_{\text{APE}}} = \frac{1}{2eC_e}
\]

Here E₀ is the electroluminescence photon energy (E₀ ≈ 1.6 eV). In the vicinity of exciton condensation, the large enhancement observed in the normalized electroluminescence power (Fig. 3c) is dominated by the enhancement in γₑ because the electroluminescence quantum yield is typically very small (that is, γₑγₚγₜ ≪ 1). The radiative recombination rate γₑ is determined by the transition dipole matrix element between the initial and final states, which are many–body quantum states in the exciton condensate phase. The radiative rate is enhanced by the number of spatially coherent excitons in the condensate, similarly to the phenomenon of superradiance. As in lasing and polariton condensation, our system is in a steady-state non-equilibrium. Unlike in lasing and polariton condensation, however, considerable narrowing of electroluminescence spectral lines does not necessarily accompany the dependence of the threshold intensity on the exciton density. In lasing or polariton condensation, although the carrier decay rate, thermal broadening and inhomogeneous broadening contribute to the spectral width below the threshold, the spectral width above the threshold is determined only by the photon decay rate in the cavity, which is typically much smaller, giving rise to substantial spectral line narrowing. In the absence of an optical cavity, the electroluminescence spectral width below and above the threshold in our devices is mainly determined by γₜ, which is not necessarily greatly suppressed above the threshold. There is therefore enhanced spatial coherence but not necessarily much enhanced temporal coherence in the condensed phase. The situation is similar to the case of a hole recombining radiatively with an electron in a superconductor, where substantial enhanced electroluminescence intensity due to the formation of a Cooper pair condensate was observed without considerable line narrowing below the critical temperature.

Data availability

The data shown in the figures and other findings of this study are available from the corresponding authors upon reasonable request.

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Author contributions Z.W. fabricated the devices, developed the experimental setup and performed the measurements. D.A.R. and J.C.H. grew the bulk TMD crystals, and K.W. and T.T. grew the bulk hBN crystals. K.F.M., J.S. and Z.W. discussed the results and commented on the manuscript.

Competing interests The authors declare no competing interests.

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Extended Data Fig. 1 | Electroluminescence image. Optical reflection image (left) and electroluminescence image (right) of device 1. The latter was measured at a fixed exciton density above threshold ($N = 0.79 \times 10^2 \ \text{cm}^{-2}$). The colour bar shows the number of electroluminescence photons.
Extended Data Fig. 2 | Analysis of electroluminescence spectra.

a. Electroluminescence (EL) spectra at different charge imbalance values. The total density is fixed at $1.34 \times 10^{12}$ cm$^{-2}$.

b, c. Energy-averaged electroluminescence peak energy (b) and linewidth (c) as a function of normalized density imbalance, $(n - p)/(n + p)$, for three fixed total charge densities. The linewidth is minimum at charge balance.

d, e. Energy-averaged electroluminescence peak energy (d) and linewidth (e) as a function of exciton density $N$ at charge balance. The dashed lines mark $N_0$. Although both the electroluminescence peak energy and linewidth depend on $N$, no clear threshold behaviour is seen.
Extended Data Fig. 3 | Effect of charge imbalance on $g^{(2)}(0)$.

Electroluminescence intensity (upper panel, left axis), intensity correlation at zero delay time $g^{(2)}(0)$ (upper panel, right axis) and normalized neutral exciton reflection contrast of the double layer (lower panel) as a function of charge imbalance near the threshold. The total charge density is fixed at $0.51 \times 10^{12} \text{ cm}^{-2}$. Photon bunching can only be observed near charge balance.
Extended Data Fig. 4 | Additional results for device 1 (basic characterization). a, Normalized neutral exciton reflection contrast of the double layer as a function of charge imbalance at different total densities. b, The corresponding electroluminescence intensity (left) and tunnelling current (right) as a function of charge imbalance.
Extended Data Fig. 5 | Additional results for device 1 (electroluminescence). a, Intensity correlation function $g^{(2)}(\tau)$ at various exciton densities. Curves are displaced vertically for clarity. Clear non-Poissonian statistics is seen near the threshold. Correlation with a longer decay time and extra oscillations are observed in this device compared to device 2. Further investigations are required to understand the microscopic mechanism responsible for these differences. b, Dependence of electroluminescence intensity and $g^{(2)}(0)$ on exciton density. c, Density imbalance dependence of the electroluminescence intensity and $g^{(2)}(0)$ (upper panel) and the normalized neutral exciton reflection contrast (lower panel) near the threshold. The total density is fixed at $1.18 \times 10^{12} \text{ cm}^{-2}$. Photon bunching is observed only near charge balance.
Extended Data Fig. 6 | Comparison between experiment and electrostatic model. **a**, Dependence of the normalized reflection contrast of the neutral exciton resonance in MoSe$_2$ (dashed lines) and WSe$_2$ (solid lines) monolayers on gate voltage for different bias voltages. When the TMDs are doped, the reflection contrast decreases like a broadened step function, reflecting the reduced oscillation strength of the neutral excitons. **b**, Bias and gate voltage combinations corresponding to $n = p$ (grey symbols), $p = 0$ (red symbols) and $n = 0$ (blue symbols). The solid lines are a guide to the eye. The dashed lines are predictions of the electrostatic model (see equations (3)–(5)).
Extended Data Fig. 7 | Photoluminescence. 

a, Contour plot of photoluminescence (PL) spectra versus gate voltage in a double-layer device with a three-layer hBN barrier (the photoluminescence intensity is shown on a logarithmic scale). Interlayer exciton photoluminescence (around 1.4 eV) is absent owing to the negligible oscillator strength.

b, Photoluminescence spectra at three representative gate voltages.
Extended Data Fig. 8 | Dependence of electroluminescence on MoSe₂–WSe₂ alignment angle. a, b, Dependence of electroluminescence intensity on exciton density (a) and on temperature (b) at charge balance for two different types of devices, one with a small (2°–5°) misalignment and the other with a large (20°) misalignment. The temperature in a is 4 K and the exciton density in b is about $0.74 \times 10^{12}$ cm$^{-2}$. 