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Valley-polarized exciton currents in a van der Waals heterostructure

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Abstract

Valleytronics is an appealing alternative to conventional charge-based electronics which aims at encoding data in the valley degree of freedom, i.e. the information over which extreme of the conduction or valence band carriers are occupying. The ability to create and control valley-currents in solid state devices could therefore enable new paradigms for information processing. Transition metal dichalcogenides (TMDCs) are a promising platform for valleytronics, due to the presence of two inequivalent valleys with spin-valley locking¹ and a direct band gap^{2,3}, which allows optical initialization and readout of the valley-state^{4,5}. Recent progresses on the control of interlayer excitons in these materials^{6–8} could offer an effective way to realize optoelectronic devices based on the valley degree of freedom. Here, we show the generation and transport over mesoscopic distances of valley-polarized excitons in a device based on a type-II TMDC heterostructure. Engineering of the interlayer coupling results in enhanced diffusion of valley-polarized excitons, which can be controlled and switched electrically. Furthermore, using electrostatic traps, we can increase exciton concentration by an order of magnitude, reaching densities in the order of 10^{12} cm^{-2} , opening the route to achieving a coherent quantum state of valley-polarized excitons via Bose-Einstein condensation.

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Contributions

A.K. initiated and supervised the project. A.C. fabricated the devices, D.U. performed optical measurements with assistance of A.C. A.C. and D.U. analysed the data. Z.S., A.C. and D.U. performed SHG measurements. K.W. and T.T. grew the h-BN crystals. A.C., D.U., A.A. and A.K. wrote the manuscript with input from all authors.

Competing Interests

The authors declare no competing interests.

Data Availability

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

Heterostructures of TMDCs such as MoSe₂ and WSe₂ can host interlayer excitons, bound electron-hole pairs where charges are spatially separated in opposite layers. These quasi-particles have long lifetimes⁹ which can reach hundreds of nanoseconds in very high quality samples, revealing sizeable diffusion lengths.¹⁰ The spatial separation of charges gives rise to a permanent out-of-plane electrical dipole moment, which allows electrical control of exciton transport up to room temperature.⁷ Moreover, the valley-dependent optical selection rules in TMDCs permit to selectively create interlayer excitons with a certain valley-state⁶ which could be used to transport and store information. Further possibilities are enabled by the lattice mismatch and relative rotation between the two layers, leading to the formation of moiré patterns.¹¹ The resulting periodic potential and locally-changing optical selection rules^{12,13} allow to obtain highly versatile emitters with electrically tuneable energy, intensity and polarisation.⁸ However, with a corrugation reaching ~150 meV,^{12,13} the moiré potential can effectively trap interlayer excitons in its local minima,^{14–16} limiting their diffusion/drift. To address these issues, we introduce an atomically thin spacer between the constituent monolayers of our heterostructure to further separate the electron- and hole-hosting layers. This tuning of interlayer interaction alters the long-range moiré pattern, while preserving the coupling necessary for hosting bright interlayer excitons.¹⁷ With this method, we realize a valley-polarized excitonic transistor, in which we can electrically control the transport of excitons carrying a certain valley state. On the other hand, using a confining electrostatic potential we can increase their concentration, a step towards the creation of a valley-polarized exciton superfluid via Bose-Einstein condensation.^{18,19}

We have fabricated artificial heterostructures based on MoSe₂ and WSe₂ monolayers, with and without an atomically-thin hexagonal boron nitride (h-BN) separator. In Fig. 1a, we show a schematic depiction of a tri-layer stack (device A), fully encapsulated by h-BN flakes which serve as a flat and clean dielectric environment between the heterostructure and the top- and bottom-gates (see Methods and Fig. S1). Multiple transparent electrodes allow us to apply laterally-changing vertical electrical fields while performing optical measurements. Fig. 1b shows an optical micrograph of device A. To directly highlight the effect of the interposed layer, we also characterize a different heterostructure without the h-BN spacer (denoted as device B, Fig. S2) fabricated using the exact same process and similarly aligned to nearly-zero stacking angle, as confirmed by second harmonic generation measurements (Fig. S1e).

We first acquire polarization-resolved micro-photoluminescence (μPL) spectra by exciting the samples with a 647 nm-laser at 4 K (see Methods). Upon photon absorption, the type-II band alignment²⁰ of MoSe₂ and WSe₂ leads to fast charge separation of photo-generated carriers²¹, followed by the formation of interlayer excitons (IXs) from electrons in MoSe₂ and holes in WSe₂. For device A we observe the appearance of a single low-energy interlayer transition at 1.39 eV which preserves the circular polarization of incoming light (Fig. 1c, left panel). This is in sharp contrast to bilayer samples without h-BN spacer like device B, where we observe an interlayer doublet, characteristic of aligned heterobilayers,^{8,13,22} with opposite helicities for the two peaks (Fig. 1c, right panel). For device A, the polarization of the emitted light ρ (measure of valley-state conservation^{5,23}) has comparable magnitude to device B, decaying with increasing temperature and being tuneable by gate voltage (Fig. S3-4). Furthermore, we can detect non-zero polarization at temperatures as

high as 150 K, while the interlayer exciton emission can be observed up to room temperature, making these structures promising for applications at elevated temperatures. Similarly to previous reports¹⁷, the energies of interlayer transitions are comparable in both cases, a result of the interplay of inequivalent binding energies and different dielectric environments. The absence of the second interlayer transition is consistent with the picture of a reduced moiré interaction due to the presence of monolayer hBN between the two active layers.

Since the interlayer exciton has a built-in out-of-plane dipole moment $\mathbf{p}$, an external electrical field $\mathbf{E}$ perpendicular to the structure shifts its energy by $\mathcal{E} = -\mathbf{p} \cdot \mathbf{E}$. We extract this Stark shift from μ PL spectra taken as a function of the applied electric field (Fig. 1d) for both devices. Here, the slope of the energy shift is proportional to the size of the IX dipole $d = \Delta \mathcal{E} / eE_z$, where e is the elementary charge. We obtain $d \approx 0.9$ nm for device A, which is considerably larger than the previously reported value for bilayer structures,¹⁰ also observed in device B ($d_B \approx 0.6$ nm). The difference is similar to the thickness of a monolayer h-BN (~ 0.3 nm). We now study the diffusion of excitons as a function of incident power. For this, we excite the corner of device A with a diffraction-limited focused laser beam (Fig. 2a, first panel) while acquiring μ PL spectra as well as spatial images of the exciton photoluminescence (see Methods). As shown in Fig. 2a, when increasing the laser power P_{in} , the size of the exciton cloud grows significantly. Simultaneously, the PL emission moves to higher energy and broadens, while the intensity grows linearly (Fig. S5). We interpret the lack of saturation as a signature of reduced exciton-exciton annihilation effects due to the h-BN separator.^{24,25} By monitoring the blue-shift of the emitted light $\mathcal{E}_{\text{BS}}$, we can estimate a lower bound for the exciton density n_X , following a simple parallel plate capacitance model²⁶:

$$\Delta \mathcal{E}_{\text{BS}} = n_X \frac{de^2}{\epsilon_{\text{HS}} \epsilon_0} \quad (1)$$

where the dipole size d was determined from the Stark shift, ϵ_0 is the vacuum permittivity and $\epsilon_{\text{HS}} = 6.26$ is the effective relative permittivity of the WSe₂/h-BN/MoSe₂ heterotrilaier (see Methods). As shown in Fig. 2b, the energy shift grows sub-linearly, but does not saturate over the explored range of powers. We extract a maximum carrier density $n_X \sim 3 \cdot 10^{11} \text{ cm}^{-2}$ that is limited by the excitation power used. For comparison, the maximum density we can achieve for the device B is considerably smaller, below 10^{10} cm^{-2} . This is most likely due to Auger recombination at high pumping power²⁷.

After characterizing the exciton density, we turn our attention to exciton diffusion. From CCD images we obtain profiles of emission intensity as a function of the distance r from the excitation spot (normalized by their intensity at $r = 0$), as illustrated in Fig. 2c. Detailed analysis (Fig. S6) reveals two distinct diffusion regimes. Closer to the excitation spot, where exciton repulsion is dominant, we observe a very slow decay and large diffusion length ($l_D > 20 \mu\text{m}$), while further away the signal falls off faster, with a universal slope similar to the one seen at low power ($l_D \sim 0.9 \mu\text{m}$). In Fig. 2d we plot the effective exciton diffusion length l_D^{eff} , defined as the distance where the emission intensity drops to $1/e$ of its initial value. The

extracted value grows with the excitation power P_{in} , reaching $l_{\text{D}}^{\text{eff}} = 2.6 \mu\text{m}$ at $P_{\text{in}} = 740 \mu\text{W}$. This trend can be explained by the density-dependent exciton-exciton repulsion (see SI 6 for more details). For comparison, we draw (grey line in Fig. 2c) the profile from device B, where we observe a much weaker diffusion for a similar laser intensity. We attribute this to the effect of the moiré pattern and stronger Auger recombination, both of which are expected to be suppressed by the h-BN separator. Therefore, in the following we will focus on the tri-layer device (A).

The long diffusion length at high incident power allows us to realize an electrically operated excitonic switch device. Using multiple back gates, we create a laterally-modulated electric field along the x direction, which produces a spatial variation of the exciton energy profile $\mathcal{E}(x)$. Here we excite IXs by parking the laser spot ($P_{\text{in}} = 500 \mu\text{W}$) on the left side of a narrow back-gate, as shown in Fig. 3. By making the gate area higher or lower in energy with respect to its surroundings, we can allow or block exciton diffusion. We show in Fig. 3a-b the calculated interlayer exciton energy modulation $\mathcal{E}(x) = -\mathbf{p} \cdot \mathbf{E}(x)$ along the lateral position x for both configurations, together with a schematic of the expected exciton motion. Fig. 3c-d illustrate the spatial extent of the PL emission, i.e. the shape of the exciton cloud, for the two cases. For $V_{\text{BG}} = -7 \text{ V}$ the gated area acts as an energy barrier, effectively blocking the excitons at its edge (OFF-state), as shown in Fig. 3c. For $V_{\text{BG}} = 0 \text{ V}$ excitons are instead free to diffuse in a flat potential and move along the “channel” (ON-state), while their emission intensity decays over distance, as in Fig. 3d. We observe a $\sim 1.4 \mu\text{m}$ difference in exciton diffusion when comparing ON- and OFF-states (Fig. 3g). To gain further insight into the drift/diffusion process, we also probe the exciton energy as a function of the spatial coordinate (Figs. S7-8) while operating the excitonic transistor device where we can clearly see the diffusion of excitons into the lower-energy region.

Combining the excitonic device operation with valley conservation, we can realize a switch that effectively controls the flow of valley-polarized excitons. For this, we optically initialize the exciton valley-state by exciting the device with σ^+ circularly-polarized light. The result is displayed in Fig. 3e-f, where spatial images of the emitted polarization $I = I_{\sigma^+} - I_{\sigma^-}$ are shown for the OFF- and ON-state. By analysing the decay of I with distance in Fig. 3h, we see that valley-polarized excitons can either be stopped before the control gate, or travel over an additional $\sim 1.3 \mu\text{m}$ -distance when in the ON-state. While here we are interested in a proof of concept, the initial degree of polarization (here $\sim 15\%$) could be further improved by resonant excitation.²² We notice that the measured polarization is slightly higher in the ON-state. Its origin could be an additional repulsion of majority excitons due to exchange Coulomb interaction.^{6,28} Also, the valley polarization seems to decay slightly faster than the PL intensity over the channel length. This is because the emission further away from the excitation spot comes from longer-lived excitons, which have higher probability to undergo intervalley scattering. As mentioned earlier, the large binding energy allows us to observe IXs at high temperatures. Indeed, we can operate the valley-polarized switch up to a temperature of 100 K and simple excitonic switch at temperatures as high as 150 K (Fig. S9), comparing favourably to 3D semiconductors.²⁹

We can use the same principle not only to control fluxes of valley-polarized excitons, but also to confine them at higher densities. Indeed, while the emission intensity rises linearly with the pumping power, the blueshift increases sub-linearly (Fig. 2b) due to exciton-exciton repulsion lowering the density. To counteract this, we generate an electrostatically-defined potential well to constrain the valley-polarized excitons and concentrate them further. Now we shine a circularly-polarized laser (720 nm) directly on the area where we apply the electric field. As shown in Fig. 4a, anti-confining splits the valley-polarized exciton cloud in two lobes, pushing excitons away from the generation point. On the contrary, when we create a potential well in the lateral direction (Fig. 4c), excitons are squeezed to a narrower area compared to their natural diffusion (Fig. 4b). We gain further information from the dependence of exciton energy on the position. In the barrier case (Fig. 4d) excitons generated in the gate area have higher energy, hence they diffuse to the sides, where they emit light at the same energy of the zero-field case (Fig. 4e). This is consistent with the strongest PL emission being localized on the two sides of the barrier, and not at the laser spot. On the other hand, when we create a well, the exciton energy is lowered, producing spatial confinement (Fig. 4f). Interestingly, the energy shift of excitons is not symmetric with respect to the applied field (as expected from the pure Stark-effect). In Fig. 4g we plot the energy of excitons in the region inside (solid) and outside (dotted) of the gate area as a function of electric field for two different excitation intensities. At zero field, increasing the incident power generates a relative blueshift of ~12 meV, in agreement with Fig. 2b. However, when we disperse excitons (negative field), this blueshift is cancelled. Even more strikingly, when we start to confine the excitons, two phenomena appear: first, the magnitude of the blueshift between low- and high-power increases; and second, the exciton energy deviates drastically from a linear behaviour, especially in the high-power case. We attribute this non-linearity to the changing density inside the trap: since excitons are confined, their average energy is not only altered by Stark-shift, but also has a strong contribution from exciton-exciton interaction depending on local density: $\Delta \mathcal{E} = -\mathbf{p} \cdot \mathbf{E} + n_{IX} d e^2 / \epsilon_{HS} \epsilon_0$.

Conversely, when we separate them, even at higher power the density is low enough to make interactions negligible. We quantify the density modulation by two methods. First, we look at how the blueshift $\mathcal{E}(\mathbf{E}) = \mathcal{E}_{500 \mu W}(\mathbf{E}) - \mathcal{E}_{66 \mu W}(\mathbf{E})$ is enhanced by the applied field $\mathbf{E}$: $K(\mathbf{E}) = \frac{\Delta \mathcal{E}(\mathbf{E})}{\Delta \mathcal{E}(0)}$. This quantifies the increase in exciton density n_{IX} induced by higher power (Fig. 4h) as a function of $\mathbf{E}$, indicating that electrostatic confinement can modulate the exciton density. However, we are mostly interested in estimating the actual exciton density in the trap. For this, we isolate the non-linear contribution to $\mathcal{E}(\mathbf{E})$, proportional to the exciton density, by removing the Stark effect (grey dashed line in Fig 4g). We show the result in Fig. 4i, which allows us to put a lower bound on the concentration of polarized excitons at $n_{IX} \sim 1.8 \times 10^{12} \text{ cm}^{-2}$, promising for the production of a degenerate Bose gas. The maximum estimated exciton density obtained at 500 μW is only marginally higher than the one obtained at 66 μW . We attribute this to the fact that for the high density of excitons in the well, the energy rise due to interaction is comparable to the depth of the confining potential, thus making the trapping less efficient. The control over the concentration of polarized excitons represents a significant step towards the realization of high-temperature Bose-Einstein condensates of valley-excitons in these systems. Further experimental work to achieve high exciton concentrations in thermal equilibrium could include engineered

potential profiles or optimized traps, enabling the collection of thermalized excitons produced by pulsed excitation at even higher densities.

Methods

Heterostructure fabrication

Thin Cr/Pt (2/3 nm) bottom gates were realized by e-beam lithography and metal evaporation on silicon substrates covered by 270 nm of SiO₂. The heterostructure was then fabricated using polymer-assisted transfer of mono- and few-layer flakes of h-BN, WSe₂ and MoSe₂ (HQ Graphene). Flakes were first exfoliated on a polymer double layer. Once monolayers were optically identified and confirmed by photoluminescence, the bottom layer was dissolved with a solvent and free-floating films with flakes were obtained. These were transferred using a home-built setup with micromanipulators to carefully align flakes on top of each other. Polymer residue was removed with a hot acetone bath. Once completed, the stack was thermally annealed under high vacuum conditions (10⁻⁶ mbar) for 6 h at 340 °C. Finally, electrical contacts were fabricated using e-beam lithography and metallization (80 nm Pd for contacts, 8 nm Pt for the top-gate).

Optical measurements

All optical measurements were performed in vacuum at 4 K, unless stated otherwise (up to 300 K for temperature dependent measurements), in a He-flow cryostat with optical access. Interlayer excitons were optically pumped with a continuous wave 647-nm diode laser focused to the diffraction limit (spot width of 0.6 μm). For resonant excitation we employed a supercontinuum laser (Fianium) at 720 nm. In order to access a specific valley, a polarizer and a quarter wave ($\lambda/4$) plate were used for generating right/left circularly- or linearly-polarized light. For μ PL measurements, the emitted light was filtered by a 650-nm long-pass edge filter and then acquired using a spectrometer (Andor Shamrock with a charge-coupled device (CCD)). Polarization-resolved μ PL measurements were performed by employing another $\lambda/4$ plate and a birefringent yttrium orthovanadate beam displacer, so that σ^+ and σ^- signals could be acquired on the spectrometer simultaneously.

Spatial imaging of the interlayer exciton emission was captured by a CCD camera (Andor Ixon) with an 850-nm long-pass edge filter that removes both the laser line and the intralayer emission from MoSe₂ and WSe₂. A similar setup with a $\lambda/4$ plate on a rotator and a fixed linear polarizer was exploited for polarization-resolved PL imaging. Finally, the spectrally-resolved PL images were acquired by the following scheme: the light from the heterostructure was transmitted through a Dove prism, an 800-nm long-pass edge filter and a slit, and then was projected on the diffraction grating of the spectrometer. The Dove prism was positioned in such a way that the longitudinal axis of the gate (y -axis) was perpendicular to both the spectrometer slit and the lines of the diffraction grating. This way, spectral cut-lines along x -axis of the device were projected on the CCD camera of the spectrometer.

The heterostructure has a global transparent Pt top gate, and several split bottom gates of different size. This allows us to establish a uniform electric field between top gate and wide bottom gates, or only a narrow region with electric field if using one of the narrow gates.

For Stark shift measurements (Fig. 1d), the top gate and a wide bottom gate were used ($V_{BG} = -2.5 V_{TG}$) to create a uniform vertical electric field. Similar configuration was used to tune the carrier concentration at zero displacement field (in this case $V_{BG} = +2.5 V_{TG}$), connecting the heterostructure to ground (Fig. 4c). In both cases, the top gate and bottom gate voltages V_{TG} and V_{BG} were applied in a ratio 1:2.5 (corresponding to the top and bottom dielectric thickness ratio) to keep either the doping level or the displacement field constant, according to the case.

To realize a laterally-modulated electric field, as in Fig.3 and 4, we employed a voltage difference between the top gate (grounded) and a narrow bottom gate (V_{BG}) (as in Fig. 1a) with the heterostructure floating in between. This way, we can create a strong electric field in the heterostructure area between the top gate and the narrow split gate, while in the adjacent areas (without bottom gate) there is no electric field (as confirmed by electrostatic simulations, see Fig. S10). This creates a lateral modulation of the interlayer exciton energy. We note that, within the voltage range employed, no significant leakage current I_G was observed ($I_G < 1$ nA).

Second harmonic generation measurements were performed at room temperature in vacuum. A femtosecond Ti:Sapphire laser (Coherent Chameleon Ultra-I) was employed, with an 800-nm excitation wavelength. The incident angle of the linear polarization was tuned by rotating a Fresnel rhomb retarder, which acts as a half-wave plate. Collected light was then transmitted through the same retarder and a linear polarizer. In order to separate the excitation beam and the SHG signal, a 650-nm long-pass dichroic beam splitter was used. The signal was then filtered by a short pass filter installed in front of the spectrometer (Princeton Instruments SpectraPro HRS-300). The SHG signal was finally acquired on a Peltier-cooled CCD camera (Princeton Instruments Blaze) mounted on the spectrometer.

Image processing

Images acquired from the CCD camera and the spectrometer were analysed and processed using the software ImageJ³⁰. The images were rotated and cropped to be all at the same scale, and the contrast was adjusted to cover the range of values in the image. In some cases, a background obtained without laser illumination was subtracted from the images, to account for ambient light noise.

Numerical simulations

The electric field plotted in Fig. 1d was calculated as follows from the voltages applied to the top V_{TG} and bottom V_{BG} gates:

$$E_z = \frac{V_{BG} - V_{TG}}{d \sum} \cdot \frac{\epsilon_{hBN}}{\epsilon_{HS}}$$

where $\epsilon_{\text{hBN}} = 4$ is the relative permittivity of h-BN crystals in the out-of-plane direction. The total thickness d was calculated as $d_{\Sigma} = d_{\text{hBN}}^{\text{top}} + d_{\text{hBN}}^{\text{top}} + d_{\text{hBN}}^{\text{spacer}} + 2d_{\text{TMDC}} = 67.6$ nm, taking into account the thickness of the top h-BN $d_{\text{hBN}}^{\text{top}} = 23$ nm, bottom h-BN $d_{\text{hBN}}^{\text{bot}} = 43$ nm, spacer h-BN $d_{\text{hBN}}^{\text{spacer}} = 0.3$ nm, and thickness of TMDC monolayers $d_{\text{TMDC}} = 0.65$ nm. In the calculations we used an effective value for the permittivity of the heterostructure ϵ_{HS} , which was calculated by considering the heterotrilaier as a series of three capacitors:

$$\epsilon_{\text{HS}} = \frac{(2d_{\text{TMDC}} + d_{\text{hBN}}^{\text{spacer}}) \cdot \epsilon_{\text{hBN}} \cdot \epsilon_{\text{TMDC}}}{2d_{\text{TMDC}} \cdot \epsilon_{\text{hBN}} + d_{\text{hBN}}^{\text{spacer}} \cdot \epsilon_{\text{TMDC}}} = 6.26$$

where $\epsilon_{\text{TMDC}} = 7.2$ is the relative permittivity of the TMDC monolayers.

For the spatial distribution of the electric field inside the heterostructure and corresponding exciton energy profile, we performed numerical simulations using COMSOL Multiphysics (see Fig. S10). The model used the exact same physical parameters as described above.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1. Device characterization.

(a) Schematic of the device structure illustrating the top and bottom split gates, as well as electrical connections. (b) False coloured optical image of the device, highlighting the different materials. Scale bar is 10 μm . (c). Polarization-resolved micro-photoluminescence spectrum from the $\text{WSe}_2/\text{h-BN}/\text{MoSe}_2$ heterotrilayer (left) and the $\text{WSe}_2/\text{MoSe}_2$ heterobilayer (right) excited with right circularly polarized light. (d) Energy of interlayer exciton emission as a function of applied vertical electric field (E_z) when sweeping at constant doping Device A (blue) or Device B (grey). Solid lines correspond to the linear Stark shift of the dipole with size $d = 0.9$ nm (0.6 nm) extracted for the heterotrilayer (heterobilayer).

Figure 2. Exciton diffusion.

(a) Leftmost figure: CCD image of the focused laser spot in the corner of the heterostructure, as indicated by the dashed line. Other figures: CCD images of the IX PL normalized emission intensity, acquired for different incident powers. Scale bar is 4 μm . (b) In blue: extracted blueshift dependence on the incident power. The solid curve is a power-law fit. In grey: full width at half maximum (FWHM) of the interlayer emission spectra at different incident powers resembles the power dependency of the blueshift. (c) Normalized PL intensity versus distance from the excitation point extracted from a. The laser profile is shown by the red area. Grey area shows diffusion of excitons in device B at 200 μW incident power. Dashed line represents the maximal emission intensity. (d) Extracted effective diffusion length (a distance from excitation spot where the emission intensity drops to $1/e$ of its initial value) as a function of incident power. The dashed line shows the diffusion length extracted from the tails in c, fitted by the convolution of the Gaussian-like laser profile with modified Bessel function of the second kind. Inset schematics demonstrate density-dependent diffusion driven by exciton-exciton repulsion.

Figure 3. Valley-polarized excitonic switch.

(a-b) Numerically simulated exciton energy profile (red line) in the OFF ($V_{TG} = 0 \text{ V}$, $V_{BG} = -7 \text{ V}$) and ON ($V_{TG} = 0 \text{ V}$, $V_{BG} = 0 \text{ V}$) states of the excitonic transistor. (c-d) Real-space CCD images of the emitted PL intensity for the OFF and ON states. (e-f) Real-space CCD images of the exciton cloud polarization measured simultaneously with c-d. (g-h) Intensity profiles of emitted intensity and polarization along a cutline in the middle of figures c-d and e-f, highlighting the operation of the valley-polarized excitonic transistor. The blue dashed rectangle in all images corresponds to the back-gate area where the vertical electric field is

modulated, as depicted in **a-b**. The white dashed area is a guide for the eye of the approximate shape of the heterostructure. The black dashed circle indicates the laser spot. Scale bar is 2 μm .

Figure 4. Electrostatic control of exciton concentration.

(a-c) Real-space CCD images of the exciton cloud polarization corresponding to the configurations of anti-confinement, free diffusion and confinement, observed at 720 nm resonant excitation with incident power of $P_{in} = 220$ W ($V_{TG} = 0$ V, $V_{BG} = -7, 0, +7$ V). The simulated energy shift for the interlayer excitons in the various cases is drawn as a yellow overlay. The red overlay shows the intensity profile along the lateral direction in the middle of the image. As a reference for the eye, the profile in the $V_{BG} = -7$ V state is replicated as a dashed line in the last two panels. PL intensity images are shown as insets. (d-f) "Energy vs x " diagram of the emission energy as a function of the lateral coordinate x in the same configuration as in a-c. The yellow overlay shows the spectra from the central region. (g) Peak emission energy in the gate area (solid line) and outside of it (dashed line) as a function of the applied electric field for 500 μ W (red) and 66 μ W (blue) incident power. In grey, the

linear stark effect extracted from Figure 1. **(h)** Ratio between the blueshift and the blueshift at zero electric field. **(i)** interlayer exciton density as a function of the applied electric field extracted from the non-linear behaviour of the energy shift for 500 μW (red) and 66 μW (blue) incident powers. The dashed rectangle in all images corresponds to the gate area, where the vertical electric field is modulated. Error bars represent the propagation of fitting uncertainty on the density. Scale bar is 2 μm .