Supplementary Information

for

Valley-polarized exciton currents in a van der Waals heterostructure

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1. **Device A - fabrication and characterization**

Fig. S1a shows optical images acquired at different steps of device fabrication. We start by realizing bottom gates by e-beam lithography and evaporation of thin Cr/Pt (2/3 nm) 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, as described in the method section. After removing polymer residue with hot acetone bath and vacuum annealing, we deposit electrical contacts using e-beam lithography and metal evaporation (80 nm Pd for contacts, 8 nm Pt for the transparent top-gate).

To probe the homogeneity of the interlayer coupling strength, we perform photoluminescence spatial mapping of the completed heterostructure. For this, we track the emission intensity at the wavelength of MoSe₂ and WSe₂ intralayer excitons, as well as the lower-energy interlayer exciton. Corresponding results are presented in Fig. S1b-d. We clearly see that intensity distribution of intralayer exciton emission coincides with exposed monolayers and is strongly quenched in the heterobilayer region where the interlayer coupling is the strongest.

In order to confirm that both the heterobilayer and heterotrilayer stacks have similar stacking angles, we perform second harmonic generation (SHG) measurements on both structures. Fig. S1e shows SHG signals acquired on exposed single layers of WSe₂ and MoSe₂ in A (top) and device B (bottom). The twist angle Δθ was calculated from the fitting of the experimental data for WSe₂ (red) and MoSe₂ (blue), revealing good alignment of the crystals.

**Supplementary Fig. S1. Fabrication details.** (a) fabrication steps of device A showing: local back-gate array evaporation; transfer of bottom h-BN and MoSe₂ on the gates; transfer of the h-BN spacer (inset: AFM image of the h-BN flake); transfer of WSe₂ and top h-BN flake; finished device with contacts and top-gate. Scale bar is 25 μm. µPL maps of device A: (b) centred around the emission energy of MoSe₂, (c) centred around the emission energy of WSe₂ and (d) around the emission energy of the interlayer exciton (IX). Scale bar is 10 μm. (e) Angle-dependent SHG measurements of the device A (top half) and device B (bottom half), together with the stacking angles calculated from the fitting of experimental data.
2. Heterobilayer reference device (Device B)

Similarly, we fabricate the device B from a well-aligned WSe$_2$/MoSe$_2$ heterostructure. Alignment of the stack is confirmed by SHG in Fig. S1e. We build the stack encapsulated in h-BN crystals on an array of local back-gates, which allow us to locally modify the electric field, generating regions with lower or higher energetic excitons. An optical image of the resulting structure is shown in Fig. S2a. First, we study free diffusion of excitons in this structure. For this, we focus the laser beam on the heterostructure above one of the split gates and record spatial images of PL emission, corresponding to the spatial distribution of the exciton cloud. The result is shown in Fig. S2b, top. We can see that the emission spot is slightly bigger compared to the excitation profile, shown in Fig. S2c (red and grey), which is a consequence of exciton diffusion.

We now apply positive voltage on the local gate and negative voltage on the silicon global gate, while keeping the top gate grounded, in order to generate a lower-energy region in close proximity to the excitation spot. As a result, excitons preferentially diffuse towards this area, as shown in Fig. S2b (bottom) and c (dashed grey). Even if we clearly see the effect of the drift transport, the initial diffusion length is rather short, and therefore the control over the excitonic transport is not efficient. For comparison, we demonstrate in Fig. S2c (solid blue curve) the free diffusion observed for device A in similar conditions. We attribute the weak diffusion of IXs in well-aligned heterobilayers to the trapping of excitons in the moiré periodic potential. This limited diffusion was consistently observed in two additional heterobilayers, all fabricated with the same procedure. We note that different diffusion profiles observed by other studies$^{1,2}$ could be related to different fabrication methods and to relative alignment between crystals, which is the main source of moiré potential.

**Supplementary Fig. S2. Details of Device B.** (a) False colour optical image of the device, highlighting the different materials and local bottom gates. Scale bar is 10 μm. (b) CCD images of the spatial distribution of the interlayer exciton emission in absence (top) and presence (bottom) of the applied electric field that promoted IX to diffuse on the left from the excitation spot upon application of $V_{BG}$ and $V_{Si}$ voltages on the local and global silicon back gates. Scale bar is 1 μm. (c) Normalized PL intensity as a function of the distance $r$ from the excitation point, extracted from b. The addition of a longitudinal electric field gradient clearly promotes the exciton diffusion (grey dashed vs grey solid curve). However, the effect is very small if compared to the diffusion observed in the Device A (blue curve), acquired in similar excitation conditions and in absence of electric field. Profile of the laser spot is corresponds to the area in red.
3. Temperature dependence of the IX emission and polarization

For device A, we notice that the polarization of the emitted light $\rho$ (which is a measure of valley-state conservation) has comparable magnitude to device B. Interestingly, temperature dependent studies presented in Fig. S3 show the decay of the polarization with increasing temperature, likely due to enhanced inter-valley scattering. Nevertheless, 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 quite promising for applications at elevated temperatures (see Supplementary Information section 9).

Supplementary Fig. S3. Temperature dependence. (a) $\mu$PL spectra of interlayer exciton measured at different temperatures ranging from 4 to 300 K. (b) Intensity of the IX emission versus temperature of the system. (c) Temperature dependence of the degree of the polarization integrated over the complete IX spectra. (d) Maps of $\mu$PL intensity (left) and polarization (right) as a function of temperature.
4. Gating dependence of the IX emission and polarization

We further study the dependence of interlayer excitons and their polarization degree on the doping level of the system in device A. Corresponding results are shown in Fig. S4.

We notice that similarly to the heterobilayer structure, excitons hosted by the heterotrilayer are tuneable by gate voltage in terms of emission intensity and degree of circular polarization (Fig. S4b). Nevertheless, we do not observe sudden polarization switching as has been reported for the WSe₂/MoSe₂ heterostructure without the h-BN spacer layer, indicating reduced moiré effects.

In Fig. S4c we perform gate dependent PL measurements in the dual-gate configuration in order to minimize the electric field. While we can observe a slight modulation of the peak intensity, we do not see the formation of any lower energy emission, which would suggest the formation of charged interlayer excitons. This is in agreement with theoretical predictions (see ref. 3), which suggest fast decrease in binding energy for charged-IX when increasing the interlayer separation distance. We can thus safely assume that charged interlayer excitons do not play a relevant role in our system.

Supplementary Fig. S4. Gating dependency. (a) Set of polarization-resolved µPL spectra acquired for different top-gate voltages ($V_{TG}$) when the heterotrilayer is excited with right-circularly polarized light. HS is kept grounded for these measurements. The backgate is not used in this set of measurements. (b) Degree of polarization versus applied gate voltage $V_{TG}$ extracted from the spectra in a. The heterostructure is grounded for this set of measurements. (c) Gate dependent measurements of IX photoluminescence. Measurements were performed in a dual gate configuration ($V_{BG} = 2.5 \ V_{TG}$, HS is grounded) in order to reduce effects from the displacement field and have pure electrostatic doping.
5. Power dependence of the IX emission

We now study how the interlayer excitons hosted in heterotrilayer (device A) change their behaviour for high incident laser powers. Fig. S5a shows the power dependency of the spectrum, from which we can extract the total PL intensity as a function of incident power, as in Fig. S5b. We interpret the lack of saturation as a signature of negligible exciton-exciton annihilation effects, as the h-BN interlayer spacer is expected to reduce density-related recombination. We also observe clear blue shift of IX energy at higher exciton density. We assign it to the exciton-exciton repulsion. We extract the magnitude of the blue shift in the main text, Fig. 2b.

Supplementary Fig. S5. Power dependency. (a) Map of photoluminescence emission as a function of incident laser power $P_n$ normalized by the acquisition time and incident power (counts/s/$\mu$W). (b) Extracted intensity of IX emission that demonstrates linear power dependency even at high excitation powers.
6. Analysing and fitting of the IX diffusion profiles

From the PL images in Fig.2a we obtain profiles of emission intensity as a function of the distance $r$ from the excitation spot (normalized by their intensity at $r = 0$), for the set of incident powers. At low excitation powers, the density profile of interlayer excitons can be fitted with the convolution of the laser Gaussian profile $e^{-r^2/w^2}$ and the modified Bessel function of the second kind $K_0$:  

$$n_{IX} = n_0 \int_{-\infty}^{+\infty} K_0 \left( \frac{r'}{l_D} \right) e^{-\frac{(r-r')^2}{w^2}} dr'$$  

(1)

where $l_D \sim 0.9 \, \mu m$ is the diffusion length of the interlayer excitons. However, the PL intensity profiles acquired for higher excitation power deviate from Eq. 1, with the appearance of two distinct regimes (SI Fig. S6): close to the excitation spot we observe a very slow decay corresponding to a diffusion length $l_D > 20 \, \mu m$, while further away the signal declines faster, with a universal slope very similar to the one seen at low power.

Near the excitation area the high density of excitons causes a strong repulsive interaction between them, proportional to the excitation power. This density-dependent repulsive force creates a drift-like motion of excitons away from the excitation spot, increasing their diffusion length at high incident powers. As the result, the extracted effective diffusion length (Fig. 2d) becomes dependent on the excitation power. Similar phenomena have indeed been observed for indirect excitons in GaAs CQWs.\textsuperscript{5}

On the other hand, at low incident powers or further away from the laser spot, as the density of these quasiparticles drops, pure exciton diffusion sets in and starts to dominate, yielding a constant far-away diffusion constant.
7. Valley-polarized excitonic transistor operation

In Fig. S7a we can see that by applying voltage to the gate electrode we can block or allow the diffusion of excitons. We notice that due to the reduced size of the gate, the maximum diffusion is obtained in the case of zero field ($V_{BG} = 0$ V). Taking the point where the intensity of the emitted light falls to $1/e$ of its maximum, we obtain a transport over $\sim 1.4$ µm in the ON state as compared to the OFF state. The profiles are taken by plotting the intensity along a horizontal cutline of the images of the PL spatial distribution. As shown in Fig. S7b, the transport works similarly for valley-polarized excitons.

Supplementary Fig. S7. Cut-lines detailing the operation of the valley-polarized excitonic transistor. (a) Intensity profile of the emitted PL for different voltage configurations from $V_{BG} = -7$ V to 0 V, $V_{TG} = 0$ V (b) Intensity profile of the polarization $\Delta I = I_{\alpha+} - I_{\alpha-}$ for the same voltage configuration.
8. Intermediate states of the excitonic transistor

In Fig. S8a the CCD images of the PL emission are shown for different gate voltages from \( V_{BG} = -7 \) V to \( V_{BG} = +7 \) V. Fig. S8b shows the exciton energy profile along the lateral coordinate (x) for the same voltage configuration.

As we can see, excitons are confined in space to the region to the left of the gate area for \( V_{BG} = -7 \) V. Energetically, they are localized around the usual value of 1.39 eV. When lowering the energy barrier (moving \( V_{BG} \) towards 0 V) the exciton cloud starts to diffuse also in the gate area, as can be seen from the panels of Fig. S8a-b. Finally, for positive gate voltage the gate area becomes energetically favourable, and thus excitons start to flow inside. This can be observed clearly in the last four panels of Fig. S8b, where the exciton cloud starts to split in energy: the excitons outside the gate stay at the usual energy, while the ones in the well are progressively lowered in energy.

Supplementary Fig. S8. Intermediate states of the excitonic switch. (a) Real-space CCD images of the emitted PL intensity corresponding to \( V_{BG} \) values from −7 V to +7 V. (b) "Energy vs x" diagrams of the emission energy as a function of the lateral coordinate x, for the same voltage configurations as in a. Top gate is grounded during these measurements. Scale bars are 2 \( \mu \)m.
9. Excitonic transistor operation at elevated temperatures

As mentioned in the main text, the relatively large binding energy of interlayer excitons allows us to observe IXs at high temperatures. Indeed, we can operate the valley-switch up to a temperature of 100 K, as shown in Fig. S9a. However, polarization conservation becomes negligible above this temperature, as we demonstrated in Fig. S9b.

Additionally, in Fig. S9c and d we demonstrate that the simple excitonic switch works at temperatures as high as 150 K. These results show the potential of 2D materials for excitonic device applications.

Supplementary Fig. S9. Exciton switch operation at elevated temperature. (a) Images of the spatial distribution of PL for $V_{BG} = -7$ V (OFF), 0 V (ON) and +7 V (collection) at 100 K. Insets show spatial distribution of $\Delta I = I_{ON} - I_{OFF}$ for each configuration, whereas the black overlay is the intensity of $\Delta I$ along the horizontal direction. (b) Plot of the intensity along a horizontal cutline in the middle of the images. Inset: intensity of $\Delta I$ along the same cutline. (c) Images of the spatial distribution of PL for $V_{BG} = -7$ V (OFF), 0 V (ON) and +7 V (collection) at 150 K. (d) Plot of the intensity along a horizontal cutline in the middle of the images. The dashed rectangle in all images corresponds to the gate area, where the vertical electric field is modulated. The top gate is grounded during these measurements. Scale bars are 2 µm.
10. Numerical simulations

To calculate the vertical electric fields in our structure in different regimes, we perform numerical simulations using COMSOL Multiphysics (see main text, Methods section). An example of the simulated electric field inside the heterostructure in the case of a confining potential \( V_{BG} = 7 \text{ V} \) is shown in Fig. S10a. We also calculate the corresponding energy of interlayer excitons, as shown in Fig. S10b. We use these calculations for the interpretation of our experimental data.

**Supplementary Fig. S10.** COMSOL simulations. (a) Spatial distribution of the electrostatic potential in the simulated structure for the case \( V_{BG} = 7 \text{ V} \). (b) Calculated energy profiles for the interlayer exciton energy for \( V_{BG} = -7 \text{ V} \), 0 V and 7 V and \( V_{RG} = 0 \text{ V} \).
11. Thickness values used in the calculations

The thickness values used in the calculations and simulations are justified by AFM measurements. In Fig. S11 we present AFM micrographs of exfoliated monolayer h-BN, h-BN transferred on MoSe₂, and a WSe₂/MoSe₂ heterobilayer structure revealing the ~0.7 nm experimental interlayer distance. Regarding this, we note here that only a clean transfer process and high-temperature annealing yield high quality vdW interfaces with interlayer separation comparable to the expected one.

Supplementary Fig. S11. AFM images of a representative WSe₂/1L-hBN/MoSe₂/hBN heterostructure prior to encapsulation and top gate deposition. (a) AFM image of isolated hBN flake. Overlay shows the height profile along the white dashed line. (b) AFM image of an area with 1L h-BN on top of MoSe₂ on top of thick h-BN. (c) AFM image of an area with WSe₂ directly on top of MoSe₂. Overlay shows the height profile along the white dashed line. After annealing in high vacuum at high temperature, the interfacial residue merges in few isolated bubbles, while the rest of the heterostructure is clean and homogeneous.

SUPPLEMENTARY INFORMATION REFERENCES

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