Polarization switching and electrical control of interlayer excitons in two-dimensional van der Waals heterostructures

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The long-lived interlayer excitons in van der Waals heterostructures based on transition-metal dichalcogenides, together with unique spin-valley physics, make them promising for next-generation photonic and valleytronic devices. Although the emission characteristics of interlayer excitons have been studied, efficient manipulation of their valley states, a necessary requirement for information encoding, is still lacking. Here, we demonstrate comprehensive electrical control of interlayer excitons in a MoSe2/WSe2 heterostructure. Encapsulation of our well-aligned stack with hexagonal boron nitride (h-BN) allows us to resolve two separate narrow interlayer transitions with opposite helicities under circularly polarized excitation, either preserving or reversing the polarization of incoming light. By electrically controlling their relative intensities, we realize a polarization switch with tunable emission intensity and wavelength. Finally, we observe large g-factors of these two transitions on application of an external magnetic field. These results are interpreted within the picture of moiré-induced brightening of forbidden optical transitions. The ability to control the polarization of interlayer excitons is a step towards the manipulation of the valley degree of freedom in realistic device applications.

Electronic devices rely on the manipulation of the charge degree of freedom to store and process information. To overcome their fundamental limitations, different degrees of freedom could be harnessed in new device concepts. Using the electron spin, for example, has been considered as an attractive alternative to charge-based devices1. Another possibility is to manipulate the valley degree of freedom2,3, associated with the minima (maxima) of the conduction (valence) band that a carrier is occupying. In this regard, two-dimensional semiconductors such as transition-metal dichalcogenides4–8 are particularly attractive for valley manipulation, because their band structure has inequivalent K and K’ valleys, which produce a binary pseudospin system9–11. Neutral and charged excitons in these heterostructures are of high interest, because they can be electrically manipulated12 and are endowed with a valley degree of freedom by their constituent electrons and holes, which can be optically addressed with circularly polarized light13–15. Previous reports have revealed that interlayer excitons16 are particularly suitable for this purpose, showing emission of polarized light with the same or the opposite handedness as used for excitation17,18. To fully exploit this property, it is nevertheless essential to find a way to manipulate the polarization (that is, valley) state of the excitons. Indeed, pioneering work by Rivera et al.19 has established that the degree of preserved circular polarization can be reduced using a gate voltage.

For full control of such manipulation, different strategies could be employed, including the creation of a moiré potential. Two-dimensional van der Waals heterostructures possess unique characteristics that arise from the moiré patterns that form when different crystals with lattice mismatch are brought into contact. This opens up a new direction for research because the moiré can dramatically change the material properties, as experimentally demonstrated for graphene on h-BN20–22. This approach has been also recently considered for exciton–moiré interactions. In particular, the prediction of a moiré-potential-induced brightening of forbidden optical transitions22,23 and its electrostatic tunability could enable full switching of the helicity state (that is, realize a logic NOT gate), crucial for logic operations.

In this Article, we demonstrate a polarization switch based on interlayer excitons in a well aligned van der Waals heterostructure, whose operation is enabled by the peculiar physics of this system. This device allows us to manipulate the helicity of light, as well as its wavelength and intensity, by the application of electric and magnetic fields.

Results

Our device consists of a contacted MoSe2/WSe2 heterobilayer encapsulated in h-BN, with a graphene bottom gate and a top transparent Pt gate (Fig. 1a). The stack is realized using the dry-transfer technique24 on a doped silicon substrate covered with 270 nm of SiO2, and the crystals are aligned to minimize the stacking angle (δθ ≤ 1˚; see Methods), thus creating a long-period moiré superlattice. This device architecture allows us to perform optical measurements on the sample while applying different voltages through the top and bottom gates as well as the global Si backgate and gives us the ability to control the doping level and the transverse electric field independently. Figure 1b shows the optical microscopy image of one of our completed heterostructure devices (see Methods for fabrication details). We note that all the measurements reported here were acquired at 4.2 K on the same device, unless stated otherwise.

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device and demonstrate intensity and energy manipulation, which
the polarization of the two transitions, we first characterize our
Electrical tuning of interlayer excitons.
Before manipulating
selection rules dictated by local atomic registry.
full-width half-maxima (FWHM) between 7 meV and 15 meV
peaks from individual monolayers (‘1L’ in insets to Fig. 1d), with
around 5 meV and an energy separation of about 25 meV between
We clearly resolve two distinct emission peaks (Fig. 1e), with FWHM
Fig. 2) around 1.39 eV owing to interlayer exciton (IX) formation.
the appearance of low-energy emission (Fig. 1d and Supplementary
(Supplementary Fig. 1). In the heterobilayer region, we observe an

h-BN-encapsulation of the heterostructure28 with a type-II band alignment 29 (Fig. 1c). Since
electrons and holes are confined to separate layers, interlayer excitons have a defined dipole moment \( \mathbf{p} \) perpendicular to the heterostructure plane. This allows us to tune their energy linearly with an
external electric field \( \mathbf{E} \) along the dipole axis: \( \Delta U \approx -\mathbf{p} \cdot \mathbf{E} \). To this end, we apply a vertical field at constant carrier concentration. As
shown in Fig. 2a, a modulation of the IX emission maximum of \( \Delta U \approx 138 \text{ meV} \) is obtained, from \( \approx 1.330 \) to \( \approx 1.468 \text{ eV} \). A linear fit of the energy shift yields a tuning rate of about 500 meV nm V\(^{-1}\), from which we obtain a qualitative estimation of the dipole size \( d \approx \Delta U/qE \approx 0.5 \text{ nm} \) (where \( q \) is the elementary charge), compatible with the expected interlayer spacing30. Although this semiclassical dipole picture is oversimplified, it captures the main effects we observe. For positive electric fields, the decrease in energy of the dipole (charges are more separated) results in reduced recombination rate and a slightly larger Stark effect. In contrast, when the exciton energy is increased, and the overlap between the electron and the hole is larger, we observe brighter photoluminescence.

If we ground the heterobilayer while applying voltage to the top gate, we can achieve control over the relative intensities of the two peaks by changing the charge carrier concentration. In contrast with previous reports17, our dual-gated configuration allows us to independently control exciton energy or relative peak intensity, while keeping the other property fixed. This geometry also allows for precise control over the doping of individual layers within the heterobilayer
(Supplementary Sections 6 and 7). We show the results of this electrostatic doping in Fig. 2b. For negative values of \( V_{\text{G}} \), the intensity of the IX\(_2\) peak is first reduced, then suppressed around \(-4\) V. At the same time, IX\(_1\) becomes broader and starts to dominate the spectrum. In contrast, at high positive voltages, we observe that IX\(_1\) becomes the dominant emission feature, while IX\(_2\) decreases in intensity and becomes quenched at higher electron density achieved by dual gating (see Supplementary Fig. 5). This closely resembles what one would expect from a two-level system, where with increased doping more electrons are driven into the upper level: here this comes from the filling of the lower spin-split conduction band and the population of the upper one. This interpretation is also supported by the observation of a faster increase in the intensity of IX\(_1\) with increasing laser power in the absence of electrostatic doping (see Supplementary Fig. 4 for details on power dependence). Further confirmation of this filling mechanism is the temperature dependence of the two transitions (Supplementary Fig. 5), with IX\(_2\) becoming stronger as the band is thermally populated.

**Polarization response.** Excitonic valleytronic devices should have an optical input and output, with the information encoded in the polarization of light. Consequently, selectively addressing the valley degree of freedom of excitons with polarized light is critically important. To this end, we characterize the polarization-resolved photoluminescence from our heterostructure. As expected, the emission intensity for positive (\( \sigma^+ \)) and negative (\( \sigma^- \)) helicity are the same in the case of linear excitation. The situation changes with circularly polarized excitation. We observe robust conservation of the incident polarization from monolayer WSe\(_2\), but not from MoSe\(_2\) (Supplementary Fig. 8). As shown earlier, our clean interfaces in h-BN-encapsulated heterostructures allow us to resolve the two different optical transitions, IX\(_1\) and IX\(_2\). Here, we observe that IX\(_1\) and IX\(_2\) have opposite behaviour under circularly polarized excitation, with polarization values up to 27% and \(-25\%\) respectively, in agreement with recent results\(^7\) (Supplementary Fig. 9). Such behaviour agrees with what is expected from a spin-conserving (spin-flipping) transition between the WSe\(_2\) valence-band maximum and the lower (upper) conduction-band minimum of MoSe\(_2\). As we discuss later in the text, in WSe\(_2\)/MoSe\(_2\), both of these transitions are allowed, with opposite polarizations and comparable intensities, for excitons localized in some energy minima of the moiré pattern.

![Fig. 2](image)

**Fig. 2** | **Electrical control of interlayer excitons.** a, Map of photoluminescence emission as a function of applied gate voltages \( V_{\text{G}} \) and \( V_{\text{BG}} \) when sweeping at constant doping. b, Map of photoluminescence emission as a function of gate voltage when electrostatically doping the device.

We now focus on the central point of our experiment, by combining the gate modulation of the two excitonic peaks with their unique polarization dependence. As we have seen, strong electron doping enhances IX\(_2\), while at small or negative gate voltages IX\(_1\) dominates. Thanks to the opposite polarization of the two peaks, this allows us to change the device operation between a polarization-inverting and polarization-preserving regime. We show the corresponding results in Fig. 3 (see Supplementary Fig. 10 for raw left and right polarization); both excitonic peaks are clearly visible in the upper (positive) half of the map, with opposite helicity. In the left panel of Fig. 3c, the spectra corresponding to \( V_{\text{G}} = 0\) V are presented. Owing to the higher intensity of the IX\(_1\) peak, the total polarization of the signal \( \Delta I_{\text{RL}} \) (net polarization \( \delta I_{\text{RL}} = I_+−I_- \) integrated over the spectrum) is positive (that is, of the same sign as the excitation). This is even more clearly visible in the right panel of Fig. 3c, where the spatial image of the exciton polarization acquired on the charge-coupled device (CCD) is shown. For strong electron doping the situation is reversed, as seen in Fig. 3b. In this configuration, IX\(_2\) emission is stronger, resulting in an overall negative value of \( \Delta I_{\text{RL}} \), and our device operates here as a polarization inverter. Even more interesting is the behaviour in the p-doped region (that is, for the application of negative gate voltage). As seen before, the higher-energy IX\(_1\) peak is suppressed at negative gate voltages, so one would expect the device to strongly preserve the helicity when electrostatically p-doped. On the contrary, IX\(_2\), polarization behaviour is now completely reversed, while IX\(_1\) shows a vanishing circular polarization (see lower half of Fig. 3a). This results from the alteration of the moiré potential induced by electrostatic doping, which shifts the exciton localization from one type of local minimum to another in the moiré pattern, with different local symmetry and thus different light coupling rules (see below). In Fig. 3d we show the spectra recorded for a strong hole-doped case, demonstrating that the polarization-inverting emission is indeed coming from the lower-energy IX\(_2\). Just as in the case of positive gate voltage, we obtain a globally negative polarization (right panel).

From our results, we can conclude that the contrasting reports in recent literature on the polarization dependence of photoluminescence in this system (observation of conservation\(^9\), reversal\(^13\) or both\(^7\) for the light helicity) could be due to different levels of natural doping occurring in the samples, over which previously there was no full control, as well as due to variable amounts of disorder. Indeed, using the same device structure, we could reliably reproduce
our observations in successive devices (Supplementary Fig. 11), obtaining qualitatively similar results.

To characterize the switching operation in more detail, we study the evolution of $\Delta I_E$ (polarization integrated over the spectrum) as a function of the applied gate voltage, as shown in Fig. 4. For $V_{\text{TG}}$ higher than 5 V, we have negative $\Delta I_E$ (‘inverter’ in Fig. 4), as a result of IX2 being the strongest transition (as in Figs. 2b and 3a). For gate values between 5 V and 0 V, IX dominates, giving positive $\Delta I_E$, that is, preserving the input polarization (‘transmitter’). It is interesting to note that within this region, application of gate voltage also allows us to modulate the amplitude of the effect, with a maximum around $V_{\text{TG}} = 1$ V. We then see a sharp transition between the two logic states happening around zero gate voltage, with a small required switching voltage around ±1 V. This threshold value depends on the gate capacitance, and could thus be considerably reduced by engineering thinner dielectric layers to obtain even higher efficiency. For $V_{\text{TG}}$ below 0 V, we have an inverting action, owing to the polarization reversal of IX1.

Zeeman splitting of the doublet. Since the valley pseudospin is associated with the magnetic moment, it is also possible to manipulate it with an external magnetic field$^{15}$. Indeed, to gain more insight into our double excitonic transition, the extraction of the effective $g$-factor could prove useful. Towards this, we performed polarization-resolved photoluminescence measurements in a magnetic field $B$, between −3 T and +3 T. The results are shown in Fig. 5. Similarly to the monolayer case$^{13}$, the $\sigma^-$ and $\sigma^+$ components of the photoluminescence peaks experience opposite energy shifts, with a splitting $\Delta E^{\sigma}$ proportional to $B$. Interestingly, the same polarization component for each peak undergoes energy shifts slightly different in amplitude (see Fig. 5). For a more quantitative analysis of the data, we extract the peak positions using a technique similar to that used in ref.$^{13}$. In Fig. 5b we show the calculated shift for IX1, IX2: $\Delta E^{\text{IX1}} = E^{\text{IX1}} - E^{\text{IX1}_0}$, $\Delta E^{\text{IX2}} = E^{\text{IX2}} - E^{\text{IX2}_0}$, together with their linear fits. Using the Zeeman energy shift $\Delta E = g_{\text{eff}} B$, where $g$ is the Bohr magneton and $B$ is the applied field, we calculate effective $g$-factors $g_{\text{IX1}} = -8.5 \pm 1.5$ and $g_{\text{IX2}} = 7.1 \pm 1.6$. Our result is considerably larger than previously reported effective $g$-factors in monolayer transition-metal dichalcogenides (normally around −4), in line with recent results on similar heterostructures$^{14}$.

**Fig. 3 | Electrical control of polarization.** a. Photoluminescence map of the difference between right and left circularly polarized emission intensities when the device is pumped with right circularly polarized light: $\delta I = I_R - I_L$, as a function of the gate voltage $V_{\text{TG}}$ in single-gate configuration. The dashed lines are the same as in Fig. 2b and serve as guides for the eye. b–d. Details of device operation in dual-gating mode. Left panels show photoluminescence spectra for $V_{\text{TG}} = +8$ V, 0 V and −8 V. Right panels show spatial imaging of $\Delta I_E$ ($\delta I_E$ integrated over the spectrum) in the corresponding gate configurations. The silicon back-gate is kept at $V_{\text{BG}} = 10 V_{\text{TG}}$ to reach higher doping densities and further enhance the effect. Scale bar, 5 µm.

**Fig. 4 | Polarization switching action.** Plot of the difference between right and left circularly polarized emission intensities when the device is pumped with right circularly polarized light: $\delta I = I_R - I_L$, as a function of the gate voltage $V_{\text{TG}}$. The resulting polarization is obtained by integrating over the entire measured emission spectrum.
Discussion

The appearance of such a double interlayer transition and its opposite polarizations have been recently captured by several theoretical studies. Here we mainly discuss our experimental results within the picture of moiré-induced effects, while a more detailed discussion covering other mechanisms can be found in Supplementary Section 12. In this model, the potential induced by the moiré creates an array of localized emitters, with different properties determined by the local symmetry. Such a periodic pattern indeed exists in our devices, owing to the good alignment and the small lattice mismatch between WSe₂ and MoSe₂. Yu et al. have calculated that the different selection rules at some of these locations can enable otherwise forbidden spin-flip transitions. We find that this mechanism could explain our experimental observations, including the existence of two optical transitions with opposite helicities separated by about 25 meV, the peculiar temperature dependence (the higher-energy IX₁ transition is enhanced at higher temperature owing to the thermal excitation of excitons) and the polarization switching with electrostatic doping. Now we discuss the distinct doping dependence in detail, by again considering the effect of the moiré potential. The energy minimum for interlayer excitons is found at the R₃₅ locations of the moiré for R-type (that is, AA- or zero angle-stacked) WSe₂/MoSe₂ (see ref. 23), where local selection rules allow both spin-conserving (IX₁) and spin-flipping transitions (IX₂), coupled with opposite polarizations of light, with comparable optical matrix elements (see also Supplementary Section 12). The different types of local energy minima R₃₅ instead show an opposite coupling with polarized light for IX₁ and a coupling with linearly polarized light for IX₂. This picture resembles our data very well: when doping our structure, we alter the moiré potential landscape (by filling some minima, or by shifting their energy electrostatically), thus causing a different type of symmetry location to become the most favourable one for excitons. This is accompanied by a change in the coupling with light for IX, sign change of ΔE₂, and in the loss of polarization for IX, in the circular basis (ΔI₂ becomes zero as it is now coupled with linearly polarized light). The observation of similar Stark shifts for IX₁ and IX₂ is also consistent with this picture: these two transitions correspond to excitons localized in moiré superlattice sites with the same symmetry, and thus are expected to have similar dipole moments. Emissions from different types of moiré sites would show different dipoles (owing to the changing interlayer distance), and consequently exhibit significantly different slopes (potentially crossing, as in ref. 23). The magnetic field-induced splitting of IX₁ and IX₂ confirms the AA-type stacking of the layers in our device (as for AB-stacked layers g₉-h≈–15 from ref. 8). Finally, we would like to stress that other theoretical investigations, including twin momentum space indirect transitions and the presence of both a direct and indirect transition also successfully explain some aspects of our observations.

In conclusion, we have demonstrated comprehensive electrical control over the polarization, wavelength and intensity of emission from interlayer excitons in a WSe₂/MoSe₂ van der Waals heterostructure. The ability to integrate all these functions in a single device to fine-tune the emitted radiation is key to practical optoelectronics and could pave the way for novel applications for valleytronic devices. Even more importantly, polarization conservation and reversal have been demonstrated to be gate-tunable, enabling a polarization-inverting action. We also report large and opposite g-factors for both IX₁ and IX₂ transitions, which fit the exciton–moiré interaction described here. We note that although this proof of principle shows the feasibility of such a device, further optimization and engineering to increase its performance and efficiency will be needed to realize practical valley devices. These results are relevant in the context of photonic and valleytronic devices, since they enable the electrical manipulation of the polarization of light and deepen our fundamental understanding of interlayer excitons in these structures.
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Author contributions
A.K. initiated and supervised the project. A.C. fabricated the devices. K.W. and T.T. grew the materials. D.U. analysed the data with input from A.A. and A.K. All authors contributed to the writing of the manuscript.

Competing interests
The authors declare no competing interests.

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Methods
Heterostructure fabrication. Few-layer graphene flakes for the bottom gate were obtained by exfoliation from graphite (NGS) on Si/SiO₂ substrates and patterned in the desired shape by electron-beam lithography and oxygen plasma etching. 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, 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 and minimize the stacking angle. For this, a homemade software was used to measure the angle between the flake edges, with a precision limited by the resolution of optical images (<1°). Polymer residue was removed with a hot acetone bath. Once completed, the stack was thermally annealed under high-vacuum conditions at 10⁻⁶ mbar for 6 h. Finally, electrical contacts were fabricated using electron-beam lithography and metallization (80-nm Pd for contacts, 8-nm Pt for the top gate).

Optical measurements. All measurements presented in the work were performed under vacuum at a temperature of 4.2 K. Excitons were optically pumped by a continuous-wave 647-nm laser diode focused to the diffraction limit with a beam size of about 1 µm. The incident power was about 200 µW. The spectral and spatial characteristics of the device emission were analysed simultaneously. The emitted light was acquired using a spectrometer (Andor Shamrock with Andor Newton CCD camera), and the laser line was removed with a long-pass 650-nm edge filter. For spatial imaging, we used a long-pass 850-nm edge filter so that the laser light and most of the emission from monolayers were blocked. Filtered light was acquired by a CCD camera (Andor Ixon). For polarization-resolved measurements, a λ/4 plate on a rotator together with a linear polarizer were used to select the polarization of incident light. A similar setup was used to image the two polarizations on the CCD camera. To acquire separate spectra for left and right circularly polarized light, a λ/4 plate and a displacer were employed. For magnetic-field-dependent photoluminescence measurements, the extraction of the peak energy shift was done using the method shown in ref. 32, which allows us to obtain better precision without making assumptions about the peak shape. A linear fit of the resulting values was then used to extract the effective g-factor, taking the error in the determination of each point as a weighting factor. The error reported in the text is the resulting uncertainty on the fit parameter.

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