Quenching the bandgap of two-dimensional semiconductors with a perpendicular electric field

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Perpendicular electric fields can tune the electronic band structure of atomically thin semiconductors. In bilayer graphene, which is an intrinsic zero-gap semiconductor, a perpendicular electric field opens a finite bandgap. So far, however, the same principle could not be applied to control the properties of a broader class of 2D materials because the required electric fields are beyond reach in current devices. To overcome this limitation, we design double ionic gated transistors that enable the application of large electric fields of up to 3 V nm$^{-1}$. Using such devices, we continuously suppress the bandgap of few-layer semiconductors with a perpendicular electric field. Owing to the huge capacitance of electrolytes (~50 F cm$^{-2}$), the required electric fields can be reached (see Supplementary Note 5 for a detailed analysis). Perpendicular electric fields can quench the bandgap of few-layer semiconductors, earlier studies have shown only a 10% gap reduction (or less) at fields reached using conventional double-gated transistors.

Electric fields that are sufficient to quench the gap of atomically thin semiconductors can be achieved in ionic gate transistors. Using the large capacitance of electrolytes (~50 μF cm$^{-2}$), ionic gated devices can accumulate charge densities—unattainable with conventional gating—that enable the observation of new physical phenomena. Associated with these high densities are extremely large electric fields, which in a double gate geometry can be controlled independently of the accumulated charge by applying opposite voltages to the top and back gates. So far, however, technical complexities have prevented the realization of double ionic gated devices. Here we demonstrate double ionic gate transistors that enable the application of electric fields up to 3 V nm$^{-1}$, and we use them to fully quench bandgaps as large as 1.6 eV in WSe$_2$ bilayers and thicker multilayers.

Double ionic gate transistors for large electric fields

In our devices (see Methods and Supplementary Note 1 for details on device fabrication), two electrolytes in contact with two independent gate electrodes are coupled to the same atomically thin semiconductor (see Fig. 1a). The top electrolyte is a commonly used ionic liquid (IL; that is, [P14]$^+$-[FAP]$^-$) and the bottom electrolyte is a lithium-ion (Li-ion) glass ceramic substrate (LASPT (Li$_2$Al$_2$SiP$_2$TiO$_{13}$)). The atomically thin semiconductor is connected to two metal source and drain electrodes and is surrounded by a ground plane (an aluminium film sandwiched between two alumina layers, that is, Al$_2$O$_3$/Al). The ground plane leaves only the 2D semiconductor exposed to the IL and eliminates any direct electrostatic coupling of the top and bottom electrolytes. This is important: if the two electrolytes were in direct contact, they would equilibrate their potential (as happens in recently realized double-side ionic gated devices), preventing the potential of the bottom and the top electrolyte being controlled independently. A perpendicular electric field with no net charge accumulation is established across atomically thin multilayers by applying a positive voltage to the IL gate (that is, the IL gate voltage, $V_{\text{IL}}$) and a negative voltage to the back gate (that is, the back gate voltage, $V_{\text{BG}}$). Because of the very large capacitance of both electrolytes, most of the applied potential difference drops across the gated material, enabling electric fields in excess of 3 V nm$^{-1}$ to be reached (see Supplementary Note 5 for a detailed analysis).

Quenching the bandgap of 2D semiconductors

In single ionic gate devices, conductivity measurements as a function of the gate voltage provide spectroscopic capabilities, enabling bandgaps of individual 2D semiconductors to be determined quantitatively. In double-gated devices, we apply the same technique to determine the presence or absence of a gap. In simple terms, gate-induced transport can be mediated by electrons when a positive gate voltage sets the chemical potential in the conduction band or by holes when a negative applied gate voltage sets the chemical potential in the valence band. The two regimes are separated by a gate voltage interval in which transport is strongly suppressed, with the chemical potential located in the gap. To determine whether or not the gap can be quenched, we look at the interval of gate...
voltages over which transport is suppressed and see if its extension can be reduced to zero by applying a perpendicular electric field. To this end, we measure the gate-induced device conductivity as a function of $V_{\text{IL}}$ and $V_{\text{BG}}$ and plot it versus $V = V_{\text{IL}} + V_{\text{BG}}$ (i.e., the potential applied to the semiconductor channel) for different values of $E^* = V_{\text{IL}} - V_{\text{BG}}$ (proportional to the applied electric field; see Supplementary Note 5). Before discussing data plotted in this way, it is, however, useful to analyse different aspects of the device behaviour.

When one gate voltage is swept whilst keeping the other gate grounded, double gate devices function as conventional transistors, as illustrated by the transfer curves (the source–drain current, $I_{\text{SD}}$, as a function of the gate voltage, $V_{\text{BG}}$ or $V_{\text{IL}}$) recorded for a bilayer (2L) WSe$_2$ device (see Fig. 1b,c), measured with $V_{\text{IL}} = 0$ V and $V_{\text{BG}} = 0$ V, respectively; for additional characterization measurements, see Supplementary Note 2). These curves are virtually identical to those measured for single-gated devices, but for increasingly negative $V_{\text{BG}}$ values (Fig. 2a), transfer curves of $I_{\text{SD}}$ versus $V_{\text{IL}}$ evolve. For $V_{\text{BG}} = -0.4$ V, the transfer curve is still similar to that measured for $V_{\text{BG}} = 0$ V (Fig. 1c): the current $I_{\text{SD}}$ increases as $V_{\text{IL}}$ is swept past the threshold for electron accumulation ($V_{\text{IL}} = 1.8$ V), and no current flows for $V_{\text{IL}} < 1.8$ V, when the chemical potential is in the gap. At $V_{\text{BG}} = -1$ V, however, the transfer curve shows qualitative differences, as current flows even when $V_{\text{IL}}$ is well below 1.8 V. As $V_{\text{BG}}$ is set to more negative values, $I_{\text{SD}}$ remains large for all values of $V_{\text{IL}}$, without ever vanishing (the square resistance $R_S \approx h/e^2$ for all $V_{\text{IL}}$, where $h$ is Planck’s constant and $e$ is the elementary charge). Analogous considerations hold true when looking at the evolution of the curve of $I_{\text{SD}}$ versus $V_{\text{IL}}$ or applying a positive voltage $V_{\text{IL}}$ to the IL gate (Fig. 2b).

The colour plot in Fig. 2c shows the complete evolution of $I_{\text{SD}}$, as a function of $V_{\text{IL}}$ and $V_{\text{BG}}$. The measurements are fully reproducible and reversible, as discussed in Supplementary Note 4. The observed behaviour is not that expected if the only effect of the gate voltages was to affect the electrostatic potential in the transistor channel, that is, the mechanism that determines the operation of a conventional transistor. In that case, both $V_{\text{IL}}$ and $V_{\text{BG}}$ would just shift the energy of the 2L WSe$_2$ bands, and a change in the voltage applied to one gate would only cause a rigid shift in the transfer curve plotted as a function of the voltage applied to the other. The data, however, do not show a rigid shift. The observed behaviour is also not that expected for devices in which the two gates are decoupled, i.e., that in which transport would be mediated by two accumulation layers of electrons and holes on opposite surfaces of the bilayer (see Supplementary Note 6 and Supplementary Fig. 10).

The reason for the unexpected behaviour is that the concomitant application of a large positive $V_{\text{IL}}$ and a large negative $V_{\text{BG}}$ generates a perpendicular electric field that quenches the bandgap. To gain further understanding, we look at the evolution of transport as $V_{\text{IL}}$ and $V_{\text{BG}}$ are varied continuously along the contour outlined by the coloured lines in Fig. 2c. At point $A$, $V_{\text{IL}} = 0$ V and $V_{\text{BG}} = -2.4$ V. The negative potential $V_{\text{BG}}$ results in the accumulation of holes (see Fig. 1b) and sets the chemical potential in the WSe$_2$ valence band. As $V_{\text{IL}}$ is increased from 0 to 2.4 V at the fixed $V_{\text{BG}}$ of $-2.4$ V, we travel from points A to B (purple line) and the electrostatic potential $V^*$ applied to the channel vanishes. Nevertheless, the current remains large. If $V_{\text{BG}}$ is then decreased from $-2.4$ to 0 V at the fixed $V_{\text{IL}}$ of 2.4 V, we move from B to C (green line), where transport is mediated by electrons accumulated by the large positive voltage $V_{\text{IL}}$ (see Fig. 1c). Transport therefore evolves from being mediated by holes at point A (with the chemical potential in the valence band) to being mediated by electrons at point C (with the chemical potential in the conduction band), without ever passing through a highly insulating state (see Fig. 2d). This is possible only if the gap closes and the
conduction and valence bands overlap along some part of the contour, with electrons and holes coexisting in the transistor channel. Indeed, this happens in the neighbourhood of B, where the electric field perpendicular to the 2L WSe₂, which is proportional to \( E^* \), is maximum.

We now plot \( I_{SD} \) as a function of \( V^* = (V_{IL} + V_{BG}) \) and \( E^* = (V_{BG} - V_{IL}) \) (Fig. 3a). Figure 3b shows \( I_{SD} - V^* \) curves measured at fixed values of \( V^* \). As expected, at small \( E^* \) (for example, 1.5 V), \( I_{SD} \) is finite for a sufficiently large negative and positive \( V^* \)—with the current carried by holes and electrons, respectively—and vanishes over an extended \( V^* \) interval as the chemical potential is swept across the 2L WSe₂ gap (the square resistance in this regime is larger than our measurement sensitivity). The \( V^* \) interval with vanishing current shrinks as \( E^* \) increases, and no highly resistive state is observed for \( E^* \) larger than a threshold value \( E^*_t \), which we determine quantitatively below. This signals a transition to a highly conductive state with increasing applied perpendicular electric field, which becomes apparent by looking at the evolution of the \( I_{SD} - E^* \) curve at fixed values of \( V^* \), that is, by looking at vertical cuts of the colour plot shown in Fig. 3a. In Fig. 3c, we plot such a cut for \( V^* = 0.5 \) V (from the dashed green line in Fig. 3a), showing that, for sufficiently large values of \( E^* (>3 \text{ V}) \), \( I_{SD} \) increases with increasing \( E^* \) without a change in the electrostatic potential applied to the channel. We conclude that in the presence of a sufficiently large electric field, a finite conductivity of the order of \( e^2/\hbar \) is present, irrespective of the position of the Fermi level, indicating that states are available at all energies to mediate transport. This implies that no gap is present and that the application of a sufficiently large electric field fully quenches the 2L WSe₂ bandgap.

To determine \( E^*_t \), we extract the electron and hole threshold voltages \( (V_{Th}^e \text{ and } V_{Th}^h, \text{ respectively, as shown in Fig. } 3b) \), plot their difference \( \delta = V_{Th}^e - V_{Th}^h \) as a function of \( E^* \) and look at when \( \delta \) vanishes (Fig. 3d). The corresponding electric field is then approximately given by \( E^*_t / \tau_{TMD} \approx 3.0 \text{ V nm}^{-1} \), where \( \tau_{TMD} = 1.3 \text{ nm} \) is the thickness of the 2L WSe₂. This is a good approximation because of the very large gate capacitance (\( C \approx 50 \mu \text{F cm}^{-2} \) (refs. 13,14,16–18)), which ensures that the voltage drop across the electrolytes is small (see discussion in Supplementary Note 5). We conclude that the electric field \( E^*_t \) needed to quench the 2L WSe₂ gap is between 2.5 and 2.7 V nm

**Fig. 2** | Electrical characteristics of a double-gated 2L WSe₂ transistor. a, \( I_{SD} \) measured for a 2L WSe₂ device as a function of \( V_{IL} \) for different negative values of \( V_{BG} \). The curves evolve from showing textbook transistor behaviour at a small negative \( V_{BG} \) value (see also Fig. 1b) to not showing any sizable current suppression at a large negative \( V_{BG} \). Same as for a, with \( I_{SD} \) measured as a function of \( V_{BG} \) for different positive values of \( V_{IL} \); the evolution of the transistor curves is fully analogous to that seen in a. c, Colour plot of \( I_{SD} \) (in logarithmic scale) as a function of \( V_{BG} \) and \( V_{IL} \). Note that the simultaneous application of a large negative \( V_{BG} \) and an equally large positive \( V_{IL} \) causes the current in the transistor to increase by between four and five orders of magnitude, despite leaving the potential of the transistor channel \( (V^* = V_{IL} + V_{BG}) \) unchanged. d, Evolution of \( I_{SD} \) along the A–B–C contour illustrated in c (where the coordinates of A, B and C in the \( (V_{IL}, V_{BG}) \) plane are indicated on the bottom axis). Transport in the transistor is mediated by holes at A and by electrons at C; finding that there is no region where the current is fully suppressed implies that, along part of the A–B–C contour, the valence and conduction bands of WSe₂ must overlap. In all measurements, \( V_{SD} \) = 0.1 V.
sufficiently large applied electric field, a finite density of states is present at all energies, that is, the gap closes. Because the multilayers are only a few atoms thick, a fully quantum mechanical calculation is needed to understand how the gap closes. In simple qualitative terms, however, a sufficiently large perpendicular electric field quenches the gap because the electrostatic potential lifts the energy of the valence-band edge at one crystal surface above the conduction band edge at the opposite surface. The conduction and valence bands then overlap, and the system becomes gapless.

One may then wonder if the closing of the gap and the gate dependence of the conductivity that we observe is not simply due to the formation of two decoupled accumulation layers—one of electrons and one of holes—at the opposite multilayer surfaces, as happens in macroscopically thick crystals. However, neither modelling the gate dependence in terms of the decoupled accumulation layers nor the experimental data measured on a 70-nm-thick double ionic gated WSe2 crystal reproduce the key aspects of the calculations (compare Fig. 1d and Fig. 1e), which show that at a large electric field, the gap closes as the minimum of the conduction band at the Q point in the Brillouin zone touches the top of the valence band at the K point (see Fig. 1e). We extracted from these calculations the full thickness dependence of the critical electric field $E_c$ needed to quench the gap and found excellent quantitative agreement with the values of $E_c$ measured experimentally (see Fig. 4h). This agreement provides strong support for our interpretation.

**Nature of the gapless state**

The discussion above provides a basic physical scenario for the nature of the gapless electronic state of atomically thin WSe2 multilayers. At charge neutrality, for applied electric fields $E > E_c$, these systems are compensated semimetals with a finite band overlap that increases with increasing $E$, with coexisting electron and hole states. The semimetal has broken inversion symmetry, owing to the large electric field present. It is uniform in the plane of the multilayer, but the ‘centres of mass’ of the electrons and holes are displaced relative to each other in the direction perpendicular to the layers. As the electric field is increased, the electron and hole states are pushed away from each other due both to the increasing potential difference

**Fig. 3** Bandgap evolution as a function of the electric field. a, Colour plot of $I_{sd}$ (in logarithmic scale) measured for a 2L WSe2 device as a function of $V^* = V_L + V_{BG}$ and $E^* = V_E - V_{BG}$ (respectively proportional to the electrostatic potential in the transistor channel and to the electric field perpendicular to the 2L WSe2 crystal). The width of the $V^*$ interval over which $I_{sd}$ is suppressed decreases monotonically on increasing $E^*$. b, For a quantitative analysis, we look at horizontal cuts of the colour plot in a for $I_{sd}$ versus $V^*$ at a fixed $E^*$ (values indicated in the figure). The curve measured at $E^* = 4.5$ V shows a complete suppression of $I_{sd}$ and the thin black lines illustrate how we determine the threshold voltage for electron conduction ($V_{th}^*$) and hole conduction ($V_{th}^*$) (by extrapolating $I_{sd}$ to zero for positive and negative values of $V^*$, and the positions of the threshold voltages are marked by the vertical dashed lines). c, Cut of the colour plot in a, taken at $V^* = 0.5$ V, that corresponds to the vertical green dashed line. The data show a transition from a highly resistive state at low $E^*$ to a state with a conductivity of the order $\hbar/2e^2$ at $E^* > 4$ V. This transition, which occurs at fixed $V^*$, is a direct manifestation of the quenching of the bandgap caused by the applied electric field. d, Plot of the difference $\delta = V_{th}^* - V_{th}^*$ as a function of $E^*$, to find the value of $E^*$ for which $\delta = 0$ V ($E^* = 4$ V in our 2L WSe2 device) to determine the condition at which the gap closes.
between the two surfaces and to the fact that, as the electron and hole densities increase, the screening length may eventually become shorter than the thickness. At that point, the system will effectively consist of electron- and hole-accumulation layers that are coupled only by Coulomb attraction. For the thinnest multilayers, such as 2L and 3L WSe₂, however, the thickness is so small that full overlap of the electron and hole wavefunctions remains relevant even at the largest experimentally accessible densities. An investigation of transport and of the optical response as a function of the perpendicular electric field and temperature will clearly be essential for understanding the properties of atomically thin WSe₂ multilayers with an electrically tunable bandgap.

**Fig. 4 | Quenching the gap in 3L, 4L and 5L WSe₂ devices.**

a,c,e. Colour plots of \( I_{SD} \) as a function of \( V^* = V_L + V_{bg} \) (the sum of the IL and back gate voltages) and the electric field \( \mathcal{E}' \) for 3L (a), 4L (c) and 5L (e) devices. b,d,f. Corresponding cuts (\( I_{SD}-V^* \) curves) for different fixed values of the electric field \( \mathcal{E}' \) across each WSe₂ multilayer for the 3L (b), 4L (d) and 5L (f) devices. g. Comparison between the maximum electric fields that can be applied across \( nL \) WSe₂ using different types of device. Green dots: field achieved in ionic gated devices for \( \mathcal{E}' = V_L - V_{bg} = 5 \) V (larger values are possible, as we repeatedly reached 5.5 and 6 V; see Supplementary Note 5 for details on the estimation of \( \mathcal{E}' \)); blue and orange dots: maximum field reachable with devices based on hexagonal boron nitride (hBN) assuming a breakdown field of 1.1 and 0.6 V nm⁻¹, respectively (1.1 V nm⁻¹ is the ultimate limit reached in ultra-thin hBN; 0.6 V nm⁻¹ is a more realistic estimate for common hBN). For 2L WSe₂, the electric field reachable with ionic gating is nearly one order of magnitude larger than the field accessible in hBN-based devices. The black crosses are the field needed to close the gap, as extracted from our measurements; for all thicknesses, it would have not been possible to close the gap with hBN-based devices. h, Comparison between the critical electric field \( \mathcal{E}_c \) values extracted from experiments (black crosses) and the corresponding theoretical values obtained from first-principles calculations (open diamonds). Ab initio calculations slightly underestimate the gap, resulting in a small underestimation of \( \mathcal{E}_c \). To take this into account, the filled diamonds show the value of the critical electric field obtained using the ratio between the known and the calculated gap to rescale \( \mathcal{E}_c \) (see Supplementary Note 8). The agreement with the experimental data is excellent.
Conclusion

We have established the ability to quench a semiconducting band-gap larger than 1.6 eV in 2D semiconductors as thin as 2L WS2 (that is, just over 1 nm thick), by applying a very large perpendicular electric field, close to 3 V nm−1. The ability to control the semiconducting gap and to reversibly apply electric fields of 3 V nm−1 onto a 2D material have great potential for future studies. Indeed, many theoretical studies have been reported, predicting that the electronic properties of a variety of atomically thin crystals can be drastically altered if fields of this strength can be reached in the experiments. Examples include topological transitions in transition metal dichalcogenides (TMDs)44–46 and other 2D materials47, the ability to switch the magnetic anisotropy48 or the topological charge of magnetic excitations49 and control of the electronic state and other properties of different van der Waals materials and of their heterostructures50–52. So far, these theoretical predictions have been considered to be exclusively of academic interest, because the required electric fields were nearly one order of magnitude larger than those reachable in practice. The results shown here change the situation as they enable a vast gamut of unexplored electronic phenomena to be investigated experimentally.

Note added at submission: at the time of submission, we became aware of a manuscript by Weintrub et al. that has just appeared on the ‘cond-mat’ public archive53 and that reports experiments similar to those discussed here.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41565-022-01183-4.

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Methods

Device fabrication. The double ionic gate transistors employed in our experiments use two electrostatically decoupled electrolytes to apply large perpendicular electric fields across atomically thin WSe2 crystals. The top gate consists of a commonly used IL, 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate ([P14]3-,[FAP]+; purchased from Merck Millipore, now VWR) coupled to a platinum/gold (Pt/Au) electrode, and the bottom gate consists of a Li-ion conductive glass ceramic (LASPT; purchased from MTI) coupled to a chromium/gold (Cr/Au) electrode. The atomically thin WSe2 crystals, ranging from 2L to 7L (and the 70 nm bulk-like crystal), were isolated via the micromechanical cleavage of bulk crystals (purchased from HQ Graphene) onto silica/silicon (SiO2/Si) substrates and then transferred onto the Li-ion conductive glass ceramic using a common dry pick-up-and-transfer technique48. As described in Supplementary Note 1, atomically thin crystals can be identified optically on the glass ceramic38, but it is preferable to transfer them from the SiO2/Si substrates to avoid compromising the integrity of the Al2O3/Al/Al2O3 trilayer that decouples the two electrolytes. The electrical contacts to the crystals, the gate and reference (see below) electrodes, are defined using electron-beam lithography, electron-beam evaporation and lift-off, or evaporation through a shadow mask (see Supplementary Note 1 for complete details on the electrode deposition).

To isolate and electrostatically decouple the Li-ion glass ceramic from the IL, we use an Al ground plane sandwiched between two Al2O3 layers (see Supplementary Note 1 for details on the deposition of the Al2O3/Al/Al2O3 trilayer). The presence of the ground plane is crucial for correct device operation, as without it the two electrolytes would equilibrate their potential and the two gates could no longer be controlled independently. The effectiveness of the electrostatic decoupling can be checked experimentally using two reference electrodes in direct contact with either the Li-ion glass or the top IL, as discussed in detail in Supplementary Note 3. The Al2O3/Al/Al2O3 trilayer also prevents diffusion of Li ions from the substrate into the IL, which would cause degradation of the IL itself.

Transport measurements. All the electrical measurements performed during this work were carried out at room temperature and under high vacuum (1 × 10−7 mbar), using home-built voltage and current amplifiers in conjunction with d.c. source-measuring units (Keithley K2400) and digital multimeters (Agilent 3410).

In the double-gated experiments, all measurements were carried out by applying a positive voltage to the IL and a negative voltage to the back gate, since under these conditions, the Li ions in the glass substrate are pushed away from the semiconducting layer and cannot intercalate. Under these conditions, the measurements are fully reversible and reproducible. If the gate voltages were swept sufficiently slowly—the typical sweeping rate in our measurements was 5 mV s−1—the measurements exhibited negligible or at most small hysteresis originating from bias stress effects (see Supplementary Note 4). In different measurements, the applied source–drain bias was varied between 10 and 100 mV, to keep it smaller or bias stress effects (see Supplementary Note 4). In different measurements, the used IL, 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate ([P14]3-,[FAP]+; purchased from Merck Millipore, now VWR) coupled to a platinum/gold (Pt/Au) electrode, and the bottom gate consists of a Li-ion conductive glass ceramic (LASPT; purchased from MTI) coupled to a chromium/gold (Cr/Au) electrode. The atomically thin WSe2 crystals, ranging from 2L to 7L (and the 70 nm bulk-like crystal), were isolated via the micromechanical cleavage of bulk crystals (purchased from HQ Graphene) onto silica/silicon (SiO2/Si) substrates and then transferred onto the Li-ion conductive glass ceramic using a common dry pick-up-and-transfer technique48. As described in Supplementary Note 1, atomically thin crystals can be identified optically on the glass ceramic38, but it is preferable to transfer them from the SiO2/Si substrates to avoid compromising the integrity of the Al2O3/Al/Al2O3 trilayer that decouples the two electrolytes. The electrical contacts to the crystals, the gate and reference (see below) electrodes, are defined using electron-beam lithography, electron-beam evaporation and lift-off, or evaporation through a shadow mask (see Supplementary Note 1 for complete details on the electrode deposition).

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First-principles simulations. To compute the critical electric field Ec at which the energy gap of a WSe2 multilayer vanishes, we carried out first-principles density functional theory simulations in the Perdew–Burke–Ernzerhof46 generalized-gradient approximation as implemented in the Quantum ESPRESSO distribution29. Here we outline the main technical aspects of our calculations.

In our calculations, the crystal structure of the multilayers is obtained from the experimentally reported structure of bulk WSe2, by cutting out a certain number of layers, without further relaxations. The Brillouin zone is sampled with a 2×2×1 k-point grid in a uniform Γ-centred Monkhorst–Pack grid. Electron–ion interactions are treated with a fully relativistic pseudopotential approach using the norm-conserving procedure as refined in the optimized norm-conserving Vanderbilt pseudopotential (or ONCVSP) approach47 with parameters from the Pseudo DOJO library50 and a cutoff energy on wavefunctions of 80 Ry. Spurious interactions with periodic replicas of the multilayers in the direction orthogonal to the layers are suppressed using a cutoff on long-range interactions46. The band structure is computed over a fine grid using Wannier interpolation46, and the gap is computed as the energy difference between the bottom of the conduction bands and the top of the valence bands. For each multilayer, the energy bandgap is computed for several values of the electric field E by putting charged sheets with uniform and opposite charge densities (±σ) at the two sides of the multilayers—thus mimicking a double gate set up—through the implementation of ref. 51. The results of the simulations are discussed in Supplementary Note 8.

Data availability

The data that support the findings of this study are available free of charge from the Yareta repository of the University of Geneva at https://doi.org/10.26037/yareta:txap4ayzibcm5hcipvvzufx72a. This repository contains the data presented in all figures, including those in the Supplementary Information.

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