

Stabilization of sliding ferroelectricity through exciton condensation

Supplementary Information

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CONTENTS

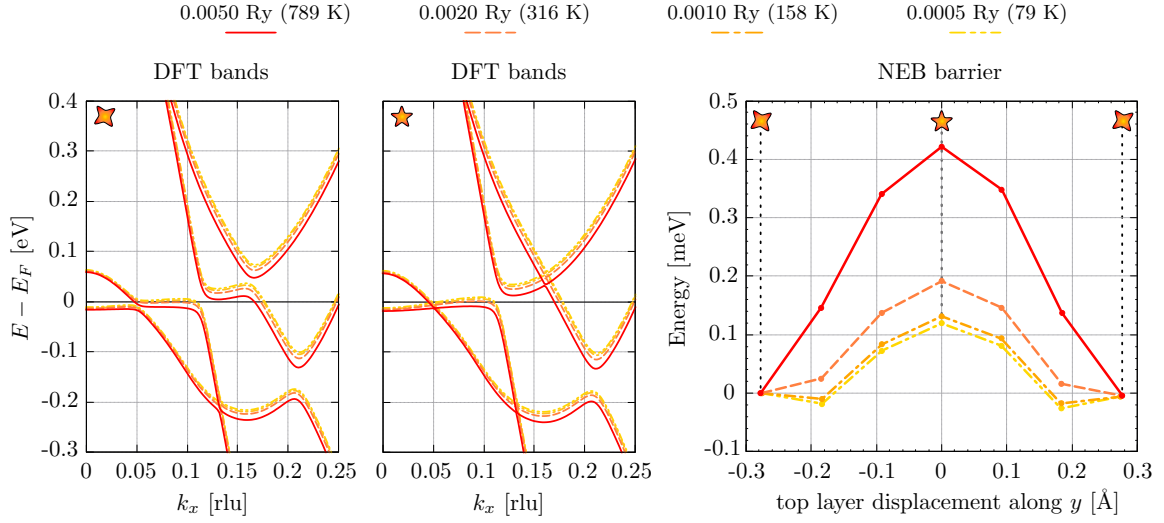
I. Estimate of the DFT-NEB ferroelectric switching barrier	2
II. Four-band model	4
A. Noninteracting Hamiltonian	4
B. Bethe-Salpeter equation and Coulomb interaction	4
C. Excitons and spontaneously broken symmetry of the EI phase	6
D. Self-consistent mean-field theory of the excitonic insulator	7
E. Discussion	8
III. Sensitivity of model to parameters and validation from first principles	9
IV. Spin-orbit coupling	11
References	12

I. ESTIMATE OF THE DFT-NEB FERROELECTRIC SWITCHING BARRIER

In this section we apply the nudged elastic band (NEB) method, as implemented in the DFT framework [1], to estimate the energy barrier of the sliding ferroelectric switching. We take special care of exploiting the Fermi-Dirac occupation function to account for the electronic temperature through the smearing parameter, and discuss how this impacts the NEB energy barrier, as well as the DFT band structure.

In the following we report results for a smearing parameter decreasing from 5×10^{-3} Ry (i.e., ≈ 800 K) down to 5×10^{-4} Ry (≈ 80 K). Though 800 K may look like a large value, this is the typical order of magnitude used to speed up the convergence of the self-consistent DFT calculation, particularly when dealing with a coarse $\mathbf{k}$ -point sampling of the Brillouin zone combined with a Fermi-Dirac occupation function. As a lower smearing parameter requires a denser Brillouin-zone sampling to achieve numerical convergence, we increased the $\mathbf{k}$ -point grid from 22×12 to 66×36 , which yields fully converged results.

Furthermore, as the interplay between $\mathbf{k}$ -point density and the smearing parameter influences the equilibrium configuration through the evaluation of forces, we performed variable-cell relaxations to optimize both the atomic positions and the lattice parameters for each given combination of smearing parameter and $\mathbf{k}$ -point grid.



Supplementary Figure 1: **DFT and NEB results at different electronic temperatures.** DFT band structures of bilayer WTe₂ (left and center panels) and NEB energy barrier of the sliding ferroelectric switching (right panel) using a 66×36 $\mathbf{k}$ -point grid for the different smearing parameters reported on top. The smearing parameter fixes the electronic temperature through the Fermi-Dirac occupation function. The band structures in the left panel correspond to the equilibrium configuration obtained via variable-cell relaxation (diamond symbol), whereas those in the center panel to the glide-mirror symmetric (GMS) structure found by the NEB calculation (star symbol). In the left and center panels the bands are shifted by their respective Fermi energy, whereas in the right panel each NEB curve is shifted so that its first image (i.e. the starting configuration of the reaction path) is at zero energy. The values on the horizontal axis of the right panel correspond to the relative sliding displacement between the layers, with zero displacement corresponding to the GMS configuration. The circles on the NEB curves mark the images used for the discretization of the reaction path, and the connecting lines are a guide to the eye.

As shown in the left and center panels of Supplementary Figure 1, the smearing parameter changes the DFT band structure—already converged for a 22×12 grid—without affecting the magnitude of the gaps at $k_x \approx 0.12$ rlu and $k_x \approx 0.21$ rlu. Therefore, our results for the excitonic renormalization of the switching barrier, presented in the main text, are unaffected. However, the smearing parameter strongly affects the occupation of valence states for $0.05 < k_x < 0.12$ rlu, which are filled only for temperatures higher than ≈ 300 K. This band region is remarkably flat not only along k_x but also along k_y (cf. Supplementary Figure 3c), hence providing a sizable contribution to the density of states in the Fermi energy window. Similarly, a significant enhancement of the density of states originates from the conduction band for $0.12 < k_x < 0.17$ rlu, though at higher temperature and sliding displacement (cf. Figure 2 of the main text).

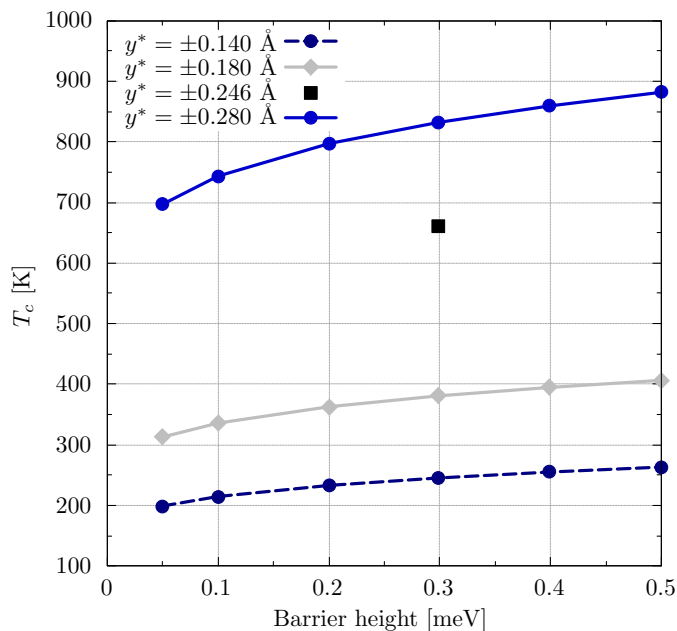
The above behavior points to a wide variation of Gibbs free energy (the extensive counterpart of the chemical potential) with temperature, and reflects in the extreme sensitivity of the NEB potential barrier to the smearing parameter. This is illustrated in the right panel of Supplementary Figure 1, where the energy zero for each curve is

fixed to the energy of the respective starting configuration. For given layer displacement coordinate, the NEB energy potential increases with the smearing parameter, the maximum variation corresponding to zero displacement (GMS structure). This may be easily rationalized, as the valence band of the GMS configuration is almost perfectly flat for a significant k_x range (center panel of Supplementary Figure 1), and hence its filling is most sensitive to temperature.

This results in a general reduction of the switching barrier with decreasing temperature. The trend shown in the right panel of Supplementary Figure 1 suggests that, at zero temperature, the barrier stabilizes to a value around 0.1 meV. The barrier “width”—the horizontal sliding displacement that connects the two ferroelectric ground states with opposite dipole—shows a similar trend, i.e., it shrinks as the electronic temperature is lowered.

The barrier height values previously published were obtained with the same methodology but were significantly higher: 0.6 meV [2, 3] and 0.3 meV [4]. We note that in those works smaller $\mathbf{k}$ -point grids were used. In conclusion, the huge suppression of the NEB potential barrier height we report may be ascribed to the very high value of the electronic density of states at Fermi energy, i.e., the extreme compressibility of Fermi liquid. We expect no such effect for the incompressible Fermi system of a large-gap semiconductor, like h-BN.

Interestingly, the increase in the switching barrier with temperature was recently measured (above 100 K) in bilayer WTe₂ and attributed to the appearance of the Fermi surface [3]. The experiment might be related to the results reported in the main text, as the predicted excitonic indirect gap varies between 3 meV (≈ 35 K, GMS structure) and 7 meV (≈ 80 K, $\bar{d}$ displacement) with sliding.



Supplementary Figure 2: **Ferroelectric switching Curie temperature (T_c) after Tang and Bauer (2023).**

The plot reports the numerical solution of equation (19) of Ref. 5. Here y^* is used to indicate the relative sliding displacement between the GMS configuration and the minimum energy configuration (i.e., half the NEB barrier width). The black square points to the estimate for bilayer WTe₂ given in Ref. 5. The other curves are estimates of T_c for both larger and lower values of barrier height and width, for a range of values relevant to the NEB barrier results of Supplementary Figure 1. The connecting lines are a guide to the eye.

According to the thermodynamic mean-field theory of Ref. 5, the key to the high Curie temperature (T_c) of sliding ferroelectrics is the rigidity of the layer sliding, which proceeds with a macroscopic mass. In addition to structural parameters (unit cell area and the 2D Lamé coefficients), presumably unchanged in our treatment with respect to the values reported in Table I of Ref. 5, T_c is most sensitive to the switching barrier height and width, as shown in Supplementary Figure 2. From Supplementary Figure 1 we estimate a barrier height of ≈ 0.1 meV and width $y^* \approx 0.180$ Å, leading to $T_c \approx 330$ K, which is half the original prediction for bilayer WTe₂ (black square dot [5]). As the mean-field treatment of the layer sliding dynamics is known to overestimate T_c by neglecting quantum fluctuations, it remains unclear whether the DFT treatment alone is able to explain the room-temperature ferroelectricity of bilayer WTe₂. This uncertainty points to the relevance of the excitonic enhancement of the switching barrier height discussed in the main text.

II. FOUR-BAND MODEL

We develop the four-band model mentioned in the main text to investigate the onset of macroscopic excitonic coherence in bilayer WTe_2 , as a function of the relative sliding of the two layers, y . This model fully complies with the space group symmetry of the bilayer for all y , in the absence of e-h interaction, and provides a transparent picture of the orbital character of the exciton wave function. This allows us to analyze the breaking of microscopic symmetries due to exciton condensation.

A. Noninteracting Hamiltonian

Within the envelope function approximation, the model provides the two lowest conduction (c) and two highest valence (v) bands by spanning the Hilbert space through a four-dimensional orbital basis. In this representation, the $\mathbf{k}$ -resolved noninteracting Hamiltonian, $\hat{H}(\mathbf{k})$, is a 4×4 matrix. The basis is made of the Bloch states at Γ obtained *ab initio* for the *glide-mirror symmetric* (GMS) configuration ($y = 0$), after a rotation. The non rotated states, plotted in Supplementary Fig. 3b, are either even or odd with respect to the symmetry operations of the space group, i.e.: (i) the glide-mirror operation, $\mathbb{G}$, a combination of mirror reflection $z \rightarrow -z$ and half-period translation $y \rightarrow y + b/2$ along the y axis (b is the lattice constant along y); (ii) the mirror reflection $x \rightarrow -x$, $\mathbb{M}$, with the vertical mirror plane yz intersecting the row of W atoms at the origin.

As apparent from Supplementary Fig. 3b, the Bloch states symmetrically spread between the two layers. Since we are interested in the charge transfer and consequent asymmetry between layers, we rotate the basis frame to introduce a layer index, σ_z , as the eigenvalue of the 2×2 z -component Pauli matrix, $\hat{\sigma}_z$, with eigenvectors $|\chi_{\sigma_z}^{\pm}\rangle$ obeying $\hat{\sigma}_z|\chi_{\sigma_z}^{\pm}\rangle = \pm|\chi_{\sigma_z}^{\pm}\rangle$. Here the pseudospinors $|\chi_{\sigma_z}^{+}\rangle$ and $|\chi_{\sigma_z}^{-}\rangle$ are linear combinations of (either conduction or valence) Bloch states, and are localized respectively in the top and bottom layer. Furthermore, as the $\mathbb{M}$ symmetry survives for $y \neq 0$, we introduce a second orbital pseudospin, τ_z , corresponding to orbitals respectively even ($\tau_z = +1$, top row of Supplementary Fig. 3b) or odd ($\tau_z = -1$, bottom row) under reflection, i.e., $\hat{\tau}_z|\chi_{\tau_z}^{\pm}\rangle = \pm|\chi_{\tau_z}^{\pm}\rangle$. The complete basis set stems from the external product of the two pseudospinors, $|\chi_{\sigma_z}^{\pm}, \chi_{\tau_z}^{\pm}\rangle \equiv |\chi_{\sigma_z}^{\pm}\rangle \otimes |\chi_{\tau_z}^{\pm}\rangle$. In this representation, the symmetry operations are

$$\mathbb{G} = \hat{\sigma}_x \otimes \mathbb{I}_{\tau}, \quad \mathbb{M} = \mathbb{I}_{\sigma} \otimes \hat{\tau}_z, \quad (1)$$

where $\mathbb{I}_{\sigma}$ and $\mathbb{I}_{\tau}$ are the 2×2 unit matrices in the layer and orbital pseudospin spaces, respectively.

The eigenvalues of the noninteracting Hamiltonian for $y = 0$, $\hat{H}_0(\mathbf{k})$, are—by construction—the DFT energy bands of the GMS structure, $\varepsilon_{M,G}^0(\mathbf{k})$, with the subscripts $M = \pm$ and $G = \pm$ labeling the parity of the corresponding eigenvector with respect to $\mathbb{M}$ and $\mathbb{G}$ operations, respectively. The explicit form of $\hat{H}_0(\mathbf{k})$ is

$$\begin{aligned} \hat{H}_0(\mathbf{k}) = & \left(\frac{\mathbb{I}_{\sigma} + \hat{\sigma}_x}{2} \right) \otimes \left(\frac{\mathbb{I}_{\tau} + \hat{\tau}_z}{2} \right) \varepsilon_{+,+}^0(\mathbf{k}) + \left(\frac{\mathbb{I}_{\sigma} - \hat{\sigma}_x}{2} \right) \otimes \left(\frac{\mathbb{I}_{\tau} + \hat{\tau}_z}{2} \right) \varepsilon_{+,-}^0(\mathbf{k}) + \\ & \left(\frac{\mathbb{I}_{\sigma} + \hat{\sigma}_x}{2} \right) \otimes \left(\frac{\mathbb{I}_{\tau} - \hat{\tau}_z}{2} \right) \varepsilon_{-,+}^0(\mathbf{k}) + \left(\frac{\mathbb{I}_{\sigma} - \hat{\sigma}_x}{2} \right) \otimes \left(\frac{\mathbb{I}_{\tau} - \hat{\tau}_z}{2} \right) \varepsilon_{-,-}^0(\mathbf{k}). \end{aligned} \quad (2)$$

The energy bands are plotted in Supplementary Fig. 3a and c respectively along the ΓX cut and in the rectangular sector of the Brillouin zone $k_x \in [0, 0.35]$, $k_y \in [-0.1, 0.1]$.

For finite displacement y , we add to the total Hamiltonian, $\hat{H}(\mathbf{k}) = \hat{H}_0(\mathbf{k}) + \hat{H}_1$, the term $\hat{H}_1$, which is the interaction between the electron and the “frozen” phonon corresponding to the rigid sliding of the two layers,

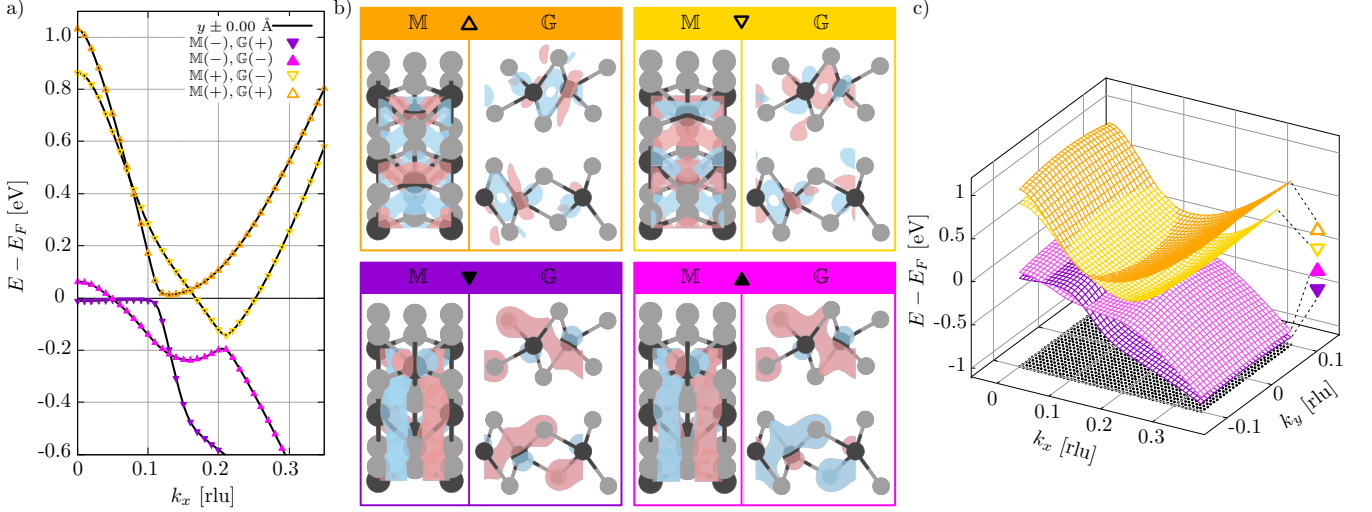
$$\hat{H}_1 = (\hat{\sigma}_z \otimes \mathbb{I}_{\tau}) \delta_{H_1}. \quad (3)$$

Here the “phonon” parameter δ_{H_1} depends on y but not on $\mathbf{k}$ (see Supplementary Section IIE for its estimate). The term (3) breaks the glide-mirror symmetry, as it does not commute with $\mathbb{G}$, and explicitly accounts for the layer polarization through the average value of $\langle \hat{\sigma}_z \rangle$ over the ground state.

B. Bethe-Salpeter equation and Coulomb interaction

Within the four-band model, the Bethe-Salpeter equation of motion for excitons that generalizes the expression (2) of the main text is

$$\sum_{v',c',\mathbf{k}'} H_{v\mathbf{c}\mathbf{k}}^{v'c'\mathbf{k}'} \Psi_x^{v'c'\mathbf{k}'} = E_x \Psi_x^{v\mathbf{c}\mathbf{k}}. \quad (4)$$



Supplementary Figure 3: **Orbital basis and symmetry.** a) Energy bands for the GMS structure along the ΓX cut of the Brillouin zone computed from density functional theory, as in Fig. 2 of the main text. The superimposed triangle symbols (open/filled and up/down) point to the symmetry classification of the states at Γ , tabulated in both legend and panel b. Since the glide-mirror symmetry is preserved along the ΓX line and there are no anticrossings, the symmetry labeling is extended to all $\mathbf{k}$ points, as the orbital character of each labeled band changes smoothly as one moves through the Brillouin zone. b) Wave functions (real part) at Γ of the highest two valence and lowest two conduction bands. The amplitude map colour points to the parity of the wave function with respect to mirror (M) and glide-mirror (G) symmetry operations, the red [blue] corresponding to the positive [negative] amplitude. In each one of the four panels, the left inset shows the top view of the bilayer (x axis in the horizontal direction) and the right inset shows the lateral view (y axis in the horizontal direction). Dark and light gray atoms are W and Te, respectively. c) Three-dimensional plot of the bands in a selected rectangular range of the Brillouin zone, illustrating the symmetry classification in the $\mathbf{k}$ space.

Now $\Psi_x^{vc\mathbf{k}}$ is the component of the exciton wave function that gives the probability amplitude for exciting an electron from the valence band v to the conduction band c , conserving the momentum $\mathbf{k}$. The indices v and c label two of the four energy bands derived from the solution of $\hat{H}_0 + \hat{H}_1$, as defined in (2) and (3). The kernel of (4) is

$$H_{vc\mathbf{k}}^{v'c'\mathbf{k}'} = [\varepsilon_c(\mathbf{k}) - \varepsilon_v(\mathbf{k})] \delta_{vv'} \delta_{cc'} \delta_{\mathbf{k}\mathbf{k}'} - W_{vc\mathbf{k}}^{v'c'\mathbf{k}'}, \quad (5)$$

and contains the screened electron-hole Coulomb attraction, W . Importantly, since the model provides us with the layer index σ_z for the eigenvectors of $\hat{H}_0(\mathbf{k}) + \hat{H}_1(\mathbf{k})$, we can generalize the form of the two-dimensional Rytova-Keldysh potential to the bilayer system [6]. Therefore, we resolve the Coulomb interaction matrix element, $W_{vc\mathbf{k}}^{v'c'\mathbf{k}'}$, as

$$W_{vc\mathbf{k}}^{v'c'\mathbf{k}'} = W(\mathbf{k} - \mathbf{k}', 0) \left[\langle \psi_v^T(\mathbf{k}) | \psi_{v'}^T(\mathbf{k}') \rangle \langle \psi_c^T(\mathbf{k}) | \psi_{c'}^T(\mathbf{k}') \rangle^* + \langle \psi_v^B(\mathbf{k}) | \psi_{v'}^B(\mathbf{k}') \rangle \langle \psi_c^B(\mathbf{k}) | \psi_{c'}^B(\mathbf{k}') \rangle^* \right] + \\ W(\mathbf{k} - \mathbf{k}', h) \left[\langle \psi_v^T(\mathbf{k}) | \psi_{v'}^T(\mathbf{k}') \rangle \langle \psi_c^B(\mathbf{k}) | \psi_{c'}^B(\mathbf{k}') \rangle^* + \langle \psi_v^B(\mathbf{k}) | \psi_{v'}^B(\mathbf{k}') \rangle \langle \psi_c^T(\mathbf{k}) | \psi_{c'}^T(\mathbf{k}') \rangle^* \right]. \quad (6)$$

Here $|\psi_i^T(\mathbf{k})\rangle$ and $|\psi_i^B(\mathbf{k})\rangle$ are the projections of the i th eigenvector ($i = c, v$) onto the top and bottom layer, respectively, and the screened interaction is

$$W(\mathbf{q}, z) = \frac{e^2}{2\varepsilon_0 A} \frac{1}{q \epsilon(q, z)}, \quad (7)$$

with

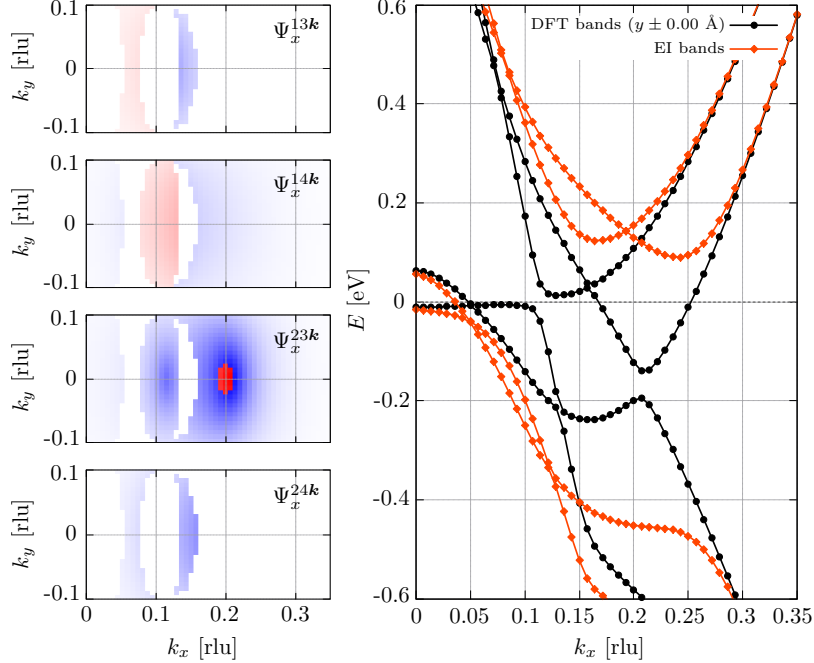
$$\epsilon(q, z) = (1 + 2\pi \alpha_{2D} q) \cosh(zq) + [2 + 4\pi \alpha_{2D} q (2\pi \alpha_{2D} q)^2] \sinh(zq)/2. \quad (8)$$

In the above equations, $h = 6.9$ Å is the distance between the two layers, chosen as the distance between the planes parallel to the WTe_2 sheets passing through the W atoms, and z is the vertical coordinate. We set $z = 0$ ($z = h$) for intralayer (interlayer) interaction.

C. Excitons and spontaneously broken symmetry of the EI phase

The lowest exciton state for the GMS structure is plotted in the left column of Supplementary Fig. 4, the four contour maps in $\mathbf{k}$ plane corresponding to different vc pairs of the wave function component $\Psi_x^{vc\mathbf{k}}$. As clear from the colour map, the amplitude with the largest weight is $\Psi_x^{23\mathbf{k}}$ (third panel from top), which qualitatively overlaps with the modulus of the exciton wave function derived within the two-band model. The latter is shown in Fig. 3a of main text for finite y ; the corresponding plot for $y = 0$ is qualitatively similar. The corrections introduced by the four-band model mainly consist in expanding the size of the Hilbert space that spans the exciton state, by means of adding wave function components. Note, though, that the weight of new components $\Psi_x^{vc\mathbf{k}}$ decreases with the increase of the e-h excitation energy for vc pair of bands located farther from Fermi energy. For the same reason, the maximum weight of $\Psi_x^{23\mathbf{k}}$ occurs in $\mathbf{k}$ space close to the position of the gaps separating bands 2 and 3.

Since the total momentum of the exciton is zero, the state must transform under symmetry operations according to the space group at Γ . It may be easily recognized from Supplementary Fig. 4 that the exciton wave function is even under the glide-mirror operation and odd under specular reflection (see for example the parity of e-h pairs at Γ). We checked that, for all $y \neq 0$, the lowest exciton state remains odd under reflection, whereas the glide-mirror symmetry is lifted by the sliding. This behavior rules the broken symmetry of the EI phase, since the noninteracting ground state wave function, which is even under specular reflection, is distorted by the hybridization with the (odd) exciton wave function at the onset of condensation [7]. It follows that the excitonic insulator must spontaneously break the mirror symmetry $x \rightarrow -x$, which is otherwise preserved in the absence of e-h attraction, for all y . This hints at “excitonic ferroelectricity” [8], which is driven by the macroscopic condensation of the interband exciton dipoles, here aligned along the x axis. Note that such exciton macroscopic dipole is orthogonal to the vertical macroscopic polarization that is observed experimentally. We expect the inclusion of spin-orbit coupling in the theory not to alter this picture, but possibly lead to additional symmetry breakings of magnetic type.



Supplementary Figure 4: **Bethe-Salpeter and EI gap equations within the four-band model.** Exciton wave function (left) and EI band reconstruction (right) within the four-band model for $\delta_{H_1} = 0$ (GMS structure). The panels on the left show the exciton wave function components, $\Psi_x^{vc\mathbf{k}}$, where $v = 1, 2$ and $c = 3, 4$ are the indices of the two valence and two conduction bands, in ascending order of energy. The range of the color scale is $[-0.1, 0.1]$, with positive [negative] values in red [blue]. As in Fig. 3 of the main text, the maximum amplitude is reached at the two locations along the ΓX cut of the energy gaps separating bands 2 and 3. The corrections to the bands in the EI phase, given by $\Delta_{ij}(\mathbf{k})$ (see main text), increase with the magnitudes of the amplitudes $\Psi_x^{vc\mathbf{k}}$. Note that, with respect to Fig. 3 of the main text, here the Brillouin zone sampling is reduced along the k_y direction. This significantly impacts the exciton binding energy, thus a lower screening ($\alpha_{2D} = 1.9 \text{ \AA}$) has been used to gap the EI band structure.

D. Self-consistent mean-field theory of the excitonic insulator

As for the two-band model illustrated in the main text, also in the present four-dimensional framework the EI ground state, $|\Psi_{\text{EI}}\rangle$, is the Slater determinant of the filled, distorted valence band Bloch states,

$$|\Psi_{\text{EI}}\rangle = \prod_{i=1,2} \prod_{\mathbf{k}} \hat{\alpha}_{i\mathbf{k}}^\dagger |\text{vac}\rangle. \quad (9)$$

Here $|\text{vac}\rangle$ is the vacuum state with no electrons, $\hat{\alpha}_{i\mathbf{k}}^\dagger$ is the Fermi operator that creates an electron of momentum $\mathbf{k}$ in the i th renormalized band of the excitonic insulator,

$$\hat{\alpha}_{i\mathbf{k}} = \sum_j a_{ij\mathbf{k}} \hat{a}_{j\mathbf{k}}, \quad (10)$$

$\hat{a}_{j\mathbf{k}}^\dagger$ creates an electron in the j th (conduction or valence) band of the pristine semimetal, and we convene that $i = 1, 2$ and $i = 3, 4$ respectively label the EI valence and conduction bands. The coefficients $a_{ij\mathbf{k}}$ occurring in (10) form a unitary matrix for given $\mathbf{k}$, which must be determined self-consistently. The $a_{ij\mathbf{k}}$'s are the eigenvectors resulting from the diagonalization of the mean-field Hamiltonian, $\hat{H}_{\text{EI}}(\mathbf{k})$, the 4×4 matrix whose eigenvalues, $E_i(\mathbf{k})$, are the renormalized EI bands:

$$H_{\text{EI}}(\mathbf{k}) = \begin{pmatrix} \varepsilon_1(\mathbf{k}) & 0 & \Delta_{13}(\mathbf{k}) & \Delta_{14}(\mathbf{k}) \\ 0 & \varepsilon_2(\mathbf{k}) & \Delta_{23}(\mathbf{k}) & \Delta_{24}(\mathbf{k}) \\ \Delta_{31}(\mathbf{k}) & \Delta_{32}(\mathbf{k}) & \varepsilon_3(\mathbf{k}) & 0 \\ \Delta_{41}(\mathbf{k}) & \Delta_{42}(\mathbf{k}) & 0 & \varepsilon_4(\mathbf{k}) \end{pmatrix}. \quad (11)$$

Here the pristine bands, $\varepsilon_i(\mathbf{k})$, are the eigenvalues of the noninteracting Hamiltonian, $\hat{H}(\mathbf{k}) = \hat{H}_0 + \hat{H}_1$ [see (2) and (3)], and the (real) self-consistent interband hybridization, $\Delta_{ij}(\mathbf{k}) = \Delta_{ji}(\mathbf{k})$, is driven by the population of the exciton condensate. This term, which generalizes to multiple bands the gap function $\Delta(\mathbf{k})$ discussed in the main text, must be determined by solving the ‘‘gap equation’’, which complements the iterative cycle:

$$\Delta_{ij}(\mathbf{k}) = \sum_{i',j',\mathbf{k}'} W_{ij\mathbf{k}}^{i'j'\mathbf{k}'} \sum_{l,l'} [a_{\mathbf{k}'}^{-1}]_{j'l} [a_{\mathbf{k}'}^{-1}]_{i'l'} \langle \hat{\alpha}_{l\mathbf{k}'}^\dagger \hat{\alpha}_{l'\mathbf{k}'} \rangle_{\Psi_{\text{EI}}}. \quad (12)$$

Note that in (12)—and only there—the indices $i, i' = 1, 2$ and $j, j' = 3, 4$ run only over the valence and conduction states, respectively, $W_{ij\mathbf{k}}^{i'j'\mathbf{k}'}$ is given by (6), and the coefficients $[a_{\mathbf{k}'}^{-1}]_{jl}$ are obtained by inverting the eigenvector matrix $a_{ij\mathbf{k}}$. We evaluate the quantum (and thermal) average $\langle \hat{\alpha}_{l\mathbf{k}'}^\dagger \hat{\alpha}_{l'\mathbf{k}'} \rangle_{\Psi_{\text{EI}}}$ occurring in (12) as

$$\langle \hat{\alpha}_{l\mathbf{k}'}^\dagger \hat{\alpha}_{l'\mathbf{k}'} \rangle_{\Psi_{\text{EI}}} = \delta_{l,l'} f[E_l(\mathbf{k})], \quad (13)$$

where $f(\cdot)$ is the Fermi-Dirac distribution function. At zero temperature, for the intrinsic excitonic insulator phase, we set f to 1 for valence states and 0 for conduction states.

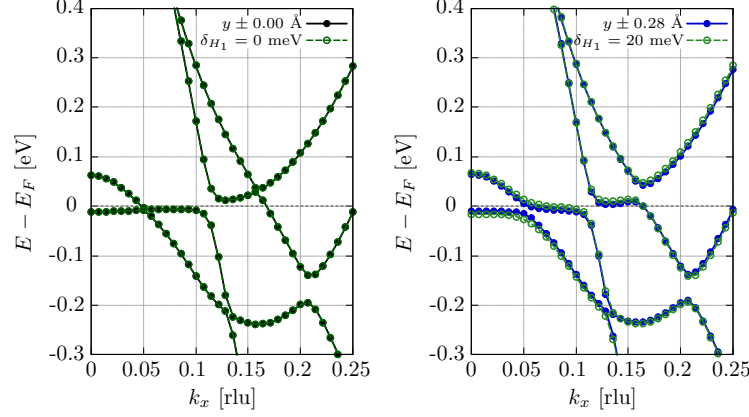
The numerical self-consistent cycle starts by using the components of the lowest-energy exciton, $\Psi_x^{ij\mathbf{k}}$, derived from the solution of the four-band Bethe-Salpeter equation (4), as a seed for $\Delta_{ij}(\mathbf{k})$ that enters the matrix (11). After $\hat{H}_{\text{EI}}$ is diagonalized, the resulting orthogonal eigenvector matrix is inverted to evaluate the coefficients $[a_{\mathbf{k}'}^{-1}]_{jl}$ entering (12). The new value of $\Delta_{ij}(\mathbf{k})$ is then inferred by performing the sum over $\mathbf{k}'$ in (12), and then used to update (11) at the next step. The cycle is repeated until $\Delta_{ij}(\mathbf{k})$ converges for all $\mathbf{k}, i, j$.

For completeness, we note that, in the presence of doping, or at finite temperature, or in the ‘excitonic semimetal’ scenario, the set of self-consistent equations reported above must be complemented by an additional condition, the total charge balance, which implicitly defines the chemical potential, μ . Such equation must be solved for μ at each step of the iterative cycle, as the chemical potential enters the average (13) through the Fermi distribution function. In principle, also the screened interaction W should be evaluated self-consistently, as the variation of μ impacts the screening of charge carriers [9]. In the simplest case of an intrinsic excitonic insulator at zero temperature, μ lies in the gap and the charge balance condition is unnecessary.

The method is illustrated in the right column of Supplementary Fig. 4, which compares the noninteracting and EI band structure for $\delta_{H_1} = 0$ and $\alpha_{2\text{D}} = 1.9 \text{ \AA}$, with a 50×29 $\mathbf{k}$ -point grid. Note that the renormalization of the bottom conduction and top valence bands due to e-h interaction compares with that assessed within the two-band model, as shown in Fig. 3 of main text.

E. Discussion

The four-band model reported in this Supplementary Note provides valuable insight into the understanding of the variation of the band structure with y , the nature of the condensing exciton, and the consequent spontaneous breaking of the crystal symmetry. On the other hand, it has a few caveats, which we discuss in the following and that ultimately led us to present the two-band results in the main text.



Supplementary Figure 5: **Comparison between model and first-principles bands.** Left: Model noninteracting bands, computed for $\delta_{H_1} = 0$ (open circles), compared to first-principles bands for the GSM structure, $y = 0$ (filled circles). Right: Model noninteracting bands, computed for $\delta_{H_1} = 20$ meV (open circles), compared to first-principles bands of the bilayer structure with the top layer displaced by ± 0.28 Å (filled circles). In both panels the bands are plotted along the ΓX cut of Brillouin zone ($k_y = 0$).

The first issue is the modeling of layer sliding through the envelope-function Hamiltonian $\hat{H}_1$ of (3), which does not depend on $\mathbf{k}$. This simplification allows for assessing the “frozen phonon” parameter, δ_{H_1} , by maximizing the overlap with the target DFT bands computed for a given sliding displacement. For example, for ± 0.280 Å displacement of the top layer from the GSM structure, we find $\delta_{H_1} = 20$ meV (right panel of Supplementary Figure 5). However, a non-local, $\mathbf{k}$ -dependent δ_{H_1} parameter would be needed to match exactly the evolution of band anticrossing with y , as predicted from density functional theory. This matter is illustrated in Supplementary Fig. 5. Whereas in the left panel the matching of first-principles (filled circles) and model (open circles) bands is complete by construction ($y = 0$), in the right panel ($y = \pm 0.28$ Å) the model conduction band crossing the Fermi energy around $k_x = (0.15)2\pi/a$ overestimates the gap opening, while the valence band underestimates it. Though the discrepancy is tiny on the energy scale of the plot, the total energy gain associated with exciton binding is amplified by condensation, leading to a strong renormalization of the sliding barrier, as discussed in the main text.

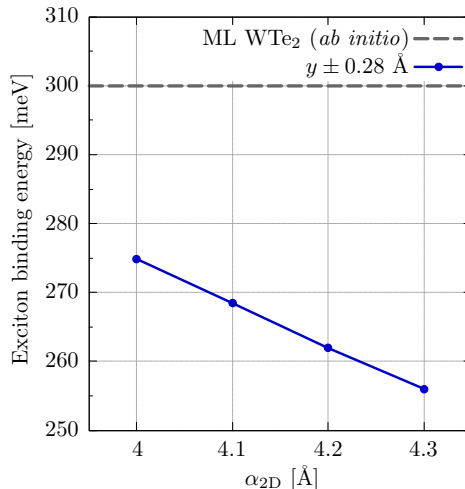
Another concern is the envelope-function approximation at the basis of the four-band model, whose reliability deteriorates as one moves from the Brillouin zone center. The issue becomes severe for $|k_y| > (0.1)2\pi/b$, due to the anticrossing behavior of bands that is not well reproduced by the model. This limits our ability to check the convergence of the solution of the Bethe-Salpeter equation (4) for large interaction strength. For this reason we relied eventually on the two-band model of the main text, based on energy bands purely derived from first principles, which allows us to extend arbitrarily the Brillouin zone sampling.

III. SENSITIVITY OF MODEL TO PARAMETERS AND VALIDATION FROM FIRST PRINCIPLES

Bilayer WTe_2 exhibits a tiny gap of a few meV, excitonic in nature [10], but is predicted to be a semimetal from DFT (Fig. 2 of main text). This poses a formidable computational challenge, since a full first-principles treatment of excitons would require the calculation of the GW correction to band structure, which includes dynamical screening effects, as well as the evaluation of the intraband contribution to electronic polarizability. Such high-level tasks are essential to capture excitonic effects in the presence of a two-dimensional Fermi surface [11–16]. Furthermore, the calculation of the EI phase is inherently self-consistent, in principle including the re-evaluation of screening at each step of the iterative cycle. This is relevant, as the static and dynamical GW correlations tend to decrease the semimetal band overlap and increase the range of the e-h attraction that binds excitons, which in turn may condense and open the gap. Eventually, the screening behavior of the reconstructed EI ground state is compatible with that of a narrow-gap semiconductor rather than that of the initial semimetal state.

Since the GW description of the bilayer is presently out of reach, we developed the two-band model approach of the main text. Luckily, two first-principles investigations recently reported consistent results for monolayer WTe_2 [10, 17], which we can use to validate the bilayer model, as we expect similar screening behaviour and exciton binding. Namely, Wu *et al.*, starting from a DFT semimetal ground state, showed [17] the tendency of the GW self-energy (in both flavors G_0W_0 and self-consistent COHSEX) to remove the Fermi surface by opening a gap. We expect a similar effect for the bilayer, which will tend to suppress the intraband contribution to screening. Importantly, the GW-corrected band structure by Wu *et al.* compares well with the one we derived previously (via hybrid DFT) to compute excitons from first principles [10]. Therefore, below we use the findings of Ref. 10 for the monolayer to benchmark those of the bilayer.

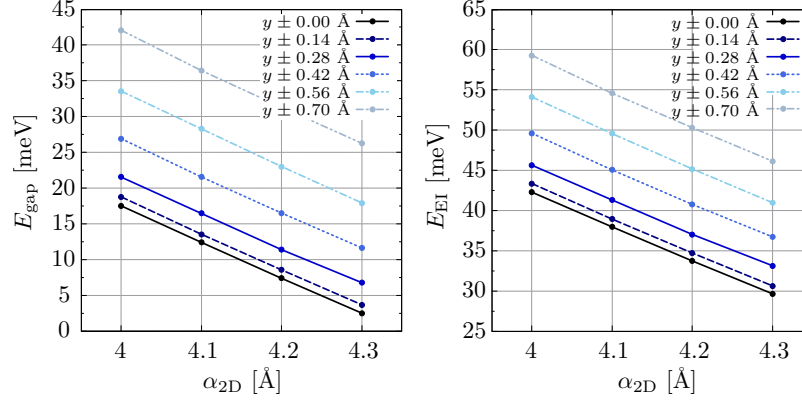
The two-band model critically relies on the parametrization of the screened e-h Coulomb attraction W via the polarizability α_{2D} [equation (3) of main text]. We cannot fix α_{2D} from the DFT calculation, since the predicted ground state is semimetallic: The occurrence of the spurious Fermi surface implies an intraband contribution to α_{2D} , which diverges at long wave length. The only consistent way to fix α_{2D} is to choose a value that results in the opening of a gap, which hence suppresses the intraband polarizability. Indeed, the chosen value of $\alpha_{2D} = 4.3 \text{ \AA}$ provides a tiny many-body indirect gap that compares with experiments [10]. The resulting gap is 3 meV for $y = 0$ and 7 meV for $y = \bar{d}$ (Fig. 3b of main text, EI band structure).



Supplementary Figure 6: **Exciton binding energy upon variation of the screening parameter.** Binding energy of the lowest exciton, resulting from the solution of Bethe-Salpeter equation (1) of main text, versus screening parameter α_{2D} . The results were obtained for sliding displacement $y \pm 0.28 \text{ \AA}$, using a 30×60 $\mathbf{k}$ -point grid sampling the region $k_x \in [0, 0.3]$ rlu and $k_y \in [-0.3, 0.3]$ rlu. The lines connecting the dots are a guide to the eye. The dashed line highlights the exciton binding energy for monolayer (ML) WTe_2 , computed from first principles [10].

We may justify further the chosen value of α_{2D} by contrasting the exciton binding energy inferred from W with the first-principle prediction for the monolayer [10]. This is illustrated in Supplementary Fig. 6, which compares the model exciton binding energy (dots) with the reference value [10] of 300 meV for the monolayer (dashed line), in the range $4 \leq \alpha_{2D} \leq 4.3 \text{ \AA}$. In general, as the screening of the e-h attractive potential increases with α_{2D} , the exciton binding energy decreases. Importantly, in the plotted range of α_{2D} the binding energy for the bilayer is smaller than the one for the monolayer, which matches the physical expectation of stronger screening in the bilayer. We disregard the outer

range of α_{2D} , since for $\alpha_{2D} \ll 4 \text{ \AA}$ the bilayer binding energy nonphysically exceeds the monolayer value, whereas for $\alpha_{2D} > 4.3 \text{ \AA}$ the resulting excitonic phase is semimetallic and the Rytova-Keldish form of W loses accuracy.

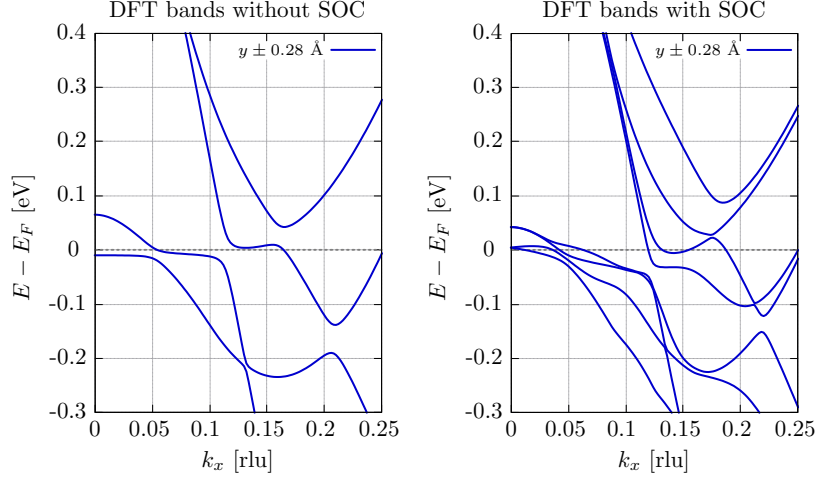


Supplementary Figure 7: **Energetics of the excitonic insulator vs screening and sliding displacement.** Many-body indirect gap in the EI phase, E_{gap} (left panel), and total energy gain, E_{EI} (right panel), versus screening parameter, α_{2D} , for various values of the sliding displacement, y . The connecting lines are a guide to the eye.

Like the exciton binding energy, also the EI gap function, $\Delta(\mathbf{k})$, converges to smaller values with increasing α_{2D} , which translates into weaker renormalization of EI bands and smaller size of the many-body indirect gap, E_{gap} . Remarkably, whereas E_{gap} is significantly sensitive to α_{2D} , the height of the sliding barrier potential is not. The reason is that the excitonic renormalization of the barrier height depends on the *variation* of E_{gap} with y , and not on its absolute magnitude. This is illustrated by Supplementary Fig. 7, which plots both E_{gap} (left panel) and the EI total energy gain, E_{EI} (right panel), vs α_{2D} , for different sliding displacements, y . Importantly, both curves are essentially parallel straight lines, namely, the variation of both E_{gap} and E_{EI} with y does not depend on the screening parameter, α_{2D} . In conclusion, the key result of our work, the excitonic renormalization of the height of the sliding barrier, is robust against the details of the two-band model.

IV. SPIN-ORBIT COUPLING

In the natural stacking of bilayer WTe_2 the inversion symmetry of the single-layer crystal structure is not preserved. As a result, spin-orbit coupling (SOC) splits the bands of bilayer WTe_2 by breaking Kramers degeneracy (Supplementary Fig. 8), while Bloch states remain doubly degenerate in monolayer WTe_2 [10]. Thus, the splitting effect must be attributed to inter-layer interaction, and may be tentatively rationalized as a Rashba-like term, $\sim \mathbf{S} \cdot (\mathbf{k} \times \mathbf{E})$, with $\mathbf{S}$ being the spin and $\mathbf{E}$ the microscopic electric field, after noting that the band splitting vanishes as $\mathbf{k} \rightarrow 0$ (right panel of Supplementary Fig. 8).



Supplementary Figure 8: **Effect of spin-orbit coupling on DFT bands.** DFT-PBE band structure without (left panel) and with (right panel) spin-orbit coupling (SOC). Results for the bilayer structure with sliding displacement $y \pm 0.28 \text{ \AA}$. The parameters of the DFT calculation with SOC are the same as in the Methods section of main text, with the exception that the corresponding full-relativistic pseudopotentials from the same library have been used [18].

Since our key findings depend on the variation of the total energy gain of the EI phase, E_{EI} , with the sliding displacement y , we expect the inclusion of SOC not to alter qualitatively our conclusions. In fact, even if the energy bands split with the SOC field, we expect the Rashba-like corrections, $\sim \pm |\mathbf{k} \times \mathbf{E}|$, to substantially cancel out when summing over occupied states to compute E_{EI} . An accurate statement would require to work out in detail the full numerical inclusion of SOC effects, as the intricacies of anticrossings shown in right panel of Supplementary Fig. 8 may compromise the reliability of any modelization. Such a task is beyond the scope of the present work.

On the other hand, SOC will qualitatively change the microscopic symmetries spontaneously broken by exciton condensation, as already mentioned in Supplementary Section II C. In particular, a magnetic instability could add to the breaking of the mirror symmetry $x \rightarrow -x$. The inclusion of SOC effects would significantly complicate the theory of the EI phase, by increasing the number of independent gap equations to solve self-consistently.

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