

Swapping exchange and spin-orbit coupling in ex-so-tic 2D heterostructures

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The concept of swapping the two most important spin interactions—exchange and spin-orbit coupling—is proposed based on two-dimensional multilayer van der Waals heterostructures. Specifically, we show by performing realistic *ab initio* simulations, that a *single* device consisting of a bilayer graphene sandwiched by a 2D ferromagnet Cr₂Ge₂Te₆ (CGT) and a monolayer WS₂, is able not only to generate, but also to swap the two interactions. The highly efficient swapping is enabled by the interplay of gate-dependent layer polarization in bilayer graphene and short-range spin-orbit and exchange proximity effects affecting only the layers in contact with the sandwiching materials. We call these structures *ex-so-tic*, for supplying either exchange (ex) or spin-orbit (so) coupling in a single device, by gating. Such *bifunctional* devices demonstrate the potential of van der Waals spintronics engineering using 2D crystal multilayers.

Keywords: spintronics, graphene, heterostructures, proximity spin-orbit coupling, proximity exchange

Introduction. In the paradigmatic spintronic device, the still elusive Datta-Das spin transistor [1], the spin-orbit coupling in a thin semiconductor channel rotates the spin of electrons flowing between two ferromagnetic electrodes. The transistor relies on one crucial aspect: a gate atop the channel modifies the magnitude of the spin-orbit interaction. As a result, one could electrically tune the spin precession between say 0 and 180 degrees, and turn the electron flow on or off. While other spin transistor designs have since been demonstrated [2–5], the principal difficulty of controlling spin interactions via electric fields remains as long as one deals with conventional semiconductor materials. Similar problems are faced by controlling exchange coupling electrically. In a pioneering experiment [6] 125 V was needed to turn the magnetic moment of ferromagnetic semiconductor InMnAs on and off.

The advent of 2D van der Waals heterostructures gave a strong push to spintronics [7, 8], with well-founded hopes for a far greater control over spin interactions such as spin-orbit and exchange couplings. The key mechanism, allowing such a control, is an interplay of the proximity effect [9] and gating. The past years have seen impressive progress in this direction, for both spin-orbit and exchange couplings individually [10–21].

Bilayer graphene (BLG) is emerging as a particularly useful platform for controlling the spin interactions by proximity effects. It was proposed that spin-orbit coupling in BLG can be turned on and off on demand [22, 23] by gating, as recently demonstrated by capacitance measurements [24]. Similar effects have been predicted for ferromagnetic encapsulation [25–27]. The next milestone would be a transport demonstration with sensitivity to the spin polarization of the bands, or the spin-orbit torque in the proximity setup [28–30].

In this paper we present a device structure which is capable of not only creating the two most important spin interactions—spin-orbit and exchange couplings—in an

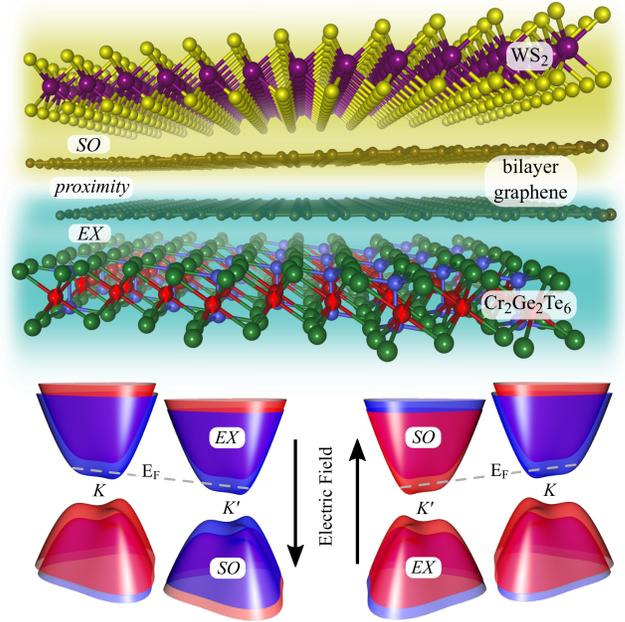


FIG. 1. Ex-so-tic van der Waals heterostructure. Top: BLG sandwiched between a monolayer TMDC (such as WS₂ or MoSe₂) and a monolayer ferromagnetic semiconductor (such as CGT or CrI₃). The magnetization of the ferromagnet is indicated by the arrows. The top layer of BLG is proximitized by the TMDC, acquiring a giant spin-orbit coupling, while the bottom layer of the BLG is proximitized by the ferromagnet, acquiring an exchange coupling. Bottom: Electric tunability of the low-energy bands at *K* and *K'*. The colors red and blue indicate out-of-plane spin. For a fixed Fermi level E_F (here in the conduction band), the Dirac electrons experience either exchange or spin-orbit coupling, depending on the electric field.

electronic system, but also swapping them on demand by an applied electric field. Being able to swap two different spin (or other effective) interactions by gating is

a striking thought, without precedence in the realm of conventional materials. While proximity spin-orbit coupling preserves time-reversal symmetry, and leads to such phenomena as topological quantum spin Hall effect [31] or giant spin relaxation anisotropy [15, 32–34], proximity exchange coupling breaks time reversal symmetry and renders a nominally nonmagnetic electronic system effectively magnetic [35]. Swapping the two interactions provides a reversible route between time-reversal symmetric and magnetic physics.

Our choice of the electronic platform for swapping the spin interactions is BLG, which is nicely suited for proximity-based devices by providing two coupled surfaces. Via layer polarization, which locks a given layer to an electronic band (or set of bands), transport properties can be strongly influenced by the environment. We use a CGT/BLG/WS₂ heterostructure, with monolayer CGT providing a strong proximity exchange effect to the bottom layer of BLG, and WS₂ to induce strong spin-orbit coupling to the top layer, see Fig. 1. Both CGT and WS₂ are semiconductors, so they contribute to transport properties of the heterostructure only by imprinting their characteristic spin properties onto the BLG.

Gating can swap spin-orbit and exchange couplings, as illustrated in Fig. 1. The applied electric field changes the layer polarization, which changes the layer-band assignment, thereby swapping the proximity exchange and spin-orbit couplings. This intuitive picture is supported below by realistic density functional theory (DFT) simulations and phenomenological modeling, predicting quantitatively the behavior of the ex-so-tic heterostructures in the presence of a transverse electric field.

Swapping spin-orbit and exchange coupling by gate. We consider a supercell stack containing BLG sandwiched between a monolayer WS₂ and a monolayer ferromagnetic CGT with an out-of-plane magnetization, as depicted in Fig. 1. We calculate the electronic states for this structure using DFT, see Supplemental Material [36]. The band structure along selected high-symmetry lines containing K point is shown in Fig. 2. The magnetization—from CGT—as well as the strong spin-orbit coupling—from TMDC—are manifested by the spin polarization of the bands. Inside the semiconductor band gap, there are well preserved parabolic electronic states of BLG. States relevant for transport form the four low-energy bands close to the Fermi level. It is these four bands that are at the focus of this work.

BLG *per se* has no band gap, has tiny spin-orbit coupling (on the order of tens of μeV s [37]), and has no magnetic exchange coupling. But sandwiched by the two monolayers, both orbital and spin properties of the low-energy bands of BLG strikingly change. First, the built-in dipole moment in the heterostructure [22, 27] separates conduction and valence bands, inducing a band gap for a given spin polarization. Next, the states exhibit a giant spin-orbit coupling induced from the TMDC layer.

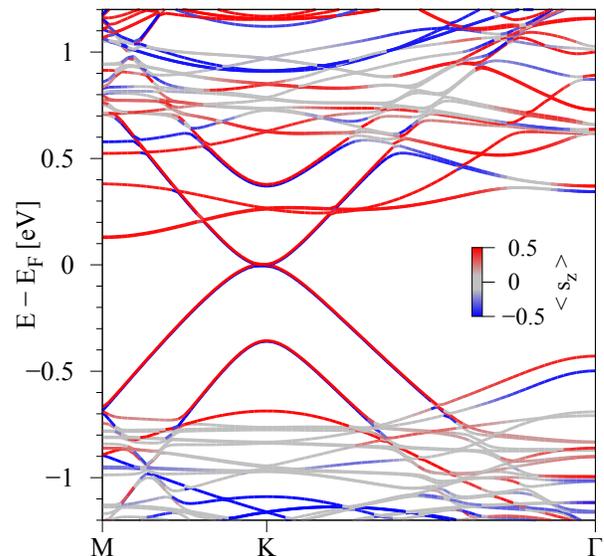


FIG. 2. Calculated electronic band structure of a WS₂/BLG/CGT stack along high symmetry path M-K- Γ . The color of bands (red and blue) corresponds to the s_z spin expectation value.

This coupling is opposite in K and K' points, as a consequence of time reversal symmetry of the spin-orbit interaction. Finally, the electronic states of BLG become magnetic, manifested by an exchange splitting, equal in K and K' , coming from the ferromagnetic CGT. It is the fascinating spectral separation of the spin-orbit coupling and exchange in the BLG which allows for their swapping.

The three aforementioned effects are nicely seen in Fig. 3 which shows a zoom to the low-energy band structure of the stack at both K and K' . Perhaps the most striking feature of the band structure is the difference in the energy dispersion at K and K' , which ultimately comes from the interplay between the induced spin-orbit and exchange couplings in BLG. While the valence and conduction bands partially overlap at K , there is a local band gap at K' . The dipole moment of the heterostructure is too weak to open a global (over the whole Brillouin zone) band gap. The spin splittings are 2–8 meV, which is experimentally significant.

An applied external electric field increases the band gap of the doubly proximitized BLG and makes explicit the effects of the TMDC and CGT layers. This is demonstrated in Fig. 3, which shows both the DFT results (for fields -0.5 , 0 , and 0.4 V/nm) and model calculations (for fields, -1 , -0.5 , 0 , 0.4 , and 1 V/nm) using an effective Hamiltonian introduced in the next section. Let us first look at negative electric fields, say -1 V/nm, which point down, towards CGT. The conduction bands are split by about 8 meV, having identical spin polarizations at K and K' , which means that this splitting is due to exchange coupling: the conduction bands are affected

TABLE I. Parameters of the model Hamiltonian \mathcal{H} , Eq. (1), fitted to the DFT low-energy dispersion data for the CGT/BLG/WS₂ heterostructure at zero applied electric field. Parameters γ are in eV, others in meV. The dipole is given in debye. Unspecified model parameters are zero. Based on the fit parameters for the individual BLG/CGT and WS₂/BLG subsystems given in the Supplementary Information, we assumed $\lambda_I^{A2} = -\lambda_I^{B2}$ and $\lambda_{\text{ex}}^{A1} = \lambda_{\text{ex}}^{B1}$ here for the fit.

γ_0	γ_1	γ_3	γ_4	V	Δ	λ_I^{A2}	λ_I^{B2}	λ_{ex}^{A1}	λ_{ex}^{B1}	E_D	dipole
2.432	0.365	-0.273	-0.164	-0.474	8.854	1.132	-1.132	-3.874	-3.874	0.348	0.398

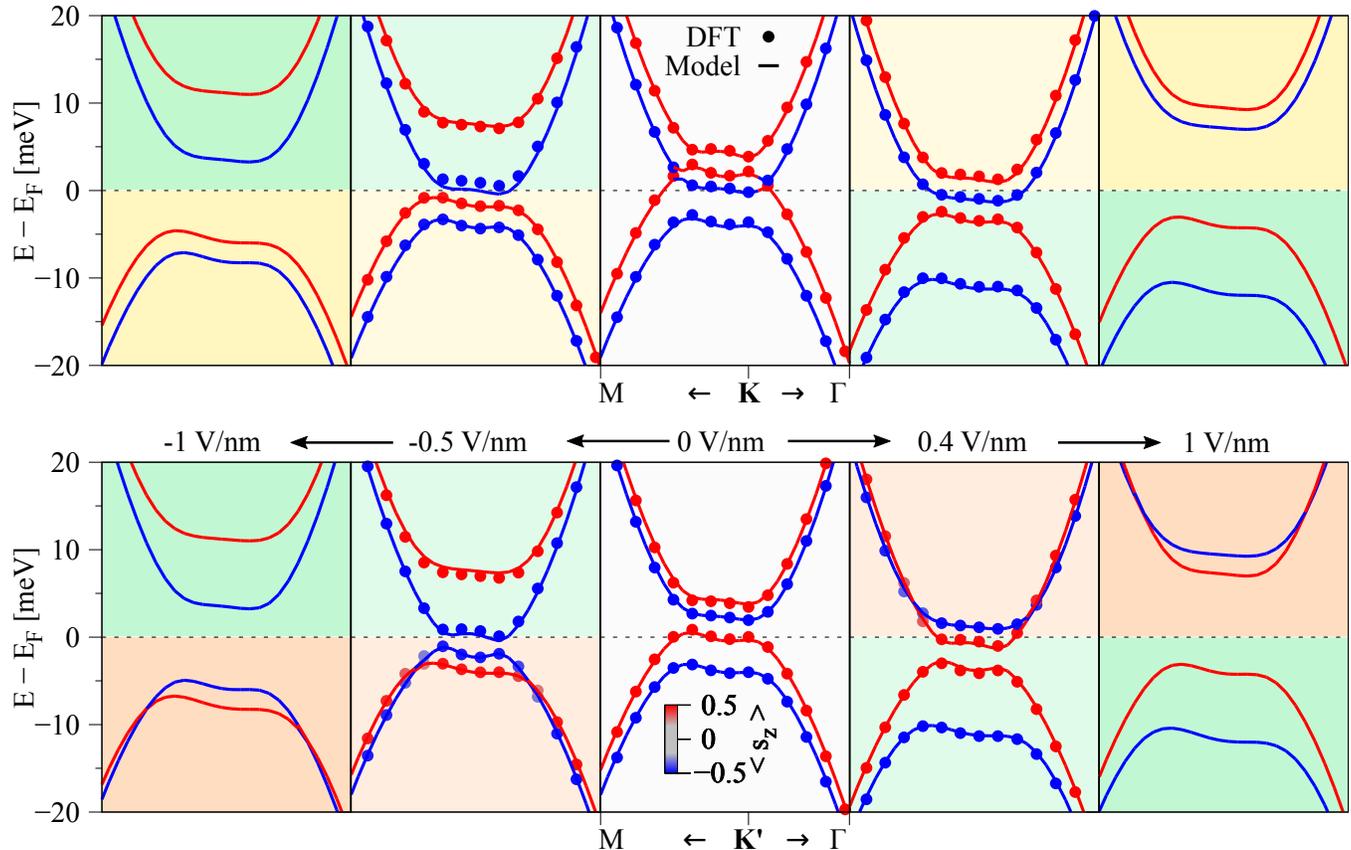


FIG. 3. Low energy dispersion curves around K (top) and K' (bottom) for the WS₂/BLG/CGT heterostructure. The color of the curves corresponds to the s_z spin expectation value: spin up is red and spin down is blue. The yellowish and reddish backgrounds indicate that the bands are split by spin-orbit coupling: the yellowish is for down-up (blue-red) and reddish for up-down (red-blue) spin ordering along increasing energy. The greenish background is for exchange coupling whose ordering is always up-down, fixed by the magnetization of the CGT layer. From left to right the transverse electric field is tuned from -1 to 1 V/nm. For electric fields of -0.5 , 0 , and 0.4 V/nm both model (solid lines) and DFT data (symbols) are plotted. For electric fields of ± 1 V/nm we use the model parameters with extrapolated values for V , assuming a linear dependence on the field. Parameters $V = 7.1, 3.2, -0.5, -3.6, -8.1$ meV correspond to the field values of $-1, -0.5, 0, 0.4, 1$ V/nm, respectively.

by the ferromagnetic CGT and conduction electrons will exhibit transport properties of magnetic conductors. On the other hand, the valence bands are split less, by about 2 meV. More important, the spin polarizations of the two bands are opposite at K and K' , signaling time-reversal symmetry: the valence bands experience spin-orbit coupling from WS₂. This makes sense. At negative electric fields electrons in the upper layer of BLG have a lower energy, and form the valence band. These electrons are affected by the TMDC layer, which gives them the strong spin-orbit coupling character. We will see in the next sec-

tion that the spin-orbit coupling is of the valley Zeeman type, which is characteristic for graphene proximitized by a TMDC. Similarly, electron orbitals in the lower layer of BLG have a higher energy, forming the conduction band, which is magnetic due to the presence of CGT.

If the applied electric field points up, towards the TMDC layer, the situation is reversed, see Fig. 3. While the orbital band gap develops again, the spin characters of the valence and conduction bands are swapped. The conduction bands are now split by spin-orbit coupling, and the valence bands by strong exchange cou-

pling. The reason is that the conduction bands are now formed by orbitals from the upper BLG layer which is close to TMDC, and the valence bands by orbitals from the lower BLG layer, close to the magnetic CGT.

We have thus demonstrated the swapping of exchange and spin-orbit couplings: at a fixed chemical potential (doping level), the investigated electronic system exhibits exchange coupling at one value of the electric field, and spin-orbit coupling at another. This gating effect controls the spin character of the states, and effectively turns time reversal symmetry on (spin-orbit coupling) or off (exchange). Such ex-so-tic structures offer on-demand exchange and spin-orbit coupling.

It is not essential for the swapping operation that the BLG is sandwiched between monolayers. Few-layer TMDCs and ferromagnets would work equally well. The short-rangeness of the proximity effect acts both ways. For example, the CGT monolayer induces exchange coupling to the adjacent (here lower) layer of BLG. The upper BLG layer is affected much less. Adding more layers of CGT will not matter. It will still be the interfacial layer which hybridizes with the carbon orbitals and induces the exchange in BLG. This picture was demonstrated explicitly by systematic DFT investigations in few-layer TMDC/CrI₃ slabs [38].

Model Hamiltonian. We now describe the low-energy band structure of the doubly proximitized BLG in the vicinity of K and K' valleys using an effective model that captures the essential physics. For this purpose we employ a Hamiltonian which was originally derived from symmetry arguments for AB stacked BLG in the presence of an external transverse electric field [37]. In our case the extrinsic effects are caused by the heterostructure: the upper BLG layer is proximitized by the TMDC, while the lower layer by the ferromagnetic CGT. Beyond these adjacent layers the proximity effect is too weak to markedly affect the band structure of BLG, which is also explicitly demonstrated by our study of individual TMDC/BLG and BLG/CGT bilayers (see Supplemental Material [36]).

The effective Hamiltonian reads

$$\mathcal{H} = \mathcal{H}_{\text{orb}} + \mathcal{H}_{\text{soc}} + \mathcal{H}_{\text{R}} + \mathcal{H}_{\text{ex}} + E_D. \quad (1)$$

It comprises orbital (orb), intrinsic spin-orbit (soc), Rashba (R), and exchange (ex) terms. We also include the Dirac-point energy E_D , which is a global energy shift to offset charge transfer in DFT results. In what follows we specify the individual Hamiltonian terms using pseudospin notation in which the physics is particularly transparent. A matrix form of \mathcal{H} is given in Supplemental Material [36].

The orbital physics is captured by

$$\begin{aligned} \mathcal{H}_{\text{orb}} = & -\frac{\sqrt{3}\gamma_0 a}{2} \mu_0 \otimes (\tau k_x \sigma_x + k_y \sigma_y) \otimes s_0 \\ & + \frac{\gamma_1}{2} (\mu_x \otimes \sigma_x - \mu_y \otimes \sigma_y) \otimes s_0 \\ & - \frac{\sqrt{3}\gamma_3 a}{4} \mu_x \otimes (\tau k_x \sigma_x - k_y \sigma_y) \otimes s_0 \\ & - \frac{\sqrt{3}\gamma_3 a}{4} \mu_y \otimes (\tau k_x \sigma_y + k_y \sigma_x) \otimes s_0 \\ & - \frac{\sqrt{3}\gamma_4 a}{2} (\tau k_x \mu_x - k_y \mu_y) \otimes \sigma_0 \otimes s_0 \\ & + V \mu_z \otimes \sigma_0 \otimes s_0 \\ & + \Delta (\mu_+ \otimes \sigma_+ + \mu_- \otimes \sigma_-) \otimes s_0, \end{aligned} \quad (2)$$

where we denote the graphene lattice constant a and the Cartesian components of the wave vector k_x and k_y measured from $\pm K$ for the valley index $\tau = \pm 1$. The Pauli matrices μ_i , σ_i , and s_i , represent layer, pseudospin, and spin, with $i = \{0, x, y, z\}$. We also define $\mu_{\pm} = \frac{1}{2}(\mu_z \pm \mu_0)$ and $\sigma_{\pm} = \frac{1}{2}(\sigma_z \pm \sigma_0)$ to shorten notation. Parameters γ_j , $j = \{0, 1, 3, 4\}$, describe intra- and interlayer hoppings of the BLG. To describe the effect of a transverse displacement field we introduce the voltage parameter V for the lower, and $-V$ for the upper layer of BLG. The parameter Δ describes the asymmetry in the energy shift of the bonding and antibonding states.

The proximity effects dramatically influence the spin states via spin-orbit coupling. The intrinsic spin-orbit coupling term, while also present in a free-standing BLG, is strongly renormalized by the proximity to the TMDC. This effect is described by

$$\begin{aligned} \mathcal{H}_{\text{soc}} = & \mu_+ \otimes \tau (\lambda_{\text{I}}^{\text{A}1} \sigma_+ + \lambda_{\text{I}}^{\text{B}1} \sigma_-) \otimes s_z \\ & - \mu_- \otimes \tau (\lambda_{\text{I}}^{\text{A}2} \sigma_+ + \lambda_{\text{I}}^{\text{B}2} \sigma_-) \otimes s_z, \end{aligned} \quad (3)$$

with parameters λ_{I} denoting the proximity spin-orbit coupling of the corresponding layer (1, 2) and sublattice (A, B) atom. Due to the short-rangeness of the proximity effect only the upper layer, so that only parameters $\lambda_{\text{I}}^{\text{A}2}$ and $\lambda_{\text{I}}^{\text{B}2}$ are significant (on the meV scale). The Rashba coupling can emerge due to the breaking of the space inversion symmetry in the heterostructure and the applied electric field. This term has the form

$$\mathcal{H}_{\text{R}} = \frac{1}{2} (\lambda_0 \mu_z + 2\lambda_{\text{R}} \mu_0) \otimes (\tau \sigma_x \otimes s_y - \sigma_y \otimes s_x), \quad (4)$$

where λ_0 describes the local (intrinsic) breaking of space inversion due to the presence of the other layer in BLG. The resulting spin-orbit fields are opposite in the two layers, giving no net effect on the spin-orbit splitting. The global breaking of space inversion due to the heterostructure and the electric field is accounted for by the proper Rashba parameter λ_{R} . For a more detailed description of the model and parameters, we refer the reader to Ref.

[37]. Finally, the magnetic proximity effect induces exchange coupling in BLG, which has the standard form,

$$\begin{aligned} \mathcal{H}_{\text{ex}} = & \mu_+ \otimes (-\lambda_{\text{ex}}^{\text{A1}} \sigma_+ + \lambda_{\text{ex}}^{\text{B1}} \sigma_-) \otimes s_z \\ & - \mu_- \otimes (-\lambda_{\text{ex}}^{\text{A2}} \sigma_+ + \lambda_{\text{ex}}^{\text{B2}} \sigma_-) \otimes s_z, \end{aligned} \quad (5)$$

where parameters λ_{ex} represent the proximity exchange for the individual sublattices and layers.

The spectrum of the effective Hamiltonian \mathcal{H} is fitted to the DFT-obtained low-energy dispersion of doubly proximitized BLG at zero applied electric field shown in Fig. 2. First, the orbital parameters γ_j from \mathcal{H}_{orb} are obtained, as well as the built-in bias V , staggered potential Δ , and the Dirac energy E_D . In the second step we analyze the fine structure of the spectrum and extract proximity induced spin-orbit and exchange parameters. The induced spin-orbit coupling in the upper layer is of the valley-Zeeman type, with opposite parameters on the two atoms of the sublattice: $\lambda_{\text{I}}^{\text{A2}} = -\lambda_{\text{I}}^{\text{B2}} \approx 1.1$ meV. The atoms of the lower BLG layer experience strong proximity exchange coupling due to the adjacent CGT, $\lambda_{\text{ex}}^{\text{A1}} = \lambda_{\text{ex}}^{\text{B1}} \approx -3.9$ meV. As for the Rashba coupling, it is negligible (on the meV scale), which is consistent with our finding that the spin polarization of the considered bands is predominantly out of plane. The fitted parameters are summarized in Tab. I; parameters not presented there were found to be negligible and set to zero. With such a minimal set of parameters the agreement of the effective model and the DFT is excellent, see Fig. 3.

The fitted parameters explicitly demonstrate the message that one layer of BLG experiences giant (on the meV scale) spin-orbit coupling, while the other layer giant exchange coupling, at zero electric field. What about the spectrum at finite electric fields? The only parameter that significantly changes when a field is applied is the voltage bias V between the two layers, as the other parameters are largely fixed by orbital hybridization. We therefore change V in our model Hamiltonian (and keep other parameters as fitted to the zero-field DFT results) to see how the spectra develop. The results are presented in Fig. 3. Their qualitative interpretation was already given above. Here we only note that the consistency of this procedure (changing only V as the electric field is applied) is checked by performing DFT simulations for two electric fields, -0.5 and 0.4 V/nm; for ± 1 V/nm we use V assuming its linear dependence on the applied electric field. The agreement with the model calculations, seen in Fig. 3, gives us full confidence in our approach. More detailed fit results can be found in the Supplemental Material [36], where we also provide results for the individual BLG/CGT and WS_2/BLG subsystems, to further validate the robustness of the model.

Summary. Doubly proximitized BLG offers a unique platform for investigating fundamental spin physics and designing multifunctional spintronics applications. Using realistic DFT simulations and phenomenological model-

ing we demonstrate swapping the two most important spin interactions—exchange and spin-orbit couplings—in a $\text{WS}_2/\text{BLG}/\text{CGT}$ multilayer. The swapping also means turning the time-reversal symmetry on and off, on demand, in the electronic states at a given doping. Since the effect is robust, we expect a variety of swapping phenomena if the bilayer sandwich comprises antiferromagnets, ferroelectricity, ferroelectrics, topological insulators, or superconductors.

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Supplemental Material:

Swapping exchange and spin-orbit coupling in ex-so-tic 2D heterostructures

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In the Supplemental Material, we show the geometries, band structures, and low energy bilayer graphene (BLG) bands fitted by the model Hamiltonian for the precursor heterostructures WS₂/BLG, and BLG/Cr₂Ge₂Te₆ (CGT). The results further validate the robustness of the model and accuracy of our fits. Furthermore, we show the exact geometry of the CGT/BLG/WS₂ heterostructure from the main text, as well as a more detailed view on the fitted low energy bands for zero and finite electric fields. We also explicitly write the Hamiltonian in matrix representation. In addition, we present details about our structural setup and the first-principles calculations.

GEOMETRY SETUP

For the considered heterostructure of the main text, we choose a 5×5 supercell of BLG in Bernal stacking, a $\sqrt{3} \times \sqrt{3}$ CGT supercell and a 4×4 supercell of WS₂. We stretch the lattice constant of graphene by roughly 2% to 2.5 Å [1] and stretch the lattice constant of CGT by roughly 6% to 7.2169 Å [2]. The WS₂ lattice constant is compressed by about 1% to 3.125 Å [3]. The supercell of WS₂/BLG/CGT has a lattice constant of 12.5 Å and contains 178 atoms in the unit cell. In Fig. S1 we show the exact geometry, akin to Fig. 1 of the main text, including relaxed interlayer distances of the CGT/BLG/WS₂ hybrid structure. The obtained interlayer distances are similar as reported in Refs. [4, 5]. The lower (upper) graphene layer, formed by sublattices A₁ and B₁ (A₂ and B₂), is proximitized by the CGT (WS₂) only, due to the short range proximity effect.

FIRST-PRINCIPLES CALCULATIONS

The electronic structure calculations and structural relaxation of the BLG-based heterostructures are performed by DFT [6] with QUANTUM ESPRESSO [7]. Self-consistent calculations are performed with the k -point sampling of $12 \times 12 \times 1$ to get converged results for the proximity exchange and spin-orbit coupling. For the CGT/BLG/WS₂ heterostructure, when spin-orbit coupling is included, a smaller k -point sampling of $6 \times 6 \times 1$ is used. We perform open shell calculations that provide the spin polarized ground state of CGT. A Hubbard parameter of $U = 1$ eV is used for Cr d -orbitals, being in the range of proposed values for CGT [5, 8]. We use an energy cutoff for charge density of 500 Ry, and the kinetic energy cutoff for wavefunctions is 60 Ry for the scalar relativistic pseudopotential with the projector augmented wave method [9] with the Perdew-Burke-Ernzerhof exchange correlation functional [10]. When SOC is included, we use the relativistic versions of the pseudopotentials. For the relaxation of the heterostructures, we add van der Waals corrections [11, 12] and use quasi-newton algorithm based on trust radius procedure. Dipole corrections [13] are also included to get correct band offsets and internal electric fields. In order to simulate quasi-2D systems, we add a vacuum of 20 Å, to avoid interactions between periodic images in our slab geometry. To determine the interlayer distances, the atoms of BLG and WS₂ are allowed to relax only in their z positions (vertical to the layers), and the atoms of CGT are allowed to move in all directions, until all components of all forces are reduced below 10^{-3} [Ry/ a_0], where a_0 is the Bohr radius.

HETEROSTRUCTURE OF BLG, CGT, AND WS₂

Here we present the Hamiltonian used to model the doubly proximitized BLG. The basis states are $|C_{A1}, \uparrow\rangle$, $|C_{A1}, \downarrow\rangle$, $|C_{B1}, \uparrow\rangle$, $|C_{B1}, \downarrow\rangle$, $|C_{A2}, \uparrow\rangle$, $|C_{A2}, \downarrow\rangle$, $|C_{B2}, \uparrow\rangle$, and $|C_{B2}, \downarrow\rangle$. In this basis the Hamiltonian from the main text is (see also Ref. 14)

$$\mathcal{H} = \mathcal{H}_{\text{orb}} + \mathcal{H}_{\text{soc}} + \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{R}} + E_D, \quad (\text{S1})$$

$$\mathcal{H}_{\text{orb}} = \begin{pmatrix} \Delta + V & \gamma_0 f(\mathbf{k}) & \gamma_4 f^*(\mathbf{k}) & \gamma_1 \\ \gamma_0 f^*(\mathbf{k}) & V & \gamma_3 f(\mathbf{k}) & \gamma_4 f^*(\mathbf{k}) \\ \gamma_4 f(\mathbf{k}) & \gamma_3 f^*(\mathbf{k}) & -V & \gamma_0 f(\mathbf{k}) \\ \gamma_1 & \gamma_4 f(\mathbf{k}) & \gamma_0 f^*(\mathbf{k}) & \Delta - V \end{pmatrix} \otimes s_0, \quad (\text{S2})$$

$$\mathcal{H}_{\text{soc}} + \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{R}} = \begin{pmatrix} (\tau\lambda_{\text{I}}^{\text{A1}} - \lambda_{\text{ex}}^{\text{A1}})s_z & i(\lambda_0 + 2\lambda_{\text{R}})s_-^{\tau} & 0 & 0 \\ -i(\lambda_0 + 2\lambda_{\text{R}})s_+^{\tau} & (-\tau\lambda_{\text{I}}^{\text{B1}} - \lambda_{\text{ex}}^{\text{B1}})s_z & 0 & 0 \\ 0 & 0 & (\tau\lambda_{\text{I}}^{\text{A2}} - \lambda_{\text{ex}}^{\text{A2}})s_z & -i(\lambda_0 - 2\lambda_{\text{R}})s_-^{\tau} \\ 0 & 0 & i(\lambda_0 - 2\lambda_{\text{R}})s_+^{\tau} & (-\tau\lambda_{\text{I}}^{\text{B2}} - \lambda_{\text{ex}}^{\text{B2}})s_z \end{pmatrix}. \quad (\text{S3})$$

We use the linearized version for the nearest-neighbor structural function $f(\mathbf{k}) = -\frac{\sqrt{3}a}{2}(\tau k_x - ik_y)$ and introduce also $s_{\pm}^{\tau} = \frac{1}{2}(s_x \pm i\tau s_y)$ for shorter notation.

TABLE S1. Fit parameters of the model Hamiltonian \mathcal{H} , Eq. (1) in the main text, for the BLG/WS₂ structure with SOC, for the CGT/BLG structure without SOC, and for the CGT/BLG/WS₂ structure without SOC. Unspecified parameters are zero.

system	BLG/WS ₂ (SOC)	CGT/BLG (no SOC)	CGT/BLG/WS ₂ (no SOC)
γ_0 [eV]	2.453	2.541	2.434
γ_1 [eV]	0.372	0.384	0.365
γ_3 [eV]	-0.270	-0.296	-0.269
γ_4 [eV]	-0.162	-0.179	-0.165
V [meV]	4.226	-8.499	-0.479
Δ [meV]	10.208	10.434	8.649
$\lambda_{\text{I}}^{\text{A2}}$ [meV]	1.070	0	0
$\lambda_{\text{I}}^{\text{B2}}$ [meV]	-1.179	0	0
$\lambda_{\text{ex}}^{\text{A1}}$ [meV]	0	-4.567	-4.257
$\lambda_{\text{ex}}^{\text{B1}}$ [meV]	0	-4.224	-3.997
E_D [meV]	-3.844	-1.003	-0.336
dipole [debye]	-1.089	1.527	0.366

In Fig. S1, we present a side view of the CGT/BLG/WS₂ heterostructure. The interlayer distance are of typical van der Waals type and similar as obtained in Refs. [4, 5]. In Tab. S1, we summarize the fit results for the CGT/BLG/WS₂ stack, when spin-orbit coupling (SOC) is not included in the calculation. The results are similar to the case with SOC, as listed in Tab. (1) in the main text, but SOC parameters are zero.

In Fig. S2 (Fig. S3), we show the calculated low energy bands with the model fit for zero (finite) electric field. Figs. S2 and S3 are more detailed views of the DFT-fitted bands, as shown in Fig. (3) from the main text. In all cases, the model agrees very well with the first-principles data and also band splittings are well reproduced. As described in the main text, we use the DFT-fitted parameters for zero electric field from Tab. (1) and tune V to match the finite electric field cases. An animation ([Efield_movie.mp4](#)) showing the evolution of the low energy bands of doubly proximitized BLG at K and K' , for a series of electric fields is also available.

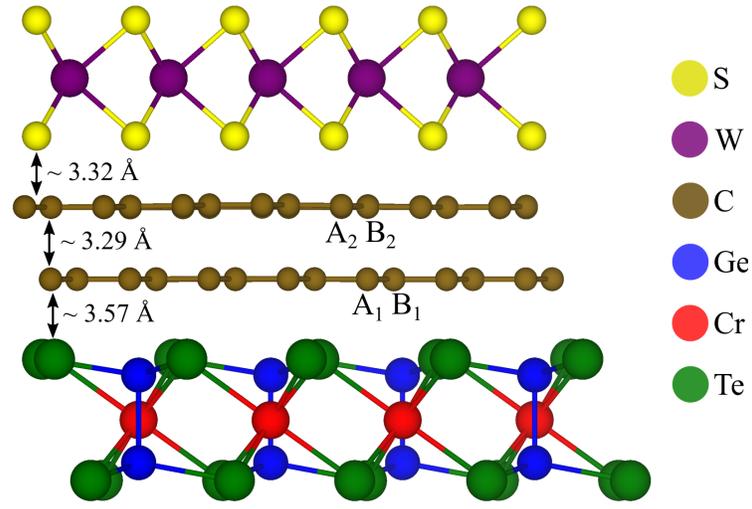


FIG. S1. Side view of the CGT/BLG/WS₂ heterostructure. Different colored spheres correspond to the different atomic species. The lower (upper) graphene layer of BLG is formed by sublattices A₁ and B₁ (A₂ and B₂). The relaxed interlayer distances are also indicated.

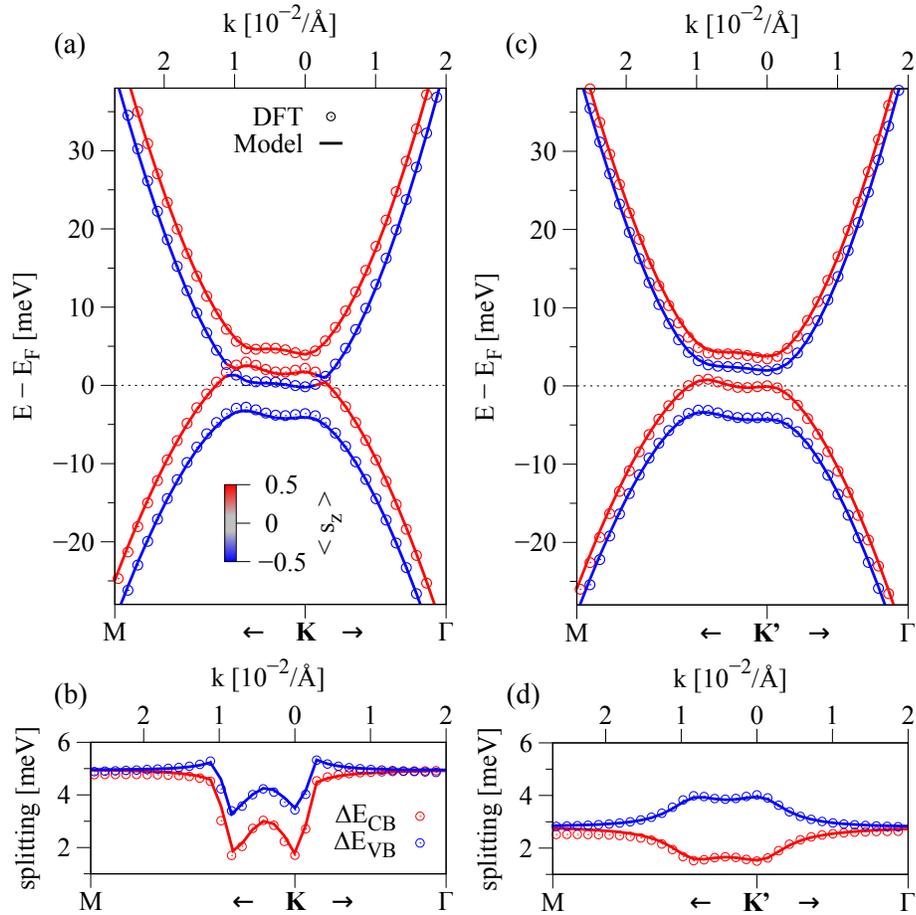


FIG. S2. (a) Zoom to the calculated low energy bands (symbols) around the K point with a fit to the model Hamiltonian (solid lines) for the WS₂/BLG/CGT heterostructure including SOC for zero electric field. (b) Energy splittings of the conduction and valence band. (c,d) Same as (a,b), but for the K' point.

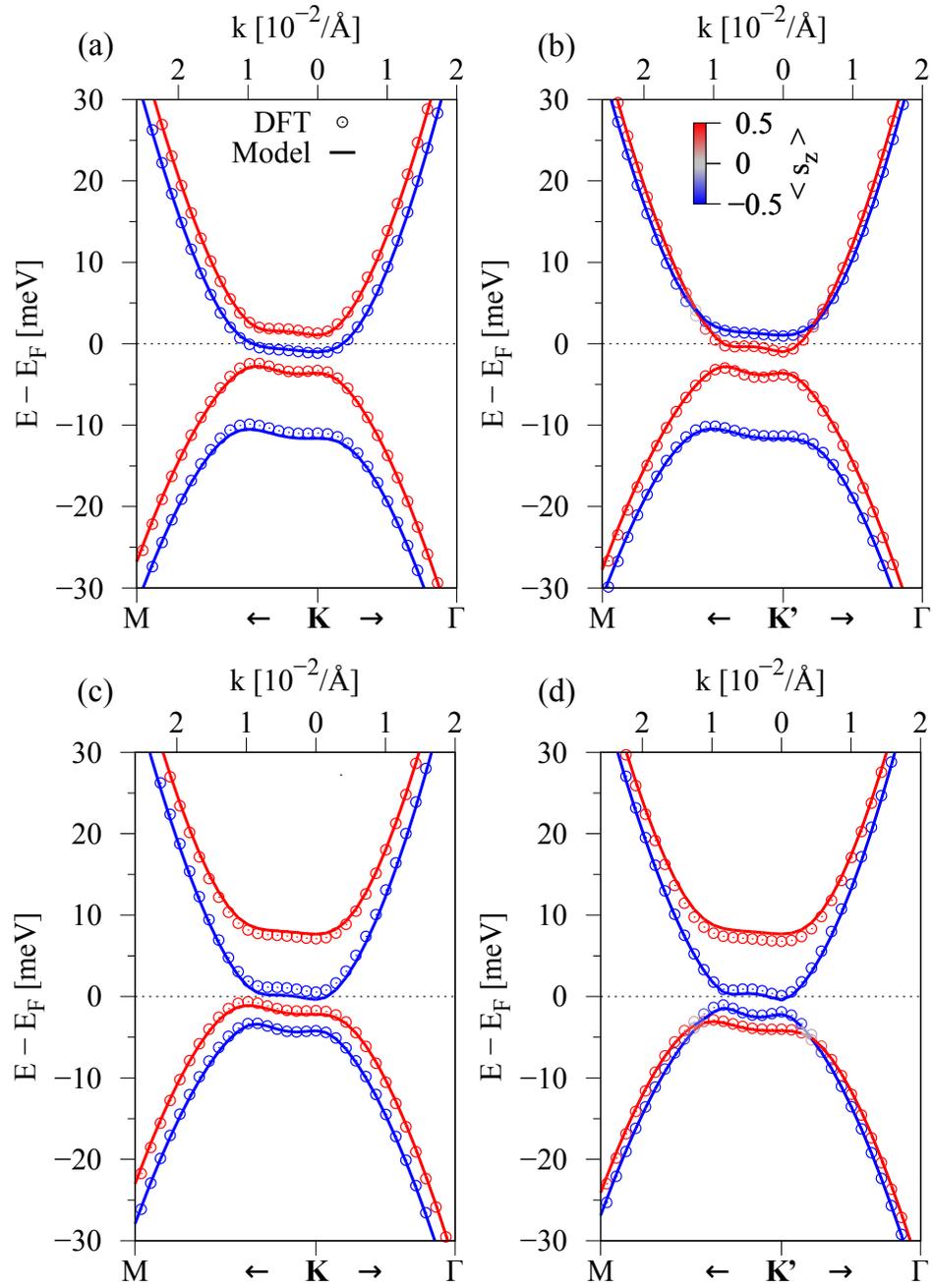


FIG. S3. Calculated (symbols) low energy band structures with a fit to the model Hamiltonian (solid lines), around K and K' points for $\text{WS}_2/\text{BLG}/\text{CGT}$ heterostructure including SOC. (a,b) For transverse electric field of $+0.4$ V/nm and a calculated dipole of 2.755 Debye. (c,d) For transverse electric field of -0.5 V/nm and a calculated dipole of -2.649 Debye.

HETEROSTRUCTURE OF BLG AND WS₂

For the WS₂/BLG heterostructure, we choose a 5×5 supercell of BLG in Bernal stacking and a 4×4 supercell of WS₂. We stretch the lattice constant of graphene by roughly 2% to 2.5 Å and the WS₂ lattice constant is compressed by about 1% from 3.153 Å [3] to 3.125 Å. The supercell of WS₂ on BLG has a lattice constant of 12.5 Å and contains 148 atoms in the unit cell.

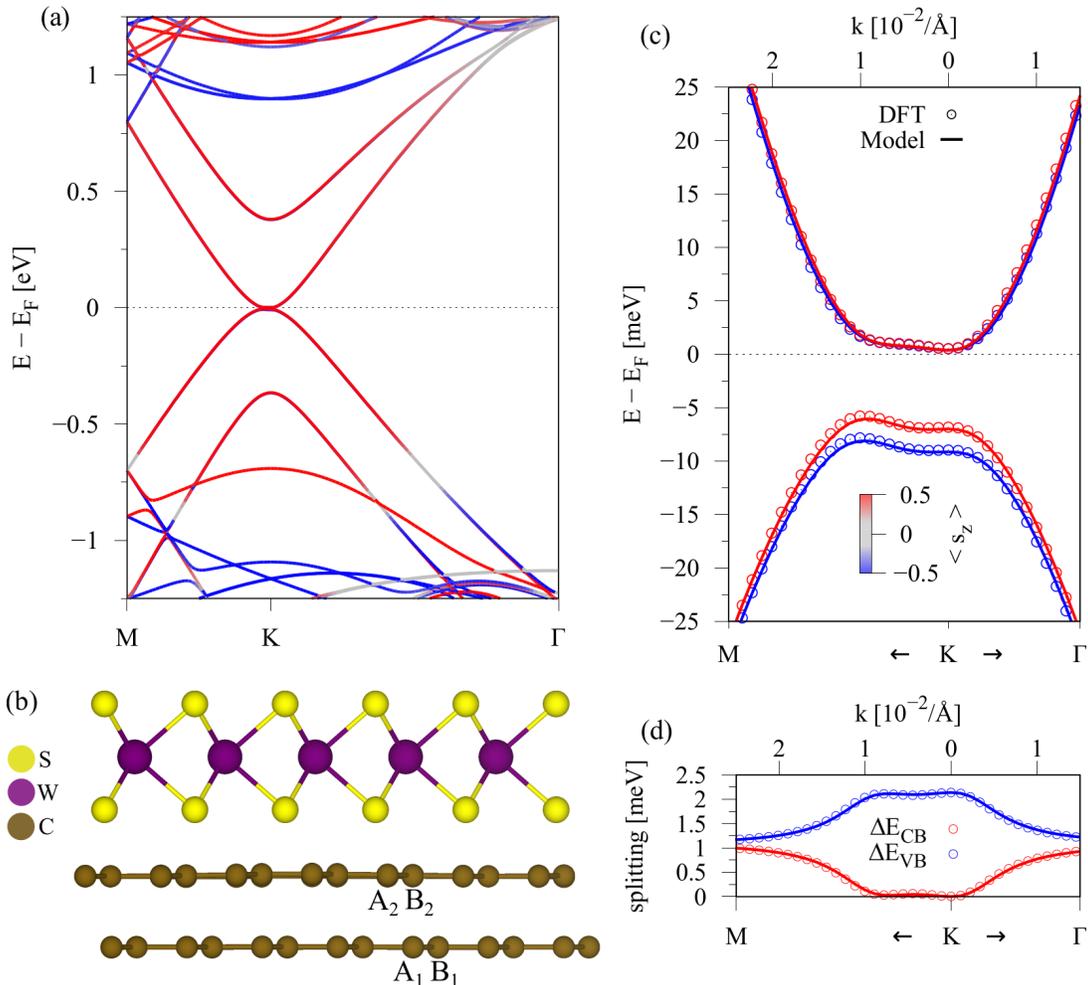


FIG. S4. (a,b) Band structure with SOC and geometry of WS₂ on BLG. (c) Zoom to the calculated low energy bands (symbols) around the *K* point with a fit to the model Hamiltonian (solid lines). The color corresponds to the s_z expectation value. (d) Energy splittings of the conduction and valence band.

Recent first-principles calculations have shown that for BLG on WS₂, there exists a spin-valve effect [4]. In Figs. S4(a,b) we show the band structure and a side view of the geometry of WS₂ on BLG. The results are in perfect agreement with Ref. [4]. We go one step further and fit our BLG low energy model Hamiltonian from the main text to the DFT calculated bands and find perfect agreement between the model and the ab-initio data, see Figs. S4(c,d). The fit parameters are summarized in Tab. S1 for the BLG/WS₂ heterostructure. The low energy valence band originates from the top graphene layer formed by sublattices A₂ and B₂. Because of the short range nature of proximity SOC, only this top layer is proximitized by the WS₂. Therefore the valence band experiences strong SOC induced spin splitting, while the conduction band splitting is much smaller near the *K* point.

Because of this short rangeness of proximity SOC and the unique BLG band structure, the splitting can be switched from the valence band to the conduction band by applying a transverse electric field across the heterostructure [4, 15], and spin relaxation of electrons and holes can be controlled fully electrically.

HETEROSTRUCTURE OF BLG AND CGT

For the BLG/CGT heterostructure, we choose a 5×5 supercell of BLG in Bernal stacking and a $\sqrt{3} \times \sqrt{3}$ CGT supercell. We keep the lattice constant of graphene unchanged at $a = 2.46 \text{ \AA}$ and stretch the CGT lattice constant by roughly 4% from 6.8275 \AA [2] to 7.1014 \AA . The supercell of BLG on CGT has a lattice constant of 12.3 \AA and contains 130 atoms in the unit cell.

Similar to before, in Figs. S5(a,b) we show the band structure and a side view of the geometry of BLG on CGT. In this case, the bottom graphene layer formed by sublattices A_1 and B_1 experiences strong proximity exchange from CGT. This system has recently been considered in Ref. [5], where an exchange valve effect has been found. Again, we fit our low energy model to the first-principles data and find perfect agreement, see Figs. S5(c,d). Here, the valence band, which originates from the bottom graphene layer formed by sublattices A_1 and B_1 , is strongly split due to proximity exchange. In contrast, the conduction band splitting is negligible, again due to short rangeness of the proximity effect. The fit parameters are also summarized in Tab. S1 for the CGT/BLG heterostructure.

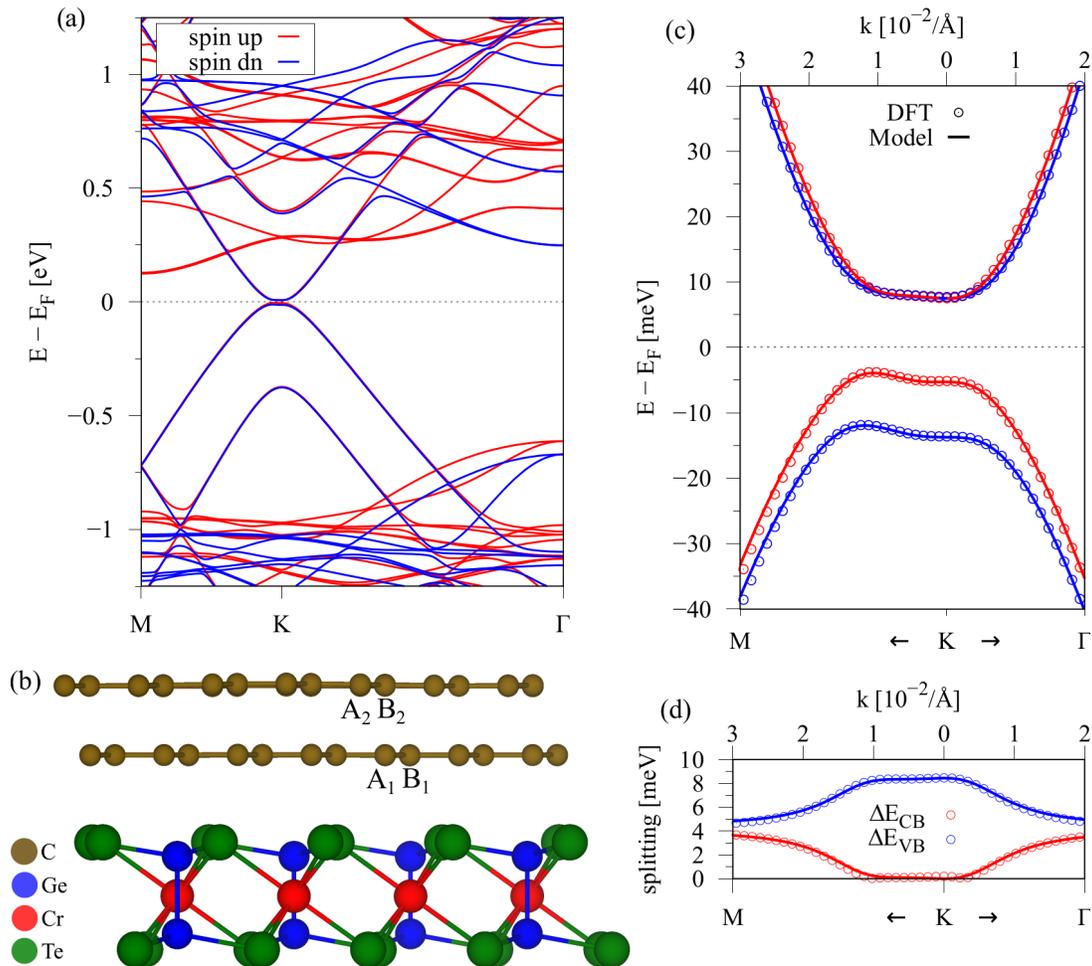


FIG. S5. (a,b) Band structure without SOC and geometry of BLG on CGT. (c) Zoom to the calculated low energy bands (symbols) around the K point with a fit to the model Hamiltonian (solid lines). Bands in red (blue) correspond to spin up (down). (d) Energy splittings of the conduction and valence band.

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