Proximity Spin–Orbit Torque on a Two-Dimensional Magnet within van der Waals Heterostructure: Current-Driven Antiferromagnet-to-Ferromagnet Reversible Nonequilibrium Phase Transition in Bilayer CrI₃

Kapildeb Dolui, Marko D. Petrović, Klaus Zollner, Petr Plecháč, Jaroslav Fabian, and Branislav K. Nikolić*

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ABSTRACT: The recently discovered two-dimensional magnetic insulator CrI₃ is an intriguing case for basic research and spintronic applications since it is a ferromagnet in the bulk but an antiferromagnet in bilayer form, with its magnetic ordering amenable to external manipulations. Using the first-principles quantum transport approach, we predict that injecting unpolarized charge current parallel to the interface of the bilayer-CrI₃/monolayer-TaSe₂ van der Waals (vdW) heterostructure will induce spin–orbit torque and thereby drive the dynamics of magnetization on the first monolayer of CrI₃ in direct contact with TaSe₂. By combining the calculated complex angular dependence of spin–orbit torque with the Landau-Lifshitz-Gilbert equation for classical dynamics of magnetization, we demonstrate that current pulses can switch the direction of magnetization on the first monolayer to become parallel to that of the second monolayer, thereby converting CrI₃ from antiferromagnet to ferromagnet while not requiring any external magnetic field. We explain the mechanism of this reversible current-driven nonequilibrium phase transition by showing that first monolayer of CrI₃ carries current due to evanescent wave functions injected by metallic transition metal dichalcogenide TaSe₂, while concurrently acquiring strong spin–orbit coupling via such a proximity effect, whereas the second monolayer of CrI₃ remains insulating. The transition can be detected by passing vertical read current through the vdW heterostructure, encapsulated by a bilayer of hexagonal boron nitride and sandwiched between graphite electrodes, where we find a tunneling magnetoresistance of ≃240%.

KEYWORDS: spin–orbit torque, 2D magnetic materials, van der Waals heterostructures, spintronics, first-principles quantum transport

INTRODUCTION

The recent discovery of two-dimensional (2D) magnets derived from layered van der Waals (vdW) materials¹,² has opened new avenues for basic research on low-dimensional magnetism³,⁴ and potential applications in spintronics⁵–⁹. Their magnetic phases can substantially differ from those in conventional bulk magnetic materials due to large structural anisotropy, which makes different signs and magnitudes of intralayer $J_{\text{intra}}$ and interlayer $J_{\text{inter}}$ exchange coupling between localized magnetic moments possible. For example, $J_{\text{intra}}$ and $J_{\text{inter}}$ are ferromagnetic and antiferromagnetic, respectively, between magnetic moments on Cr atoms within the bilayer of CrI₃, which eventually becomes an antiferromagnetic insulator with Néel temperature $T_N \approx 61$ K.² Hence, antiferromagnet spins have an opposite orientation in the two monolayers, whereas monolayer, trilayer, and bulk CrI₃ are ferromagnetic. Thus, the bilayer of CrI₃ can also be viewed as two monolayer ferromagnets that are antiferromagnetically

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coupled to each other. The monolayer CrI₃ circumvents the Mermin-Wagner theorem,¹⁰ where thermal fluctuations destroy long-range magnetic order in 2D, by exhibiting strong uniaxial perpendicular magnetic anisotropy (PMA) that removes rotational invariance and effectively makes it a realization of the Ising model.³ The PMA is also required for high-density device applications.

The layer and stacking order¹¹,¹² dependence of electronic and spin structure as a new knob, together with possibilities for external manipulation via gating, straining, and coupling to other 2D materials within vdW heterostructures, allows for dramatic changes of magnetic ordering of 2D magnets, which is not possible with conventional bulk magnetic materials. For example, very recent experiments¹³−¹⁵ have demonstrated antiferromagnet (AFM) to ferromagnet (FM) phase transition in the bilayer of CrI₃ by applying an external electric field via gate voltage or by electrostatic doping. While these effects offer potential building blocks¹⁶ for an ultralow-dissipation nonvolatile memory, at present, they employ cumbersome external magnetic fields that cannot be generated on the nanoscale as required for integration with other elements of a circuit. Furthermore, to read the change of magnetic state of such a device within a circuit, it is still required to pass a current through it,¹⁷ as demonstrated recently using unconventional magnetic field coupling to the proximity effect in graphene) with current carrier (see the Supporting Information) up to the first sight not suitable for SOT-operated devices.

Here, we employ first-principle quantum transport framework, which combines⁴⁻⁶ nonequilibrium Green functions (NEGFs)⁷ for two-terminal devices with noncollinear density functional theory (ncDFT) calculations,⁸,⁹ to predict that the AFM–FM nonequilibrium phase transition can be induced by SOT in the bilayer-CrI₃/monolayer-TaSe₂ vdW lateral heterostructure depicted in Figure 1 where unpolarized charge current is injected parallel to the interface. The monolayer of TaSe₂ is chosen in 1H phase for which lattice mismatch between TaSe₂ and CrI₃ is as small as 0.1%, while inversion symmetry of TaSe₂ is broken to create large spin−orbit coupling (SOC).

Both conventional spin-transfer torque (in the absence of SOC and in geometries with two FM layers with noncollinear magnetizations⁴⁻⁶,³⁰) and SOT (in geometries with one FM layer but requiring interfacial or bulk SOC effects⁴⁻⁶,³¹,³²) can be described microscopically and independently of a particular physical mechanism,²²,²⁶ as a consequence of the interaction between current-driven (CD) nonequilibrium spin density⁵³⁻⁵⁵ of conduction electrons S_{CD}(x) and a nonzero exchange-correlation (XC) magnetic field B_{XC}(x)²⁶,²⁹ present in equilibrium. Their cross product, S_{CD}(x) × B_{XC}(x), is local torque at some point in space x, so that the total torque is obtained by integration⁴⁻⁶,³⁰

\[
T = \int d^3r S_{CD}(r) \times B_{XC}(r)
\]

While B_{XC}(x) is nonzero in both the monolayer and bilayer of CrI₃ due to long-range magnetic ordering, S_{CD}(x) appears only on the monolayer of CrI₃ that is in direct contact with the monolayer of TaSe₂, as demonstrated by Figure 2. This is due to the proximity effect where evanescent wave functions from TaSe₂ penetrate into it by TaSe₂. The unit vectors of magnetizations on the two CrI₃ monolayers are denoted by m₁ and m₂. The heterostructure is assumed to be infinite in the xy-plane.

![Figure 1. Schematic view of the CrI₃-TaSe₂ vdW heterostructure consisting of an insulating antiferromagnetic bilayer of CrI₃ and a nonmagnetic metallic monolayer TMD TaSe₂. The unpolarized charge current is injected parallel to the interface by a small applied bias voltage V_b between the left and right macroscopic reservoirs. The current flows through the monolayer of TaSe₂ as well as through the first monolayer (see the Supporting Information) of CrI₃ due to evanescent wave functions injected into it by TaSe₂. The unit vectors of magnetizations on the two CrI₃ monolayers are denoted by m₁ and m₂. The heterostructure is assumed to be infinite in the xy-plane.](https://dx.doi.org/10.1021/acs.nanolett.9b04556)

![Figure 2. Current-driven nonequilibrium spin density S_{CD} = (S_{CD}S_{CD}S_{CD}) in the linear-response regime within a bilayer-CrI₃/monolayer-TaSe₂ vdW heterostructure for (a) m₃ = |m₃|; (b) m₅ = |m₅|; (c) m₇ = |m₇|. Vertical dashed lines indicate the position of each atomic plane. The area of the common rectangular supercell of the vdW heterostructure in Figure 1 is denoted by |= 2√3 a², where a = 6.85 Å is the lattice constant of bulk CrI₃. Shaded green areas represent rescaled S_{CD} in the spatial region of the first monolayer of CrI₃, which is in direct contact with the monolayer of TaSe₂.](https://dx.doi.org/10.1021/acs.nanolett.9b04556)
profiles of local charge current density), while also bringing\(^{36}\) SOC from TaSe\(_2\) to ensure that \(S_{CD}(r)\) is not collinear to \(B_X(r)\). The giant SOC hosted by TaSe\(_2\) itself due to inversion symmetry breaking in ultrathin layers of transition metal dichalcogenides (TMDs)\(^{37,38}\) is confirmed by large \(S_{CD}(r)\) within the spatial region of the TaSe\(_2\) monolayer in Figure 2.

The SOT vector in eq 1, with its complex angular dependence [Figure 4] on the direction of magnetization (along the unit vector \(m_1\) in Figure 1) of the first monolayer of CrI\(_3\), is combined in a multiscale fashion\(^{30,39}\) with the classical dynamics of magnetization governed by the Landau-Lifshitz-Gilbert (LLG) equation to demonstrate reversible switching of \(m_1\) [Figure 5b and the accompanying movie in the Supporting Information] from the \(-z\) to \(+z\) direction by current pulses and, thereby, transition from the AFM to FM phase of the CrI\(_3\) bilayer. The dynamics of \(m_1\) can be detected by passing the vertical read current along the \(z\)-axis,\(^{17}\) where we compute the tunneling magnetoresistance (TMR) of \(\approx 240\%\) [Figure 6] due to the AFM–FM transition of the CrI\(_3\) bilayer. For such a scheme, we assume that the bilayer-CrI\(_3\)/monolayer-TaSe\(_2\) vdW heterostructure is sandwiched between two metallic semi-infinite graphite electrodes along the \(z\)-axis with hexagonal BN (hBN) bilayers inserted between the leads and vdW heterostructure [inset of Figure 6].

### METHODOLOGY

We employ the interface builder in the QuantumATK\(^{40}\) package to construct a unit cell of the vdW heterostructure in Figure 1 while starting from experimental lattice constants of...
The heterostructure in Figure 1 is split into the central region and left (L) and right (R) semi-infinite leads, all of which are composed of the same CrI3/TaSe2 trilayer. The self-energies of the leads $\Sigma_{L,R}(E)$ and the Hamiltonian $H_{KS}$ of the central region are obtained from ncDFT calculations within the QuantumATK package using PBE parametrization of GGA for the XC functional; norm-conserving fully relativistic pseudopotentials of the type SG15-SO40,48 for describing electron–core interactions; the SG15 (medium) numerical linear combination of atomic orbitals (LCAO) basis set.38 Periodic boundary conditions are employed in the plane perpendicular to the transport direction with grids of $1 \times 101$ k-point (lateral device setup in Figure 1 for SOT calculations) and $25 \times 25$ k-point (vertical device setup in the inset of Figure 6 for TMR calculations). The energy mesh cutoff for the real-space grid is chosen as 100 hartree.

The lesser Green function (GF), $G^-(E) = \Sigma G(E) - i\delta_{E,E_f}G(E)$, of NEGF formalism makes it possible to construct the nonequilibrium density matrix

$$
\rho_{eq} = \frac{1}{2\pi i} \int_{-\infty}^{\infty} dE G_{\uparrow}(E) G_{\uparrow}(E)
$$

in the steady state and elastic transport regime. Here, $G = [E\Lambda - H_{KS} - \Sigma_{\downarrow}(E,V_L) - \Sigma_{\uparrow}(E,V_R)]^{-1}$ is the retarded GF; $\Gamma(E) = \sum_{\downarrow}(E,V_L) - \Sigma_{\downarrow}(E,V_R)$ is the shifted Fermi function of the macroscopic reservoirs into which semi-infinite leads terminate; $V_L = V_\downarrow - V_\uparrow$ is the applied bias voltage between them; $\Gamma_{\uparrow\downarrow}(E) = \left[\Sigma_{\uparrow}(E) - \Sigma_{\downarrow}(E)\right]$ is the level broadening matrix. For the lateral heterostructure [Figure 1], all matrices, $H_{KS}$, $G$, $\Gamma_{\uparrow\downarrow}$, and $\rho_{eq}$ depend on $k_y$ while for vertical the heterostructure [inset of Figure 6], they depend on $(k_x,k_z)$. Due to the nonorthogonality of the LCAO basis set $\hat{\phi}_i$, we also use the overlap matrix $A$ composed of elements $\langle \phi_i|\phi_j \rangle$.

The CD part of the nonequilibrium density matrix $\mu_{CD}(k_x,k_y,k_z)$ is obtained by subtracting the equilibrium density matrix $\rho_{eq}(k_x,k_y,k_z)$ for $V_L = V_R$. This yields

$$
S_{CD}(k_x,k_y,k_z) = Tr[\rho_{CD}(k_x,k_y,k_z) \sigma A^{-1}]
$$

and SOT

$$
T = \frac{1}{\Omega_B} \int_{BZ} dk_x S_{CD}(k_x,k_y,k_z) \times B_{XC}(k_y,k_z)
$$

which we compute by performing trace in the LCAO basis [instead of in real space as in eq 1], and additional integration over the one-dimensional Brillouin zone (BZ) of length $\Omega_B$ is performed.

**NONEQUILIBRIUM SPIN DENSITY**

The xy-plane averaged $S_{CD}$ is plotted in Figure 2 for three representative orientations of the magnetization $\mathbf{m}_i$ on the first monolayer of CrI3. The nonequilibrium spin

\[ \text{CrI}_3/\text{monolayer-TaSe}_2 \text{vdW heterostructure.} \]
density is zero on the second monolayer of CrI$_3$ which confirms that evanescent wave functions originating from the metallic TaSe$_2$ monolayer and the spin–orbit proximity effect carried by them decay exponentially fast, so they are able to reach only the first monolayer of CrI$_3$. Independently of the orientation of $\mathbf{m}_1$, the magnitude of $S_{\text{CD}}$ within the first monolayer of CrI$_3$ is mainly dominated by its y-component, which is an order of magnitude smaller than $S_{\text{CD}}$ within TaSe$_2$. Concurrently, the magnetic proximity effect from CrI$_3$ induces a small magnetization into the monolayer of TaSe$_2$ with magnetic moments on Ta and Se atoms being 0.008 and 0.001 $\mu_B$, respectively, where $\mu_B$ is the Bohr magneton. In comparison, we compute magnetic moments on Cr and I atoms as $\mu_{\text{Cr}} = 3.43$ $\mu_B$ and $\mu_{\text{I}} = 0.14$ $\mu_B$ respectively. The component $S_{\text{CD}}$ within the TaSe$_2$ monolayer is insensitive to $\mathbf{m}_1$, while $S_{\text{CD}}$ remains negligible.

Unlike the surface of the topological insulator or heavy metals where $S_{\text{CD}}$ is confined to the plane in accord with the phenomenology of the standard inverse spin-galvanic (or Edelstein) effect, TaSe$_2$ exhibits large out-of-plane component $S_{\text{CD}}$, which is sensitive to the orientation of $\mathbf{m}_1$ and is highly sought for SOT-operated device applications. This can be traced to the large $S_{\text{CD}}$ component of the spin texture in k-space for an isolated monolayer TaSe$_2$, which remains large when TaSe$_2$ is covered by the bilayer CrI$_3$ while also acquiring nontrivial in-plane components $S_{\text{eq}}$ and $S'_{\text{eq}}$ (see the Supporting Information for more details).

### ANGULAR DEPENDENCE OF SOT

The SOT vector can be decomposed into odd ($o$) and even ($e$) components with respect to the orientation of $\mathbf{m}_1$, so we find $\mathbf{p}_o$ ($\theta = 88^\circ, \phi = 98^\circ$) instead of often naively assumed $\mathbf{p}$ ($\theta = 90^\circ, \phi = 90^\circ$) in eq 6 is conventional field-like torque, while higher terms can have properties of both field-like and damping-like torque [the lowest order term $\mathbf{r}^\alpha(p \times \mathbf{m}_1)$ in the expansion of $T^o$ is conventional damping-like torque]. The value of $\mathbf{r}^\alpha_o$ for the SOT driven classical dynamics of magnetization, equation for more details).

#### SOT-DRIVEN CLASSICAL DYNAMICS OF MAGNETIZATION

The effective anisotropic classical Heisenberg model for magnetic moments $\mathbf{m}_1$ and $\mathbf{m}_2$ on Cr atoms within two monolayers of CrI$_3$ in Figure 1 is given by

$$
\mathcal{H} = -J_{12}(\mathbf{m}_1 \cdot \mathbf{m}_2) - \sum_{i=1,2} A_i (m_i^z)^2
$$

where the values for $J_{12} = -0.05$ meV, as the interlayer AFM exchange coupling, and $A_1 = 1.5$ meV, as the PMA constant in the presence of TaSe$_2$ monolayer, are extracted from ncDFT calculations. They are close to the corresponding values obtained for the isolated CrI$_3$ bilayer in previous ncDFT calculations. We simulate the classical dynamics $\mathbf{m}_1(t)$ by solving the LLG equation

$$
\frac{d\mathbf{m}_1}{dt} = -\gamma \mathbf{m}_1 \times \mathbf{B}_{\text{eff}} + \lambda_1 \mathbf{m}_1 \times \frac{d\mathbf{m}_1}{dt} + \frac{\gamma T^o}{\mu_0}
$$

assuming that current flows along the z-axis as in Figure 1. Note that other expansions, such as in terms of orthonormal vector spherical harmonics, can also be employed to define the fitting function. Here, $\mathbf{r}^\alpha_{o,n}$ are the fitting parameters, and $p$ is the unit vector along the reference direction set by current-induced nonequilibrium spin density, such that $T^o = 0$ when $\mathbf{m}_1 \parallel p$. In simple systems, like the Rashba spin-split 2D electron gas or metallic surface of the topological insulator in contact with the FM layer, $p \parallel y$ (assuming injected current along $\hat{x}$) is determined by symmetry arguments. However, for more complicated systems, it has to be calculated, and we find $p \equiv (\theta = 88^\circ, \phi = 98^\circ)$ instead of often naively assumed $p \equiv (\theta = 90^\circ, \phi = 90^\circ)$.
\( \mathbf{m}_1 \parallel \mathbf{z} \) to \( \mathbf{m}_2 \parallel \mathbf{z} \) while magnetization of the second layer remains \( \mathbf{m}_2 \parallel \mathbf{z} \). Such current-induced FM phase is stable in-between two pulses, on the proviso that \( \lambda_1 > |J_z| \) in eq 7, and can be reversed back to the AFM phase by the next pulse [Figure 5b and movie in the Supporting Information]. We assume different Gilbert damping parameters \( \lambda_1 = 0.01 > \lambda_2 = 0.0001 \) on two monolayers of CrI\(_3\) due to the presence of the TaSe\(_2\) monolayer, but the actual value \( \lambda_1 \) on the first monolayer of CrI\(_3\) is likely smaller. Thus, we anticipate that the time needed to stabilize the FM phase would be on the order of ~1 ns instead of ~100 ps in Figure 5 (where \( \lambda_1 \) was tuned for such numerical convenience).

Since \( T^0 \) forces precession of magnetization around the axis defined by \( \mathbf{p} \), magnetization reversal in the voltage pulse setup is achieved by fine-tuning the pulse duration \( \Delta t_{\text{ON}} \) to half of the period of that precession. The Gilbert damping term, \( \lambda_1 \mathbf{m}_1 \times \mathbf{d}\mathbf{m}_1 / dt \), does not play a role in this type of switching, although \( \lambda_1 \) together with the PMA constant is critical to stabilize the FM phase after the pulse is switched off, as shown in the movie in the Supporting Information. For instance, when Gilbert damping is set to zero, the magnetizations in both monolayers never fully align with the \( \mathbf{z} \)-axis and instead continue to precess around it which renders the FM phase unstable. Note that more detailed LLG simulations would be required to simulate more than two magnetic moments and their inhomogeneous switching in a particular device geometry, as often observed experimentally in ferromagnet/heavy-metal heterostructures,\(^{56}\) but our two-terminal device is homogeneous and translationally invariant within the \( xy \)-plane in Figure 1. Also, in the presence of disorder and thereby induced voltage drop across the central region,\(^{25,26,53}\) we expect that \( T^0 \) would become nonzero\(^{54}\) and contribute to switching.

\section*{TMR AS A PROBE OF AFM–FM TRANSITION}

Finally, in analogy with the experiments\(^{17}\) where SOT-driven magnetization switching has been probed by passing additional vertical read current through SOT devices operated by lateral current, we investigate the angular dependence of TMR for vertical current assumed to be injected between semi-infinite graphite leads along the \( z \)-axis sandwiching bilayer-CrI\(_3\)/monolayer-TaSe\(_2\) [see inset in Figure 6 for illustration]. We define angular dependence of TMR as \( TMR(\theta) = |R(\theta)| - R(0)/R(0) \), where \( R(0) \) is the resistance of the FM phase with \( \mathbf{m}_1 \parallel \mathbf{z} \) and \( R(\theta) \) is the resistance for angle \( \theta \) between them.

We compute \( R(\theta) = -\int \frac{\mathbf{E}_p}{\mathbf{E}_0} \mathbf{m}_1 \phi \left( \mathbf{E}_p, \theta \right) \mathbf{m}_2 \phi \left( \mathbf{E}_p, \theta \right) \int_{\text{BZ}} T(\mathbf{E}_p, \theta) \int_{\text{BZ}} T(\mathbf{E}_p, \theta) \int_{\text{BZ}} T(\mathbf{E}_p, \theta) \). This integration is done over the forward transfer function of the BZ, \( T(\mathbf{E}_p, \theta) = \frac{1}{T_{\text{total}}} \int_{\text{BZ}} dk_x T(\mathbf{E}_p, \theta; k_x) \), akin to eq 5. Thus, \( R(\theta = 180^\circ) \) corresponds to the AFM phase. Note that TMR(\( \theta = 180^\circ \)) recovers the conventional definition of TMR using only parallel and antiparallel configurations of the magnetizations. In Figure 6, we obtain TMR(\( \theta = 180^\circ \)) \( \approx 240\% \) when using additional hBN bilayers inserted between graphite leads and the vdW heterostructure. When hBN is removed, TMR drops to TMR(\( \theta = 180^\circ \)) \( \approx 40\% \), while exhibiting a peculiar change of sign for the angles between \( \theta = 0^\circ \) and \( \theta = 180^\circ \) in accord with the experimental observation reported in ref 18 of few-layer-graphene/bilayer-CrI\(_3\)/few-layer-graphene junctions.

\section*{ASSOCIATED CONTENT}

\section*{Supporting Information}

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.9b04556.

Movie, accompanying Figure 5a,b, animates the time evolution of magnetizations, \( \mathbf{m}_1(t) \) and \( \mathbf{m}_2(t) \) in Figure 1, driven by a sequence of rectangular voltage pulses (MP4).

Additional details for Figure 2, such as the spatial profile of local current density on different monolayers of the bilayer-CrI\(_3\)/monolayer-TaSe\(_2\) vdw heterostructure, as well as for Figure 3, such as spin textures in the bilayer-CrI\(_3\)/monolayer-TaSe\(_2\) vdw heterostructure vs spin textures in an isolated monolayer-TaSe\(_2\) (PDF).

\section*{AUTHOR INFORMATION}

\section*{Corresponding Author}

Branislav K. Nikolić — Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, United States; orcid.org/0000-0002-5793-7764; Email: bnikolic@udel.edu

\section*{Authors}

Kapildeb Dolui — Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, United States

Marko D. Petrović — Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, United States

Klaus Zollner — Institute for Theoretical Physics, University of Regensburg, Regensburg 93040, Germany

Petr Plecháč — Department of Mathematical Sciences, University of Delaware, Newark, Delaware 19716, United States

Jaroslav Fabian — Institute for Theoretical Physics, University of Regensburg, Regensburg 93040, Germany

Complete contact information is available at: https://pubs.acs.org/doi/10.1021/acs.nanolett.9b04556

\section*{Notes}

The authors declare no competing financial interest.

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Supporting Information for “Proximity Spin-Orbit Torque on a Two-Dimensional Magnet within van der Waals Heterostructure: Current-Driven Antiferromagnet-to-Ferromagnet Reversible Nonequilibrium Phase Transition in Bilayer CrI₃”

Kapildeb Dolui,¹ Marko D. Petrović,¹,² Klaus Zollner,³ Petr Plecháč,² Jaroslav Fabian,³ and Branislav K. Nikolić¹,∗

¹Department of Physics and Astronomy, University of Delaware, Newark, DE 19716, USA
²Department of Mathematical Sciences, University of Delaware, Newark, DE 19716, USA
³Institute for Theoretical Physics, University of Regensburg, Regensburg 93040, Germany

I. ADDITIONAL INFORMATION FOR FIGURE 2 IN THE MAIN TEXT

The real-space nonequilibrium spin density within different layers of bilayer-CrI₃/monolayer-TaSe₂ van der Waals (vdW) heterostructure shown in Figure 2 in the main text demonstrates that evanescent wavefunction originating from the monolayer TaSe₂ reach only the first monolayer of CrI₃. This is further clarified by spatial profile of local charge current density [Figure S1] which we extract from nonequilibrium Green function (NEGF) combined with noncollinear density functional theory (ncDFT) calculations. As shown in Figure S1, injected unpolarized charge current flows only through the monolayer TaSe₂ and (a much smaller fraction of it) through the first monolayer of CrI₃ which is in direct contact with the monolayer TaSe₂.

![Figure S1](image)

**Figure S1.** The NEGF+ncDFT-computed spatial profile of local charge current density flowing parallel to the interface of bilayer-CrI₃/monolayer-TaSe₂ vdW heterostructure from Figure 1 in the main text.

* bnikolic@udel.edu
II. ADDITIONAL INFORMATION FOR FIGURE 3 IN THE MAIN TEXT

The electronic and spin structure from Figure 3(a),(b) in the main text for bilayer-CrI$_3$/monolayer-TaSe$_2$ vdW heterostructure, computed by ncDFT+U methodology using $U = 2.0$ eV, is replotted in Figure S2 with additional details of spin structure along different bands. That is, the colored segments within each band signify three components of the spin expectation value. We also mark (open circles) band projections onto the three different monolayers. Figure S2(d) highlights four bands near the Fermi level, b$_1$–b$_4$, that originate from the monolayer TaSe$_2$. For those bands, Figure S3 plots the corresponding *spin textures* in $k$-space as the equilibrium expectation value

$$
(S_{\text{eq}}^x, S_{\text{eq}}^y, S_{\text{eq}}^z) = \langle \Psi_k | \mathbf{\sigma}/2 | \Psi_k \rangle,
$$

(1)

of the spin operator in the eigenstates $|\Psi_k\rangle$ of ncDFT Hamiltonian, where $\mathbf{\sigma} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ is the vector of the Pauli matrices. We also denote their contribution to the Fermi surface. For example, in Figure S3(c) the green colored regions indicate the contribution of band b$_3$ to the Fermi surface, i.e., when $E_{b_3} - E_F$ is within the energy window $[-0.03, 0.03]$ eV. The arrows represent the in-plane $S_{\text{eq}}^x$ and $S_{\text{eq}}^y$ components, while the red/blue colored regions show the $S_{\text{eq}}^z$ component.

Although band structure in Figure S2 near the Fermi level $E_F$ resembles that of an isolated monolayer TaSe$_2$ shown in Figure S4, due to hybridization with states from the first monolayer of CrI$_3$ band anticrossings emerge in Figure S2. Furthermore, bands originating from the monolayer TaSe$_2$ acquire in-plane spin components $S_{\text{eq}}^i$ ($i = x, y$) and $S_{\text{eq}}^z$. In contrast, they are too small to be visualized in Figure S5 for an isolated monolayer TaSe$_2$. Overall, the threefold symmetry of the heterostructure is also reflected in the spin textures. Especially the hexagonal shaped green region in Figure S3(c) around the $\Gamma$ point looks quite interesting since it exhibits a nontrivial in-plane contribution to the spin textures. The contribution of band b$_3$ to the Fermi surface, shown in Figure S3(b), is the largest of all bands and the corresponding spin texture is even more complicated. Bands b$_1$ and b$_4$ are giving only minor contributions to the Fermi surface.

The relevant scales of the in-plane and out-of-plane components of spin textures are unraveled by taking the spin expectation values for the bands b$_1$–b$_4$ and by averaging their absolute values within the Brillouin zone (BZ):

$$
\overline{S_{\text{eq}}^i} = \frac{1}{N} \sum_{k \in \text{BZ}} |S_{\text{eq}}^{i,\alpha}(k)|,
$$

(2)

where $i = \{x, y, z\}$, $\alpha = \{b_1, b_2, b_3, b_4\}$ and $N$ is the number of $k$-points that we use to sample the BZ. Table S1 summarizes the averaged spin expectation values, where we find that $\overline{S_{\text{eq}}^y}$ component is the dominant contribution for all four bands. Still, we find also sizable in-plane spin components which are around 5% of the out-of-plane one.

<table>
<thead>
<tr>
<th></th>
<th>b$_1$</th>
<th>b$_2$</th>
<th>b$_3$</th>
<th>b$_4$</th>
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<td>$\overline{S_{\text{eq}}^x}$</td>
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<td>0.4586</td>
<td>0.4622</td>
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</tbody>
</table>

For comparison, Figure S4(a)–(c) shows the electronic and spin structure of an isolated monolayer TaSe$_2$. The bands near the Fermi level are strongly $S_{\text{eq}}^z$-polarized. The in-plane spin components, $S_{\text{eq}}^x$ and $S_{\text{eq}}^y$, can be only found for the bands near the $\tilde{\Gamma}$ point at energies around 0.6 eV, which is not relevant for transport properties. In Figure S4(d) we highlight four bands near the Fermi level, b$_1$–b$_4$, similar to Figure S2(d). For those bands, we calculate spin textures and their contribution to the Fermi surface, as shown in Figure S5. We find that the in-plane spin components are too small to be visualized and thus are not shown in Figure S5. For an isolated monolayer TaSe$_2$, the out-of-plane

Table S1. The spin expectation values of the bands b$_1$–b$_4$ averaged over the BZ for the bilayer-CrI$_3$/monolayer-TaSe$_2$ vdW heterostructure.
Figure S2. First-principles-computed bands of bilayer-CrI$_3$/monolayer-TaSe$_2$ vdW heterostructure with SOC turned on in ncDFT+U calculation with $U = 2.0$ eV. In panels (a)–(c), the green open spheres emphasize the projections onto different monolayers, while the color of the solid lines correspond to (a) $S_{eq}^x$, (b) $S_{eq}^y$, and (c) $S_{eq}^z$ spin expectation value. In panel (d), we emphasize four bands near the Fermi level, $b_1$–$b_4$, with different colors that originate from the TaSe$_2$ layer.

Spin strongly dominates the bands, similar to the vdW heterostructure case. Also the contribution of the four bands to the Fermi surface are very similar, but in-plane components of the spin texture are strongly suppressed.

Similar to the vdW heterostructure case, we also calculate the average spin expectation values of the bands, which are summarized in Table SII. In contrast to the vdW heterostructure case, the in-plane spin-components are at maximum 0.5% of the out-of-plane one. Consequently, the monolayer TaSe$_2$ within the vdW structure shows roughly a 10-fold enhancement of the in-plane components of spin texture.
Figure S3. Spin textures and contribution to the Fermi surface of the four bands b1–b4, defined in Figure S2(d), for the bilayer-CrI3/monolayer-TaSe2 vdW heterostructure. In panel (d), the first BZ is indicated with the dashed hexagon. The green color indicates the contribution of band b4 to the Fermi surface, i.e., when $E_{b4} - E_F$ is within the energy window [-0.03, 0.03] eV. The arrows represent the in-plane spin components, via the scaled unit vector $0.04 \times (S_{eqx}^x, S_{eqy}^y)/\sqrt{(S_{eqx}^x)^2 + (S_{eqy}^y)^2}$ for better visualization, plotted only for the green Fermi surface. The red/blue color represents the out-of-plane spin component $S_{eqz}^z$. In panels (a)–(c), we plot the same as in panel (d), but for bands b1–b3, respectively.

Table S II. The spin expectation values of the bands b1–b4 averaged over the BZ for an isolated monolayer TaSe2.

<table>
<thead>
<tr>
<th></th>
<th>b1</th>
<th>b2</th>
<th>b3</th>
<th>b4</th>
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<tr>
<td>$S_{eqx}$</td>
<td>0.0013</td>
<td>0.0014</td>
<td>0.0021</td>
<td>0.0024</td>
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<tr>
<td>$S_{eqy}$</td>
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<td>0.0005</td>
<td>0.0015</td>
<td>0.0020</td>
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<tr>
<td>$S_{eqz}$</td>
<td>0.4874</td>
<td>0.4906</td>
<td>0.4908</td>
<td>0.4912</td>
</tr>
</tbody>
</table>
Figure S4. First-principles-computed bands of monolayer TaSe$_2$ with SOC turned on in ncDFT calculations. In panels (a)–(c), the color of the lines correspond to (a) $S_x^{eq}$, (b) $S_y^{eq}$, and (c) $S_z^{eq}$ spin expectation value. In panel (d), we emphasize four bands near the Fermi level, $b_1$–$b_4$, with different colors, similar to Figure S2(d).
Figure S5. Same as Figure S3 but for an isolated monolayer TaSe$_2$ whose bands $b_1$–$b_4$ are defined in Figure S4(d). The in-plane components of the spin expectation value, $S_{eq}^x$ and $S_{eq}^y$, are too small to be visualized.