Spin-orbit torque switching of Néel order in two-dimensional CrI$_3$

Fei Xue$^{1,2}$ and Paul M. Haney$^1$

$^1$Physical Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
$^2$Institute for Research in Electronics and Applied Physics & Maryland Nanocenter, University of Maryland, College Park, MD 20742

(Dated: April 13, 2021)

Spin-orbit torque enables electrical control of the magnetic state of ferromagnets or antiferromagnets. In this work we consider the spin-orbit torque in the 2-d Van der Waals antiferromagnetic bilayer CrI$_3$, in the $n$-doped regime. In the purely antiferromagnetic state, two individually inversion-symmetry broken layers of CrI$_3$ form inversion partners, like the well-studied CuMnAs and Mn$_2$Au. However, the exchange and anisotropy energies are similar in magnitude, unlike previously studied antiferromagnets, which leads to qualitatively different behaviors in this material. Using a combination of first-principles calculations of the spin-orbit torque and an analysis of the ensuing spin dynamics, we show that the deterministic electrical switching of the Néel vector is the result of dampinglike spin-orbit torque, which is staggered on the magnetic sublattices.

**Introduction.**— Spin-orbit torque is a mechanism for electrically switching thin-film magnets, and has the potential to enable scalable magnetic random access memory and devices for next-generation computing [1]. The effect occurs in magnetically ordered systems that lack inversion symmetry - such as heavy metal-ferromagnet bilayers [2, 3] - when a DC current or electric field is applied. Spin-orbit torque can be decomposed into a component that is even under time-reversal, known as the “fieldlike” torque, and a component that is odd under time-reversal, known as the “fieldlike” torque [4]. Knowledge of the dominant component of spin-orbit torque can help to identify the microscopic source of the torque and can assist in optimizing the effect [5].

In addition to switching ferromagnets, spin-orbit torque has been shown to switch antiferromagnets [6–11]. Antiferromagnets are of particular interest due to their insensitivity to stray magnetic fields and the fast time scales of their excitations [1, 8, 10]. It was shown [6] that spin-orbit torque is present in bulk antiferromagnets in which inversion symmetry is locally broken on individual magnetic sublattices, while the crystal lattice retains global inversion symmetry. More precisely, in antiferromagnets that are invariant under the combined operations of inversion and time-reversal, the spin-orbit torques acting on the magnetic sublattices cooperatively switch the antiferromagnetic Néel vector $\mathbf{L}$ [6, 7]. In these types of materials studied so far, such as CuMnAs and Mn$_2$Au, the magnetic exchange energy is much larger than other energy scales, and the mechanism for switching is a uniform fieldlike torque present on both magnetic sublattices [6, 7].

In this work we consider the spin-orbit torque in the recently discovered class of two-dimensional Van der Waals magnetic materials [12–14], exemplified by antiferromagnetic bilayer CrI$_3$. In this semiconducting material, the two magnetic CrI$_3$ layers are antiferromagnetically coupled and the ground state Néel vector is oriented perpendicular to the plane [12] (see Fig. 1). There is intense recent interest in this material due to its unique and potentially useful properties, including the tunability and control of its magnetic state through gating and doping [15–17] and its spin-filtering effects [18–20]. From the structure shown in Fig. 1(b), it’s clear that bilayer CrI$_3$...
shares some characteristics with previously mentioned antiferromagnets, such as CuMnAs: Inversion symmetry is locally broken on the magnetic sublattices (denoted A and B), while in the purely antiferromagnetic state, the bilayer is invariant under the combined operations of inversion plus time-reversal.

There is a key difference between this and previously studied materials, namely that the magnetic exchange constant is similar in magnitude to the magnetic anisotropy energy \([21, 22]\). This allows for significant current-induced canting of the sublattice magnetization and the development of a net magnetization, which breaks the combination of inversion plus time-reversal symmetry. For this material, we show that the important symmetry is instead a 2-fold rotational symmetry about the \(y\)-axis; this symmetry constrains the magnetic dynamics to a subspace of spin configurations and enables spin-orbit torque switching of the Néel vector. We also show that the dominant mechanism for spin-orbit torque switching of the Néel vector is the dampinglike torque, which is staggered on the two sublattices of the antiferromagnet. These conclusions are based on first-principles calculations of the spin-orbit torque as a function of the magnetic configuration, and an analysis of the ensuing spin dynamics.

**Spin dynamics in CrI\(_3\)** — We first consider the spin dynamics in CrI\(_3\) and show that the crystal symmetry confines the spins a subspace of magnetic configurations. In the general case, the time evolution of the spin orientations \(\hat{m}^{A,B}\) are described by the coupled set of Landau-Lifshitz-Gilbert (LLG) equations \([23–25]\):

\[
\frac{d\hat{m}^{A,B}}{dt} = \hat{m}^{A,B} \times \left( \frac{\gamma}{m} \frac{\delta E}{\delta \hat{m}^{A,B}} \right) + \mathbf{T}^{A,B},
\]

where \(m\) is the magnitude of the magnetic moment (assumed equal on both sublattices), \(\gamma\) is the absolute value of the gyromagnetic ratio, and \(\alpha\) is the Gilbert damping parameter. The energy \(E\) is comprised of an easy-axis anisotropy (along \(\hat{z}\)) and Heisenberg exchange coupling: \(E(\hat{m}^{A}, \hat{m}^{B}) = -\frac{1}{2}mH_A[(\hat{m}^{A} \cdot \hat{z})^2 + (\hat{m}^{B} \cdot \hat{z})^2] + mH_E(\hat{m}^{A} \cdot \hat{m}^{B})\), where \(H_A\) and \(H_E\) are the effective magnetic fields from anisotropy and exchange, respectively. \(\mathbf{T}^{A,B}\) is the spin-orbit torque on the A,B sublattice, which we discuss in more detail in the next section.

The dynamics of the coupled spin system can be quite complex in the general case and analytical treatments are often not possible. A commonly studied limit is \(H_E \gg H_A\), which is applicable to many metallic antiferromagnets. In this case the system can be simplified to a single vector equation of motion for the Néel vector \(\hat{L} = (\hat{m}^{A} - \hat{m}^{B})/2\), while the net magnetization \(\mathbf{M} = (\hat{m}^{A} + \hat{m}^{B})/2\) is no longer an independent variable \([24, 25]\). For CrI\(_3\), \(H_E \approx H_A\) \([21]\) and this simplification is no longer valid. We next discuss an alternative simplification that follows the system crystal symmetry.

We consider an applied electric field along the \(y\)-axis and spin configurations that are staggered in \((x, z)\) and uniform in \(y\): \(m_{x/z}^A = -m_{x/z}^B\) and \(m_y^A = m_y^B\). In this case, the system retains 2-fold rotation symmetry about the \(y\)-axis (see Fig. 1). Any torque on the spins is therefore symmetry-constrained to satisfy \(T_{x/z}^A = -T_{x/z}^B\) and \(T_y^A = T_y^B\). The \(x\) and \(z\) components of the spins then remain staggered and the \(y\) components remain uniform. The trajectory of the spins is thus symmetry-confined to the subspace \((L_x, M_y, L_z)\). This motivates the definition of a “mixed” order parameter \(\hat{N} \equiv (L_x, M_y, L_z)\) \([26]\). Eq. 1 leads to the following equation of motion for \(\hat{N}\):

\[
\frac{d\hat{N}}{dt} = \hat{N} \times \left( \frac{\gamma}{m} \frac{\delta E}{\delta \hat{N}} + \alpha \frac{d\hat{N}}{dt} \right) + \mathbf{T}^{\text{odd}}(\hat{N} \times \hat{p}) + \mathbf{T}^{\text{even}}(\hat{N} \times (\hat{p} \times \hat{N})),
\]

where the energy is comprised of the easy-axis anisotropy along \(\hat{z}\) and an effective hard-axis anisotropy along \(\hat{y}\), which encodes the magnetic exchange:

\[
E(\hat{N}) = -\frac{1}{2}mH_A(\hat{N} \cdot \hat{z})^2 + mH_E(\hat{N} \cdot \hat{y})^2.
\]

The last two terms on the right-hand-side of Eq. 2 are the spin-orbit torques, consisting of terms that are odd and even under time reversal, as indicated (note the prefactors \(T^{\text{odd,even}}\) are assumed to be constants, independent of \(\hat{N}\)). \(\hat{p}\) is a direction related to the system symmetry, and the spin-orbit torque vanishes when \(\hat{N}\) is aligned to \(\hat{p}\). We discuss the spin-orbit torque terms in detail in the next section; for now they can be taken as a general representation of a torque which is spanned by the two vectors perpendicular to \(\hat{N}\). We have verified that fluctuations away from the \(\hat{N}\) subspace do not alter the steady state dynamics \([27]\). One important feature of this system which enables this simplification is that the easy-axis anisotropy is perpendicular to the axis of 2-fold rotation symmetry. If this were not the case, then the anisotropy torque would immediately drive the spin configuration out of the subspace.

The simple form of the time evolution of \(\hat{N}\) allows for an analytical treatment of fixed points and an intuitive description of the dynamics. For \(\hat{p}\) along \(\hat{z}\), the dampinglike torque \(\mathbf{T}^{\text{even}}\) competes with \(\gamma\alpha(H_E + H_A)\) while the fieldlike torque \(\mathbf{T}^{\text{odd}}\) competes with \(\gamma H_A\). For CrI\(_3\), \(H_A \approx 1.77\) T, \(H_E \approx 0.76\) T, and \(\alpha \approx 0.04\) \([21]\), so that the dampinglike torque is the dominant mechanism for switching. In the next section we show that \(\hat{p}\) has the standard \(x\) component, and due to additional mirror plane symmetry breaking in CrI\(_3\), also has a \(z\) component. For \(\hat{p} = (p_x, 0, p_z)\), the fixed points to lowest order in spin-orbit torque are \(\hat{N} = (-\frac{T^{\text{odd}}}{\gamma H_A} \pm \frac{T^{\text{even}}}{\gamma(2H_E + H_A)}, \pm 1)\). The instability threshold
to switch between fixed points is:

$$|T^{\text{even}}_{ij}| > \gamma \alpha (H_E + H_A). \quad (4)$$

A typical switching trajectory is shown in Fig. 1(d): the spin-orbit torque drives $\mathbf{N}$ from north pole to the fixed point close to south pole. For previously studied antiferromagnets, $H_E$ is sufficiently large so that switching with $T^{\text{even}}$ is not feasible, and the switching relies instead on $T^{\text{odd}}$ exceeding $H_A$. The utilization of $T^{\text{even}}$ for switching the Néel vector of CrI$_3$ is one of the material’s distinguishing features.

**Microscopic calculations of spin-orbit torques** — Having established the relevant degrees of freedom for the spin configuration in CrI$_3$ as $\mathbf{N}$, we next present microscopic calculations of the spin-orbit torque per applied electric field - a quantity known as the “torkance” - as a function of $\mathbf{N}$. The procedure for this calculation is well-established [28, 29], and we briefly provide a description here and refer the reader to the Supplemental Information for more technical details. We first obtain the Hamiltonian in a localized atomic orbital basis using a combination of Quantum Espresso [30] and Wannier90 [31]. We then utilize linear response theory to compute the torkance on each magnetic sublattice. The torkance is classified as even or odd according to its behavior under time-reversal. We denote the $j$th component of the torkance on atom A,B in response to an electric field along the $i$-direction with $\tau_{ij}^{AB}$. The even and odd components of the torkance are given by:

$$\left(\tau_{ij}^{AB}\right)^{\text{even}} = 2e \text{ Im} \sum_{n,m \neq n} f_n \left(\frac{\partial H}{\partial k_i}\right)_{n,m,n,m} \left(T_j^{AB}\right)_{m,n}, \quad (5)$$

$$\left(\tau_{ij}^{AB}\right)^{\text{odd}} = -e \sum_n \left(\frac{\partial f_n}{\partial E_n}\right) \left(\frac{\partial H}{\partial k_i}\right)_{n,n} \left(T_j^{AB}\right)_{n,n} \quad (6)$$

The sum in Eqs. 5-6 is over eigenstates $|\psi_n\rangle$ of the $\mathbf{k}$-dependent Hamiltonian $H_{\mathbf{k}}$, where $\mathbf{k}$ is the Bloch wave vector and the eigenstate label $n$ includes $\mathbf{k}$ and band index. $(O)_{n,m} = \langle \psi_n | O | \psi_m \rangle$ is the matrix element of the operator $O$, and $f_n = (e^{(E_n - \mu)/k_B T} + 1)^{-1}$ is the equilibrium Fermi-Dirac distribution function. $\mu$ is the Fermi level, $\eta$ is the broadening parameter, and $e$ is the electron charge. The atom-resolved torque operator is $\mathbf{T}^{AB} = \frac{i}{\hbar} \left\{ \mathbf{S}, \Delta \right\}$, where $\mathbf{S}$ is the spin operator, $\Delta$ is the spin-dependent exchange-correlation potential, and $\mathbf{P}^{AB}$ is the projection operator onto the orbitals centered on atomic site A (B). To compute the torque as a function of $\mathbf{N}$, we manually rotate the spins on A and B sublattices.

Figure 2 shows the $\mathbf{N}$-dependence of the torkance with (a) and (b) showing the dampinglike (time-reversal even) and fieldlike (time-reversal odd) torkance, respectively. The fixed points of both dampinglike and fieldlike torkance lie in the $L_z - L_x$ plane, away from the $L_z = 0$ equator. This is an important feature and is a consequence of the lack of mirror symmetry with respect to the $yz$ plane. This position of the fixed point ensures that the spin-orbit torque drives $\mathbf{N}$ to a point in the northern or southern hemisphere; after the spin-orbit torque is removed, $\mathbf{N}$ then relaxes to the nearest easy-axis along $+z$ or $-z$. Previous studies on systems with similar in-plane mirror symmetry breaking, such as WTe$_2$-Py heterostructures [29, 32–34], have verified that this symmetry breaking results in a spin-orbit torque that drives the magnetic order parameter to a point away from the equator. Exploiting this property has emerged as an approach for deterministically switching perpendicularly magnetized thin films with spin-orbit torque, and we show here that this also enables switching of the perpendicular Néel vector.

We note that the $\mathbf{N}$-dependence of the torkance is quite complex, deviating substantially from the simple, lowest order form used in the analysis of the previous section. In the Supplemental Information, we provide the full symmetry-allowed expansion of the torkance and quantify the substantial contribution from higher order terms. We additionally find that the fixed points for even and odd torkance are different. These features of the microscopically computed torkance have important consequences for the details of the dynamics of $\mathbf{N}$ under spin-orbit torque, which we show in the next section.

We next consider the torkance versus Fermi level for $\mathbf{N}$ along $z$ and $x$ directions, shown in Figs. 3 (a) and (b), respectively. Both even and odd components are peaked for Fermi energies near the conduction band minimum. For $\mathbf{N} = \hat{z}$, the even torkance is approximately $1 \ e a_0 / \hbar$ ($a_0 \approx 0.0529$ nm is the Bohr radius) at 0.1 eV above the conduction band minimum, which is larger than the even torkance in the ferromagnetic Pt/Co bilayer ($\approx 0.6 \ e a_0 / \hbar$) [28]. This large magnitude is due to band
crossings in the conduction band from $p$-orbitals of the heavy Iodine atoms. For $\mathbf{N} = \hat{x}$, the even torkance magnitude is around $0.4 \text{ e}a_0/\hbar$. The even torkance for this $\mathbf{N}$ configuration is solely a consequence of the in-plane mirror symmetry breaking. This value is notably larger than the corresponding torkance derived from in-plane mirror symmetry breaking in the ferromagnetic 1T'-WTe$_2$/Co bilayer ($\approx 0.1 \text{ e}a_0/\hbar$) [29].

**Magnetization dynamics** — Given the significant deviation of the $\mathbf{N}$-dependence of the microscopically computed spin-orbit torque from the simple form given in the earlier analysis, it's worthwhile to compute the spin dynamics with the *ab initio* spin-orbit torque (Fig. 2) as input into the coupled LLG equations (Eq. 1). $\mathbf{N}$ is parameterized by spherical coordinates ($\theta, \phi$), and we use a bilinear interpolation of a dense $80 \times 80$ mesh of spin-orbit torque obtained from first-principles to obtain the full $\mathbf{N}$-dependence.

Figure 4 shows the spin-orbit torque driven dynamics. We find that the spin-orbit torque can either induce switching or induce steady state oscillations of $\mathbf{N}$. Figure 4(a) shows that for an applied electric field $E = -2.3 \text{ V}/\mu\text{m}$, the spin-orbit torque switches the Néel order $L_z$ from the north pole to the southern hemisphere within $100 \text{ ps}$ and generates a finite in-plane magnetization $M_y$. By separately turning off the fieldlike (odd) or damping-like (even) contributions to the spin-orbit torque, we find that the switching of $L_z$ originates from the damping-like torque, while the fieldlike torque helps to accelerate the switching dynamics and reduce the $E$-field threshold. Figure 4(b) shows an oscillating steady state for $E = 3 \text{ V}/\mu\text{m}$, with a frequency of approximately $80 \text{ GHz}$. We find that both dampinglike and fieldlike torque are required to induce steady state oscillation.

We summarize the final steady states as a function of the applied field $E$ for two initial magnetization configurations $L_z = +1$ and $L_z = -1$ in Fig. 4 (c) and (d), respectively. The switching of the Néel vector occurs at approximately $|E| = 2 \text{ V}/\mu\text{m}$. This threshold compares well with the estimate provided by Eq. 4. Reaching the larger scale oscillations at large applied $E$ will rely on the material to sustain large power dissipation, which depends in turn on factors such as the carrier mobility. The flatness of the conduction bands implies a low mobility, as seen experimentally [35], which should enable larger applied electric fields. Fig. 4 (c) and (d) demonstrate hysteretic switching of the Néel vector, and are related by mirror symmetry about the $xz$ plane.

**Discussion** — The experimental detection of the Néel vector reversal is challenging. For bilayer CrI$_3$, out-of-plane magnetic-optical Kerr effect (MOKE) imaging has previously been used to discriminate between $L_z = +1$ and $L_z = -1$ [16], and transport effects such as nonlinear anisotropic magnetoresistance can also detect $\mathbf{N}$ [11]. We also note that the moderate exchange energy leads to the development of a substantial steady state in-plane magnetization of the driven system, which may be detected experimentally with in-plane MOKE. Aside from the particulars of CrI$_3$, we show generally that antiferromagnets in the weak to moderate exchange coupling regime exhibit different behaviors from their more commonly studied large $H_E$ counterparts. The switching cri-

---
teria for these antiferromagnets is reduced by a factor of magnetic damping, offering potentially easier routes to electrical manipulation. Continued progress in the field of Van der Waals antiferromagnets should provide further opportunities for unique modes of electrical control of these materials.

F.X. acknowledges support under the Cooperative Research Agreement between the University of Maryland and the National Institute of Standards and Technology Physical Measurement Laboratory, Award 70NANB14H209, through the University of Maryland.

[1] A. Manchon, J. Železný, I. M. Miron, T. Jungwirth, J. Sinova, A. Thiaville, K. Garello, and P. Gambardella, Reviews of Modern Physics 91, 035004 (2019).
[2] L. Liu, T. Moriyama, D. Ralph, and R. Buhrman, Physical review letters 106, 036601 (2011).
[3] I. M. Miron, K. Garello, G. Gandin, P.-J. Zermatten, M. V. Costache, S. Auffret, S. Bandiera, B. Rodmacq, A. Schuh, and P. Gambardella, Nature 476, 189 (2011).
[4] As described in Ref. [36], time-reversal even (odd) torque coincides with dampinglike (fieldlike) torque only for odd values of $\ell$ in the expansion of torque in vector spherical harmonic basis (which includes the lowest order term). Nevertheless, we use these terms interchangeably for ease of reference.
[5] D. Go, F. Freimuth, J.-P. Hanke, F. Xue, Q. Gomonay, K.-J. Lee, S. Blügel, P. M. Haney, H.-W. Lee, and Y. Mokrousov, Physical Review Research 2, 033401 (2020).
[6] J. Železný, H. Gao, K. Výborný, J. Zemen, J. Mašek, A. Manchon, J. Wunderlich, J. Sinova, and T. Jungwirth, Phys. Rev. Lett. 113, 157201 (2014).
[7] P. Wadley, B. Howells, J. Železný, C. Andrews, V. Hills, R. P. Campion, V. Novák, K. Olejník, F. Maccherozzi, S. Dhesi, et al., Science 351, 587 (2016).
[8] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, Nature Nanotechnology 11, 231 (2016).
[9] M. Meinert, D. Graulich, and T. Matalla-Wagner, Phys. Rev. Applied 9, 064040 (2018).
[10] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, Rev. Mod. Phys. 90, 015005 (2018).
[11] J. Godinho, H. Reichlová, D. Kriegner, V. Novák, K. Olejník, Z. Kašpar, Z. Šobiáň, P. Wadley, R. P. Campion, R. M. Otzoa, P. E. Roy, J. Železný, T. Jungwirth, and J. Wunderlich, Nature Communications 9, 4686 (2018).
[12] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, and X. Xu, Nature 546, 270 (2017).
[13] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, C. Wang, Y. Wang, Z. Q. Qiu, R. J. Cava, S. G. Louie, J. Xia, and X. Zhang, Nature 546, 265 (2017).
[14] Y. Deng, Y. Yu, Y. Song, J. Zhang, N. Z. Wang, Z. Sun, Y. Yi, Y. Z. Wu, S. Wu, J. Zhu, J. Wang, X. H. Chen, and Y. Zhang, Nature 563, 94 (2018).
[15] S. Jiang, J. Shan, and K. F. Mak, Nature Materials 17, 406 (2018).
[16] B. Huang, G. Clark, D. R. Klein, D. MacNeill, E. Navarro-Moratalla, K. L. Seyler, N. Wilson, M. A. McGuire, D. H. Cobden, D. Xiao, W. Yao, P. Jarillo-Herrero, and X. Xu, Nature Nanotechnology 13, 544 (2018).
[17] S. Jiang, L. Li, Z. Wang, K. F. Mak, and J. Shan, Nature Nanotechnology 13, 549 (2018).
[18] T. Song, X. Cai, M. W.-Y. Tu, X. Zhang, B. Huang, N. P. Wilson, K. L. Seyler, L. Zhu, T. Taniguchi, K. Watanabe, M. A. McGuire, D. H. Cobden, D. Xiao, W. Yao, and X. Xu, 360, 1214 (2018).
[19] D. R. Klein, D. MacNeill, J. L. Lado, D. Soriano, E. Navarro-Moratalla, K. Watanabe, T. Taniguchi, S. Manni, P. Canfield, J. Fernández-Rossier, and P. Jarillo-Herrero, 360, 1218 (2018).
[20] Z. Wang, I. Gutiérrez-Lezama, N. Ubrig, M. Kroner, M. Gibertini, T. Taniguchi, K. Watanabe, A. Imamoğlu, E. Giannini, and A. F. Morpurgo, Nature Communications 9, 2516 (2018).
[21] X.-X. Zhang, L. Li, D. Weber, J. Goldberger, K. F. Mak, and J. Shan, Nature Materials 19, 838 (2020).
[22] N. Richter, D. Weber, F. Martin, N. Singh, U. Schwingenschlögl, B. V. Lutsch, and M. Kläui, Physical Review Materials 2, 020404 (2018).
[23] M. D. Stiles and J. Militat, Spin-transfer torque and dynamics, in Spin Dynamics in Confined Magnetic Structures III, edited by B. Hillebrands and A. Thiaville (Springer Berlin Heidelberg, Berlin, Heidelberg, 2006) pp. 225–308.
[24] H. V. Gomonay and V. M. Loktev, Phys. Rev. B 81, 144427 (2010).
[25] A. Manchon, Journal of Physics: Condensed Matter 29, 104002 (2017).
[26] For spin configurations in the $N$-subspace ($m^{A}_{x/y} = -m^{B}_{x/y}$, $m^{A}_{z} = m^{B}_{z}$), it’s easy to show that $|N| = 1$. This spin configuration subspace can also be represented with $\hat{m}^{A}$ alone, with the value of $\hat{m}^{B}$ implied by symmetry.
[27] To determine the robustness of the projection of the spin configuration to the $N$ subspace, we solved for the steady state for $20^{4}$ possible initial conditions in the 4-dimensional space ($\theta^{A}, \phi^{A}, \theta^{B}, \phi^{B}$), which characterize the orientations of the spins on A and B. We find that the steady states from all initial conditions belong to the $N$ subspace.
[28] F. Freimuth, S. Blügel, and Y. Mokrousov, Phys. Rev. B 90, 174423 (2014).
[29] F. Xue, C. Rohmann, J. Li, V. Amin, and P. Haney, Phys. Rev. B 102, 014401 (2020).
[30] P. G. et al., Journal of Physics: Condensed Matter 29, 465901 (2017).
[31] A. A. Mostofi, J. R. Yates, G. Pizzi, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, Computer Physics Communications 185, 2309 (2014).
[32] D. MacNeill, G. M. Stiehl, M. H. D. Guimaraes, R. A. Buhrman, J. Park, and D. C. Ralph, Nature Physics 13, 300 (2016).
[33] D. MacNeill, G. M. Stiehl, M. H. D. Guimaraes, N. D. Reynolds, R. A. Buhrman, and D. C. Ralph, Phys. Rev. B 96, 054450 (2017).
[34] G. M. Stiehl, R. Li, V. Gupta, I. E. Baggari, S. Jiang, H. Xie, L. F. Kourkoutis, K. F. Mak, J. Shan, R. A. Buhrman, and D. C. Ralph, Phys. Rev. B 100, 184402 (2019).
[35] R. A. Patil, H.-W. Tu, M.-H. Jen, J.-J. Lin, C.-C. Wu, C.-C. Yang, D. Van Pham, C.-H. Tsai, C.-C. Lai, Y. Liou, et al., Materials Today Physics 12, 100174 (2020).

[36] K. Belashchenko, A. A. Kovalev, and M. van Schilfgaarde, Physical Review B 101, 020407 (2020).
Spin-orbit torque switching of Néel order in two-dimensional CrI₃: Supplemental Materials

Fei Xue¹,² and Paul M. Haney¹

¹Physical Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
²Institute for Research in Electronics and Applied Physics & Maryland Nanocenter, University of Maryland, College Park, MD 20742

(Dated: April 13, 2021)

FIRST-PRINCIPLES DETAILS

We use Quantum ESPRESSO [1] to compute the electronic structure of bilayer CrI₃. We adopt the experimental unit cell parameters [2] of bilayer CrI₃ (space group C2/m): \(a = 0.6904\) nm, \(b = 1.1899\) nm, \(c = 0.7008\) nm, and \(\beta = 108.74^°\). In the Quantum ESPRESSO implementation, we use the pseudopotentials from PSLibrary [3] generated with a scalar relativistic calculation using Projector Augmented-Wave method [4] and Perdew-Burke-Ernzerhof exchange correlations [5]. We utilize a \(7 \times 12 \times 1\) Monkhorst-Pack mesh [6], \(1360\) eV cutoff energy, \(1.36 \times 10^{-3}\) eV total energy convergence threshold, and \(0.08\) eV/nm force convergence threshold. We add a Hubbard on-site energy \(U = 3\) eV on Cr atoms [7]. We next utilize Wannier90 [8] to obtain the Hamiltonian in an atomic basis. We project plane-wave solutions onto atomic \(s,d\) orbitals of Cr atoms, \(p\) orbitals of I atoms. We then symmetrize the Wannier-like tight-binding Hamiltonian using TBmodels [9] since the presence of slight asymmetry in the tight-binding Hamiltonian results in symmetry-disallowed torque, and we remove small spin-dependent hopping terms. The final symmetrized tight-binding band structures match those obtained with plane-wave methods. We add on-site spin-orbit coupling terms \(\alpha \mathbf{L} \cdot \mathbf{S}\), where \(\mathbf{L}\) and \(\mathbf{S}\) are the orbital angular momentum and spin operators, respectively. We use \(\alpha = [90, 580]\) meV for Cr, and I [10]. Adding spin-orbit coupling “by hand” in this manner requires that Wannier orbitals are not localized in order to ensure their forms are spherical harmonics consistent with the standard representation of \(\mathbf{L}\). We adopt this approach because it is technically easier to achieve a good Wannier projection of a collinear magnetized Hamiltonian, and the on-site spin-orbit coupling approximation yields accurate results (see Fig. 1 to see a comparison of band structure obtained with Quantum ESPRESSO and Wannier orbitals). We use a dense \(k\) mesh of \(400 \times 232\) to evaluate the torque, given by Eqs. 5 and 6 of the main text. In the implementation of Eqs. 5 and 6, we adopt the approximation [11] that Wannier orbitals are perfectly localized on atomic sites and spin matrix is half of Pauli matrix in the space spanned by Wannier orbitals. We use a constant broadening parameter \(\eta = 25\) meV for the results presented. The corresponding constant electron momentum relaxation time \(\tau = \hbar/2\eta = 13\) fs. Since...
the critical Néel temperature of bilayer CrI$_3$ is around 40 Kalvin, we adopt a low temperature $k_B T = 3$ meV.

STABILITY ANALYSIS

The dynamics of two coupled spins A and B are described the set of Landau-Lifshitz-Gilbert (LLG) equations: [12–14]:

$$\frac{d\mathbf{m}^A}{dt} - \gamma H_A \left( \mathbf{m}^A \times \mathbf{z} \right) \left( \mathbf{m}^A \cdot \mathbf{z} \right) + \gamma H_E \left( \mathbf{m}^A \times \mathbf{m}^B \right) + \mathbf{T}^A,$$

$$\frac{d\mathbf{m}^B}{dt} - \gamma H_A \left( \mathbf{m}^B \times \mathbf{z} \right) \left( \mathbf{m}^B \cdot \mathbf{z} \right) + \gamma H_E \left( \mathbf{m}^B \times \mathbf{m}^A \right) + \mathbf{T}^B$$

(1)

where $\gamma$ is the absolute value of the electron gyromagnetic ratio, $H_A$ is the magnetic anisotropy field strength, $\mathbf{z}$ is the magnetic easy-axis, $H_E$ is the antiferromagnetic exchange field, $\alpha$ is the damping parameter, and $\mathbf{T}^{(A,B)}$ is the spin-orbit torque on the (A,B) sublattice. It’s convenient to work in spherical coordinates, where the magnetization vector is given by $\mathbf{m} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$. The torque is always perpendicular to the magnetization, so that it can be expressed in terms of the $\mathbf{e}_\theta, \mathbf{e}_\phi$ components, where $\mathbf{e}_\theta \equiv (\cos \theta \cos \phi, \cos \theta \sin \phi, -\sin \theta)$ and $\mathbf{e}_\phi \equiv (-\sin \phi, \cos \phi, 0)$. The matrix form of Eq. 1 is

$$\begin{pmatrix} \dot{\phi}^A \\ \dot{\theta}^A \\ \dot{\phi}^B \\ \dot{\theta}^B \end{pmatrix} = \frac{1}{1 + \alpha^2} \begin{pmatrix} \frac{1}{\sin \theta \alpha} & \frac{\alpha}{\sin \theta \alpha} & 0 & 0 \\ \frac{1}{\sin \theta \alpha} & \frac{1}{\sin \theta \alpha} & 0 & 0 \\ 0 & 0 & \frac{1}{\sin \theta \alpha} & \frac{\alpha}{\sin \theta \alpha} \\ 0 & 0 & -\alpha & \frac{1}{\alpha \sin \theta \alpha} \end{pmatrix} \begin{pmatrix} T^A_{\phi} \\ T^A_{\theta} \\ T^B_{\phi} \\ T^B_{\theta} \end{pmatrix},$$

(2)

where $T^{\phi,\theta}_{A,B}$ is obtained by projecting the right hand side of Eq. 1 to the $\mathbf{e}_\phi,\mathbf{e}_\theta$ directions on the A and B sublattices.

The fixed points and their stability are determined by the set of torque expressions $\Gamma = (T^A_{\phi}, T^A_{\theta}, T^B_{\phi}, T^B_{\theta})$. A fixed points satisfies $\Gamma = 0$, and its stability is determined by the eigenvalues of the dynamic matrix $D$. $D$ is given by the product of the matrix given on the right-hand-side of Eq. 2 and the Jacobian matrix derived from $\Gamma$ evaluated at the fixed point. A fixed point goes from stable to unstable as the real part of its eigenvalue goes from negative to positive.

Analysis of the N subspace

As discussed in the main text, the 2-fold rotational symmetry about the $y$-direction constrains the spins to the subspace spanned by $\mathbf{N} = (L_x, M_y, L_z)$. Due to the lack of mirror symmetry about the $yz$ plane, the lowest order fieldlike and dampinglike torque have the form of $\mathbf{m} \times \mathbf{p}$ and $\mathbf{m} \times (\mathbf{p} \times \mathbf{m})$, respectively, where $\mathbf{p} = (p_x, 0, p_z)$. 2-fold rotational symmetry about the $y$-axis leads to the following relation between the torque on A and B sublattices:

$$T^A_y = T^B_y,$$

$$T^A_{x,z} = -T^B_{x,z}.$$  

(3)  

(4)

The anisotropy field gives rise to the stable initial state $(L_x, M_y, L_z) = (0,0,\pm1)$ and we are interested in the condition where the spin-orbit torque drives the system away from the equilibrium state. To avoid the singular spherical coordinates near these points, we perform an index permutation $(x, y, z) \rightarrow (z, x, y)$, so that the magnetic subspace is now labelled by $(M_x, L_y, L_z)$.

In the subspace of $(M_x, L_y, L_z)$, $\theta^B = \pi - \theta^A, \phi^B = -\phi^A$. We can verify that the torque in Eq. 2 are staggered, so the $4 \times 4$ matrix form of LLG equation becomes two identical $2 \times 2$ matrices:

$$\begin{pmatrix} \dot{\phi}^A \\ \dot{\theta}^A \\ -\dot{\phi}^A \\ -\dot{\theta}^A \end{pmatrix} = \frac{1}{1 + \alpha^2} \begin{pmatrix} \frac{1}{\sin \theta \alpha} & \frac{\alpha}{\sin \theta \alpha} & 0 & 0 \\ \frac{1}{\sin \theta \alpha} & \frac{1}{\sin \theta \alpha} & 0 & 0 \\ 0 & 0 & \frac{1}{\sin \theta \alpha} & \frac{\alpha}{\sin \theta \alpha} \\ 0 & 0 & -\alpha & \frac{1}{\alpha \sin \theta \alpha} \end{pmatrix} \begin{pmatrix} T^A_{\phi} \\ T^A_{\theta} \\ -T^A_{\phi} \\ -T^A_{\theta} \end{pmatrix},$$

(5)  

(6)
Now we can drop the sublattice subscript and the equilibrium state is obtained by solving the equations $(T_\phi, T_\theta) = 0$. We can find solutions to this set of nonlinear equations with the ansatz $\theta = \pi/2 + a, \phi = \pi/2 + b$ where $a, b \ll 1$ by assuming small spin-orbit torque terms. By expanding all terms up to the first order of spin-orbit torques, we find:

$$a = \frac{T^{\text{odd}} p_z}{\gamma H_A}, \quad b = -\frac{T^{\text{even}} p_z}{\gamma(2H_E + H_A)}.$$  \hfill (7)

This equilibrium corresponds to the magnetization configuration $(M_x, L_y, L_z) = \left(\frac{T^{\text{even}} p_z}{\gamma(2H_E + H_A)}, 1, -\frac{T^{\text{odd}} p_z}{H_A}\right)$.

The dynamic matrix $D$ up to the linear order of $\alpha, a, b$, and the spin-orbit torque terms is

$$D = \begin{pmatrix}
-T^{\text{even}} p_y - \alpha(2H_E + H_A) & H_A - T^{\text{odd}} p_y \\
-2H_E - H_A + T^{\text{odd}} p_y & -T^{\text{even}} p_y - H_A \alpha
\end{pmatrix}. \hfill (8)
$$

The two eigenvalues are

$$\lambda = -T^{\text{even}} p_y - (H_E + H_A)\alpha \pm i \left[\sqrt{H_A(2H_E + H_A)} - \frac{T^{\text{odd}} p_y H_E}{\sqrt{H_A(2H_E + H_A)}}\right]. \hfill (9)
$$

The switching condition is then $p_y T^{\text{even}} < -\alpha(H_E + H_A)$. This analysis reveals the key ingredients of staggered dampinglike torque: the torque component along the direction perpendicular to the easy-axis drives the net magnetization along the direction perpendicular to both torque direction and easy-axis direction while the torque component along the direction parallel to the easy-axis switches the Néel order from one hemisphere to the other. Comparing to the fieldlike torque, the dampinglike torque only needs to compete with the total strength of exchange and anisotropy field multiplying a small Gilbert damping factor. The staggered dampinglike torque is therefore more favored to drive the AFM system when the exchange and anisotropy field have the same order of magnitude.

**Analysis of general case with PT symmetry**

For completeness, we also present a stability analysis for a system which is symmetric under inversion+time reversal operations (i.e. invariance under PT, where $P$ is parity and $T$ is time reversal). For this case, we consider easy-axis anisotropy along $y$. The assumption of PT symmetry leads to spin-orbit torques of the form $T^{\text{odd}} = \pm T^{\text{even}}(m^{A,B} \times y)$ and $T^{\text{A,B}} = \pm T^{\text{even}}(m^{A,B} \times (y \times m^{A,B}))$, where the plus (minus) sign corresponds to the torque on the A (B) sublattice. The fieldlike (time-reversal odd) and dampinglike (time-reversal even) torques are uniform and staggered, respectively. The fixed points are given by $L_y = \pm 1$, or $\theta^A = \theta^B = \pi/2$, $\phi^A = -\phi^B = \pm \pi/2$. For this case it’s necessary to evaluate the full $4 \times 4$ Jacobian derived from Eq. 2.

We first consider purely dampinglike torque (so that $T^{\text{odd}} = 0$). We obtain two unique values of the eigenvalues of the dynamic matrix $D$ evaluated at $L_y = +1$:

$$\lambda = -T^{\text{even}} - \alpha(H_E + H_A) \pm \sqrt{-2H_E H_A - H_A^2 + 2(H_E + H_A) T^{\text{even}} \alpha + H_A^2 \alpha^2 - T^{\text{even}}2\alpha^2}, \hfill (10)$$

where we have expanded the above to lowest order in $\alpha$. The real part of $\lambda$ changes sign for $T^{\text{even}} < -\alpha(H_E + H_A)$. This threshold is difficult to achieve in the limit of large exchange coupling strength.

We next consider purely fieldlike torque (so that $T^{\text{A,B}} = 0$). The eigenvalues of the resulting dynamic matrix are:

$$\lambda = T^{\text{odd}} \alpha - \alpha(H_E + H_A) \pm \sqrt{H_E^2 + H_A^2 \alpha^2 - (T^{\text{odd}} - (H_A + H_E))^2}, \hfill (11)$$

The condition of having positive real part of eigenvalues is $T^{\text{odd}} > K$.

**SYMMETRY-CONSTRAINED FORMS OF SPIN-ORBIT TORKANCE**

In this section we provide the symmetry-constrained forms of the spin-orbit torque and fit the ab initio results to these forms. In each layer of CrI$_3$, we only have one mirror plane $xz$. The time-reversal even and odd torque under
the applied field in $y$ direction are described by the symmetry-constrained expansion:

$$\tau_{\text{even}} = \sum_{m,n} [A_{mn}^e \cos(2m\theta) \sin((2n+1)\phi) + B_{mn}^e \sin(2m\theta) \sin(2n\phi)] e_\phi$$

$$\tau_{\text{odd}} = \sum_{m,n} [C_{mn}^o \cos((2m+1)\theta) \cos((2n+1)\phi) + D_{mn}^o \sin((2m+1)\theta) \cos(2n\phi)] e_\phi$$

where $m(n) = 0, 1, 2, ...$. Note that coefficients $A, B, C, D$ are related since we need to ensure that the torque is independent of $\phi$ when $\theta = 0, \pi$. We can immediately find that the conventional dampinglike and fieldlike forms of the torque correspond to the lowest order contributions:

$$\tau_{\text{even}} = A_{00}^e \sin \phi e_\phi - A_{00}^o \cos \theta \cos \phi e_\phi + D_{00}^o \sin \theta e_\theta = \tau_{\text{even}}^e (m \times (m \times (p_x, 0, p_z)))$$

$$\tau_{\text{odd}} = A_{00}^o \cos \theta \cos \phi e_\phi - A_{00}^o \sin \phi e_\theta + B_{00}^o \sin \theta e_\phi = \tau_{\text{odd}}^o (m \times (p_x, 0, p_z))$$

The unconventional symmetry direction $(p_x, 0, p_z)$ is a consequence of the absence of mirror symmetry in both $xy$ and $yz$ planes. Fig. 2 clearly shows the substantial higher-order contributions to both even and odd torques. These higher order terms complicate the global torque sphere described in the main text.

**SPIN-ORBIT TORQUE IN THE PURE NÉEL SPACE**

Here we present our first-principle results of spin-orbit torque in the pure Néel space, i.e., $m(A) = -m(B)$. In this case, the invariance under inversion+time reversal relates the torque on the magnetic sublattices: The time-reversal even (dampinglike) torque is staggered while the time-reversal odd (fieldlike) torque is uniform. Figure 3 summarizes our numerical results for $\mu = 60$ meV above the conduction band edge. The results show similar features compared to the torque in $N$-space shown in the main text, with fixed points in the $xz$ plane. However, knowledge of the torques in $L$-space is not sufficient for determining the spin dynamics since the anisotropy term immediately drives
FIG. 3. (Color online) Angular dependence of the dampinglike (a) and fieldlike (b) torkance on the Néel order direction ($\theta, \phi$) for one layer of bilayer CrI$_3$ under an external electric field along the $\hat{y}$ direction at Fermi level $\mu = 0.36$ eV. The arrow(color) on the sphere indicates the direction(magnitude) of the torkance under the given magnetization direction. We use $k_B T = 3$ meV, $\eta = 30$ meV in the calculations. $a_0$ is the Bohr radius.

the system out of the pure Néel space. Note that the Néel space state is the same as the $\mathbf{N}$-space state at the $z$ and $x$ axes.

CHEMICAL POTENTIAL DEPENDENCE OF STEADY STATES

In this section we include additional plots of final steady states at different Fermi levels summarized in Fig. 4. Both switching and oscillating behaviors can be observed at various chemical potentials and electric-field strengths. The chemical potential can be tuned by perpendicular gate voltage in principle and Fig. 4 indicates bilayer CrI$_3$ can have tunable functions by controlling both in-plane and out-of-plane fields.

FIG. 4. (Color online) Final steady state of $\hat{N}$ as a function of applied field with staring point at the $L_z = +1$ for various chemical potentials respectively. The spread in the $y$ coordinate indicates the oscillation amplitude, and the color of the spread represents the oscillation frequency.
[1] P. Giannozzi, O. Andreussi, T. Brumme, O. Bunau, M. B. Nardelli, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, M. Cococcioni, N. Colonna, I. Carnimeo, A. D. Corso, S. de Gironcoli, P. Delugas, R. A. DiStasio, A. Ferretti, A. Floris, G. Fratesi, G. Fugallo, R. Gebauer, U. Gerstmann, F. Giustino, T. Gorni, J. Jia, M. Kawamura, H.-Y. Ko, A. Kocolo, E. Küçükbilenli, M. Lazzeri, M. Marsili, N. Marzari, F. Mauri, N. L. Nguyen, H.-V. Nguyen, A. O. de-la Roza, L. Paulatto, S. Pons, D. Rocca, B. Santra, M. Schlipf, A. P. Seitsonen, A. Smogunov, I. Timrov, T. Thonhauser, P. Umari, N. Vast, X. Wu, and S. Baroni, Journal of Physics: Condensed Matter 29, 465901 (2017).
[2] X.-X. Zhang, L. Li, D. Weber, J. Goldberger, K. F. Mak, and J. Shan, Nature Materials 19, 838 (2020).
[3] A. D. Corso, Computational Materials Science 95, 337 (2014).
[4] G. Kresse and D. Joubert, Phys. Rev. B 59, 1758 (1999).
[5] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
[6] H. J. Monkhorst and J. D. Pack, Phys. Rev. B 13, 5188 (1976).
[7] N. Sivadas, S. Okamoto, X. Xu, C. J. Fennie, and D. Xiao, Nano Letters 18, 7658 (2018), pMID: 30408960, https://doi.org/10.1021/acs.nanolett.8b03321.
[8] A. A. Mostofi, J. R. Yates, G. Pizzi, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, Computer Physics Communications 185, 2309 (2014).
[9] D. Gresch, Q. Wu, G. W. Winkler, R. Häuselmann, M. Troyer, and A. A. Soluyanov, Phys. Rev. Materials 2, 103805 (2018).
[10] T. A. Tartaglia, J. N. Tang, J. L. Lado, F. Bahrami, M. Abramchuk, G. T. McCandless, M. C. Doyle, K. S. Burch, Y. Ran, J. Y. Chan, and F. Tafti, Science Advances 6, 10.1126/sciadv.abb9379 (2020), https://advances.sciencemag.org/content/6/30/abb9379.full.pdf.
[11] J. H. Ryoo, C.-H. Park, and I. Souza, Phys. Rev. B 99, 235113 (2019).
[12] M. D. Stiles and J. Miltat, Spin-transfer torque and dynamics, in Spin Dynamics in Confined Magnetic Structures III, edited by B. Hillebrands and A. Thiaville (Springer Berlin Heidelberg, Berlin, Heidelberg, 2006) pp. 225–308.
[13] H. V. Gomonay and V. M. Loktev, Phys. Rev. B 81, 144427 (2010).
[14] A. Manchon, Journal of Physics: Condensed Matter 29, 104002 (2017).