Magnon-phonon interactions open a gap at the Dirac point in the spin-wave spectra of CrI₃ 2D magnets

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Recent neutron-diffraction experiments in honeycomb CrI₃ 2D ferromagnets have evinced the existence of a gap at the Dirac point in their spin-wave spectra. The existence of this gap has been attributed to strong Dzyaloshinskii-Moriya or Kitaev (DM/K) interactions and suggested to set the stage for topologically protected edge states to sustain non-dissipative spin transport. We perform state-of-the-art simulations of the spin-wave spectra in this system, based on time-dependent density-functional perturbation theory and fully accounting for spin-orbit couplings (SOC) from which DM/K interactions ultimately stem. Our results are in fair overall agreement with experiments, but fail to account for the observed gap. The magnitude of the DM/K interaction parameters, determined from constrained density-functional theory (CDFT), is not consistent with observations either. Lattice-dynamical calculations, performed within density-functional perturbation theory (DFpT), indicate that a substantial degeneracy and a strong coupling between vibrational and magnetic excitations exist in this system, thus providing an alternative path to understanding the observed gap. In order to pursue this path, we introduce an interacting magnon-phonon Hamiltonian featuring a linear coupling between lattice and spin fluctuations, enabled by the magnetic anisotropy induced by SOC. Upon determination of the relevant interaction constants by DFpT and CDFT, this model allows us to propose magnon-phonon interactions as the most likely mechanism to explain the observed gap.

I. INTRODUCTION

The recent discovery of two-dimensional (2D) magnets [1, 2] has opened new paths in nanodevice fabrication and engineering, thanks to the high tunability of their magnetic state and the ease of their combination into functional heterostructures [3–5]. In these materials, the magnetic anisotropy induced by spin-orbit couplings (SOC) stabilizes the 2D long-range magnetic order against thermal fluctuations, by gapping the magnon Goldstone mode and thus dodging the conclusions of the Mermin-Wagner theorem [6, 7]. The van der Waals crystal CrI₃ was the first compound reported to display long-range magnetic order down to the monolayer limit, where it behaves as a ferromagnetic semiconductor with a Curie temperature of 45 K and a sizeable out-of-plane anisotropy [1]. The impressive variety of its magnetic response to the most diverse probes has stimulated an intense research effort aiming to better understand and exploit its low-dimensional magnetism. Indeed, spintronics applications are envisaged for bilayer-CrI₃, a layered antiferromagnet which can be gradually turned ferromagnetic by applied pressure [8, 9], electrical field [10, 11], or electrostatic doping [12], thus realizing an intrinsic spin filter tunnel junction with a reported high figure of merit [9, 13–15]. Moreover, the optical properties of ultrathin CrI₃ have been shown to be exceptionally sensitive to its magnetic state [16–18], stimulating new ideas for optoelectronic [19] and photovoltaic [20] devices.

Fascinating prospects ultimately arise from the potential of CrI₃ to sustain collective magnetic excitations, namely magnons. The insulating character of this material strongly reduces Landau damping, thus ensuring a magnon lifetime longer than that of any known 2D metal by one order of magnitude [21], with tremendous implications in the field of magnonics [22]. Even more interestingly, the honeycomb structure of the 2D Cr lattice is such that a system of Heisenberg magnets localized at the atomic sites would feature two spin-wave dispersions crossing at the K/K’ corners of the Brillouin zone (BZ), remnant of the Dirac points occurring in the electron energy bands in graphene. When subject to a suitable inversion symmetry-breaking perturbation, such a system would develop topologically protected edge spin waves able to sustain dissipation-less spin transport [23–26], in full analogy with fermionic topological states originally proposed to occur in graphene [27].

Indeed, inelastic neutron scattering (INS) has provided evidence of a gap of a few meV at the Dirac points in the spin-wave spectrum of thick CrI₃ samples [28]. While the existence of multiple magnon states at the center of the BZ has been confirmed by (magneto-) Raman spectroscopy [21, 29, 30] the nature and very existence of a gap at the zone border is still controversial. Early
suggestions quite naturally explained the occurrence of this gap in terms of SOC-enabled inversion-symmetry-breaking exchange interactions, such as second-nearest-neighbor Dzyaloshinskii-Moriya (DM) [28] or first-nearest-neighbor Kitaev (K) [31]. While this scenario has received support from itinerant-electron models based on tight-binding Hamiltonians [32], the robustness of these conclusions is as strong as the reliability of the semiempirical SOC and screened Coulomb repulsion parameters on which they are based. Moreover, the direct estimation from first-principles of the anisotropic exchange couplings has yielded quite diverse results according to the details of the downfolding methodology [33], with the more recent calculations pointing towards very weak [34], if not negligible [35], DM/K interactions, thus further questioning their role in the opening of the observed gap. Different mechanisms are therefore being explored, including e.g. correlation-induced asymmetries of the interlayer couplings, which however would not hold in the monolayer limit [36].

In this work we provide hefty evidence that the gap observed at the Dirac points in the spin-wave spectrum of CrI$_3$ is likely due to strong spin-lattice interactions affecting degenerate magnetic and vibrational excitations, resulting in a sort of polaritonic hybridization between magnon and phonon bands near, but not quite, at, the Dirac points. Our conclusions are robust with respect to dimensionality and do not subside down to the monolayer limit. Our first step is the evaluation of the INS magnetic cross section of monolayer CrI$_3$, performed at clamped nuclei by means of full-fledged time-dependent density-functional perturbation theory (TDDFpT) and wholly accounting for relativistic effects [37, 38]. Our approach, not relying on any adiabatic spin decoupling, avoids the intricacies of downfolding to an effective spin model and so includes the complexity of SOC-induced exchange couplings directly into the excitation spectra without the need of introducing any semi-empirical parameters. Our results reveal two dispersive magnon branches with quite different cross-section intensities, in fair agreement with INS data [28]. We clearly resolve the Goldstone gap at the zone center, the hallmark of magnetic anisotropy, but no gap is found at the Dirac points, corroborating the negligible role of DM/K interactions in spin-wave spectra of monolayer CrI$_3$ [35, 36]. Next, following recent indications that a strong spin-lattice coupling may exist in ultrathin CrI$_3$ [30, 39, 40], we determine the phonon dispersions in this system, using (static) density-functional perturbation theory (DFpT) [41], and reveal that a bundle of flat phonon bands intersects the acoustic magnon band just below the energy of the Dirac magnon. Building on this finding and leveraging constrained DFT (CDFT), we single out of the phonon bundle a flat branch with frequency $\hbar \omega \approx 14$ meV, in fair agreement with the position of the observed gap at $\hbar \omega \approx 12$ meV [28], and displaying a strong linear spin-lattice interaction. This coupling, which is enabled by SOC, can be captured by a generalized Heisenberg Hamiltonian whose eigenstates are spin-lattice polaritons featuring a gap where the free magnon and phonon bands cross. Along the $1K$ line, a gap $\Delta \approx 1.5$ meV is found to occur at $q_0 \approx 0.6q_K$. The magnitude of the gap depends on the strength of the spin-lattice coupling, whereas the shape and location of the locus of points in the BZ where it occurs depends on the details of the phonon and magnon dispersions. While thirty years of successful practice of DFpT give us confidence in the accuracy of our predictions for the phonon bands, density-functional theory, particularly when based on the local spin-density approximation (LSDA) or spin-polarized generalized-gradient approximation (GGA), is known to overestimate the magnitude of magnetic moments and exchange interactions, as recently reported also for bulk CrI$_3$ [36]. This expectation is confirmed by our overestimate of the energy of optical magnons with respect to experiment, whose mitigation would result in a shift of the gap towards the border of the BZ, as discussed in the following.

The paper is organized as follows. Section II summarizes the theoretical framework and presents the results of our TDDFpT calculations; in Sec. III our DFpT calculations of the phonon dispersion and CDFT calculations of the magnon-phonon coupling are reported; Sec. IV discusses the implications of the predicted spin-lattice polaritons on the magnon dispersion based on a generalized Heisenberg Hamiltonian. Finally, Sec. V presents our conclusive remarks.

II. INELASTIC NEUTRON-SCATTERING CROSS SECTIONS FROM TDDFPT

In INS experiments a neutron beam with wavevector $k_i$ and energy $E_i$ is inelastically scattered by the target sample to a final state characterized by the wavevector $k_f = k_i - q$ and energy $E_f = E_i - \hbar \omega$, where $\hbar q$ and $\hbar \omega$ are the momentum and energy transferred to the sample, respectively. In the first Born approximation [42, 43], the double-differential cross section corresponding to magnetic excitations of electrons can be written in the compact form as [44, 45]:

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{\hbar}{\pi} \left( \frac{g_n e}{2\hbar} \right)^2 \frac{k_f}{k_i} S(q, \omega),$$  \hspace{1cm} (1)$$

where

$$S(q, \omega) = -\text{Im} \text{Tr} \left[ P^+(q) \chi(q, q; \omega) \right].$$  \hspace{1cm} (2)$$

Here, $-e$ and $g_n \approx 3.826$ are the electron charge and neutron $g$-factor, respectively, $P^+(q)$ is the $3 \times 3$ matrix, $P_{\alpha\beta}^+(q) = \delta_{\alpha\beta} - q_\alpha q_\beta / q^2$ (with $\alpha, \beta = x, y, z$), which is a projector onto the plane perpendicular to the direction of $q$, and $\chi(q, q; \omega)$ is the $3 \times 3$ spin susceptibility matrix. The poles of $S(q, \omega)$ are fingerprints of the spin excitations of the system, both of the magnon and Stoner type, thus allowing one to characterize various magnetic
FIG. 1. Magnon dispersions along high-symmetry directions in the BZs computed using TDDFpT including SOC in a single-layer ferromagnetic CrI$_3$. The color code describes the intensities of peaks in the magnetic spectrum. The inset shows adjacent BZs and the high-symmetry directions in the $(h, k)$ plane and labels of high-symmetry points.

spectroscopies, either in the bulk, as probed by INS, or at surfaces, as probed by spin-polarized electron energy loss spectroscopy [46]. A fully ab initio determination of $S(q, \omega)$ requires the computation of the dynamical spin susceptibility by using, e.g., TDDFpT [38, 47–54]. In this work, we have computed the INS magnetic spectrum of a CrI$_3$ monolayer using the Liouville-Lanczos approach to TDDFpT of Ref. [38], as implemented within the LSDA in the plane-wave pseudopotential framework of the QUANTUM ESPRESSO™ suite of codes [55]. Further technical details of our computations are presented in Appendix A, we stress however that our implementation applies to a general non-collinear spin-polarized framework, essential in materials containing heavy elements, such as CrI$_3$, where SOC is expected to play an important role [33].

The spin-wave spectrum of CrI$_3$ monolayer thus obtained is reported in Fig. 1: two magnon branches, which can be interpreted as arising from two Heisenberg moments arranged in a honeycomb lattice, can be clearly distinguished. In particular, an intense acoustic band is present in the first BZ along the $\Gamma M$ direction, whereas the optical branch takes over when entering the second BZ. Our results are in fair agreement with the experiment of Ref. [28], the main discrepancies consisting in: i) an overestimation of the spin stiffness of the system, resulting in too broad a band width of $\approx 28$ meV (consistent with the linear-response DFT-based calculations of Ref. [36], reporting a value of $\approx 31$ meV in bulk CrI$_3$ without SOC), to be compared with an experimental value of $\approx 20$ meV [28, 56]; and, most importantly, ii) the failure to predict the existence of a gap at or in the vicinity of the Dirac ($K$) point, as observed experimentally. While too large a spin stiffness is a common feature of LSDA- and GGA-based models [36, 57, 58], we stress that our theoretical framework, being based on a fully-relativistic approach to (time-dependent) DFT, naturally accounts for the SOC that is eventually responsible for the DM/K interactions allegedly resulting in the observed gap. If anything, our treatment of relativistic effects results in too large a SOC, as witnessed by the too large value of the Goldstone gap opened by magnetic anisotropy (the acoustic magnon at the BZ center is predicted to have an energy of $\approx 1.5$ meV, to be compared with an experimental value of $\approx 0.3$ meV [56]).

We conclude that DM/K interactions at clamped nuclei are likely too small to account for the origin of the observed gap at the Dirac ($K$) point, as it was already suggested in previous theoretical studies [34, 35] and confirmed by our calculation of the relevant exchange interactions constants performed within constrained DFT and reported in Appendix A 1.

III. PHONON DISPERSIONS AND SPIN-LATTICE COUPLING

Having ruled out DM/K anisotropies as the cause of the gap opening in monolayer CrI$_3$, we now turn to examine an alternative mechanism, namely magnon-phonon coupling. Indeed, phonons often lie in the same energy range of spin waves, and when magnetic anisotropies are present, hybridization between these two excitations may
occur \[59, 60\]. Recently, it was pointed out that CrI$_3$ might fall into this category \[34\], this suggestion being supported by some earlier calculations of the phonon dispersion in monolayer CrI$_3$, displaying $i$ a sizeable peak in the vibrational density of states (vDOS) at $\approx 14$ meV, in close proximity to the position of the magnon gap observed at $\approx 12.5$ meV \[28\], and $ii$ a strong spin-lattice coupling \[39\].

In order to elaborate on this surmise, we have started by computing the phonon spectrum of CrI$_3$. For further reference, and in order to fix the notation, let us first state the lattice Hamiltonian in the harmonic approximation:

$$H_{ph} = \frac{1}{2} \sum_{ns} \frac{p_{ns}^2}{2M_s} + \frac{1}{2} \sum_{ns \neq mt} u_{ns} \cdot K_{st}(R_{nm}) \cdot u_{mt},$$  \hspace{1cm} (3)

where $n$ and $m$ enumerate the elementary cells of the crystal lattice, $R_{nm} = R_n - R_m$ is the distance between two such cells, $u_{ns}$ is the displacement of the $s$-th atom in the $n$-th unit cell, $p_{ns}$ and $M_s$ are its momentum and mass, respectively, and $K_{st}(R_{nm})$ is the matrix of the interatomic force constants, which we compute using DFpT \[41\]. Bold symbols indicate 3-vectors (Cartesian indices are suppressed and a dot “.” stands for a scalar product), whereas bold calligraphic symbols are $3 \times 3$ tensors. The vibrational normal modes are obtained from the eigenvalue equation (a tilde, “~”, on top of various quantities indicates their Fourier transforms, when needed):

$$\sum_t \tilde{K}_{st}(q) \cdot e^\mu_t(q) = M_s \Omega^2(q) e^\mu_s(q),$$ \hspace{1cm} (4)

where $q$ is the phonon wavevector, $\Omega^2(q)$ and $e^\mu(q)$ are the eigenvalues (frequencies) and eigenvectors of the $\mu$-th vibrational mode. The atomic displacements in the $\mu$-th normal mode are defined in terms of the normalized eigenvectors of Eq. (4) \[\sum_s M_s e^{\nu*}_s(q) \cdot e^\mu_s(q) = \delta_{\mu\nu}\] as:

$$u_{ns} = \frac{1}{N} \sum_{q\mu} e^{i q \cdot R_s} \xi^\mu(q) e^{\nu*}_s(q),$$ \hspace{1cm} (5)

where $\xi^\mu(q) = \sum_{ns} M_n e^{-i q \cdot R_n} u_{ns} \cdot e^{\nu*}_s(q)$ is the amplitude of the $\mu$-th normal mode, and $N$ the number of unit cells in the crystal. For future reference, we remark that the dimensions of $\xi$ are a length times the square root of a mass (say, Å × √AMU). We solved the eigenproblem of Eq. (4) within DFpT using LSDA, including SOC self-consistently. All the relevant technical details of the computations reported in this section are presented in Appendix A.

In Fig. 2 we display our computed phonon dispersions and vibrational density of states (vDOS) for the CrI$_3$ monolayer, and compare them with the INS magnetic spectrum from Ref. \[28\]. Our results confirm that a high vDOS exists in the energy range where a magnetic gap is observed, thus pointing at magnon-phonon interactions as a candidate mechanism that could drive its opening. In order to ascertain whether this can indeed be the case, we have computed the dependence of the crystal magnetization on the lattice distortion along the normal modes in the relevant energy range, using CDFT. We find that the vDOS peak is populated with vibrational modes strongly coupling with the magnetization, as reported in Fig. 8 of Appendix A.2. The most intense spin-lattice coupling
is found with the 16-th normal mode, whose energy is \( h\Omega_{16}(q_K) \approx 14 \text{ meV} \). Displacing the atoms along its eigenvectors \( u_{ns} \) at \( q = q_K \), shown in green in Figs. 3(a) and 3(b), induces a tilt of the crystal magnetization with respect to the easy-magnetization (\( z \)) axis according to the pattern presented in Fig. 3(c). In Fig. 3(d) we report the magnitude of such a tilt angle as a function of the amplitude of the lattice distortion. Not unexpectedly, no such dependence is detected when neglecting SOC, which is the origin of the magnetic anisotropy. When fully accounting for SOC, instead, a strong linear spin-lattice coupling is observed, further confirming that SOC-mediated spin-lattice couplings may be the culprit for the observed gap.

IV. MIXED SPIN-LATTICE EXCITATIONS

A. Spin-lattice Hamiltonian

In order to derive the minimal Hamiltonian accounting for a spin-lattice coupling, we consider the most general quadratic spin Hamiltonian, whose interaction parameters depend on the spin-spin distance:

\[
H_{sp} = -\frac{1}{2} \sum_{ns \neq mt} S_{ns} \cdot J_{st}(R_{nm}) \cdot S_{mt} - \sum_{ns} \left( \sum_{\nu} J_{ns}^{zz} \cdot D_{s} \cdot S_{ns} \right), \tag{6}
\]

where \( S_{ns} \) is a classical spin residing at the \( sn \) lattice site, \( J_{st}(R_{nm}) \) the exchange couplings and \( D_{s} \) the onsite magnetic anisotropy. The primed summations run on the magnetic sites only, in contrast to the summations in Eq. (3), which run over all the atomic positions. Both the exchange couplings and the onsite magnetic anisotropy depend implicitly on the atomic displacements and can be expanded in powers of \( u_{ns} \). The zeroth order (\( u_{ns} = 0 \)) corresponds to the equilibrium case, where our CDFT calculations have shown that only the isotropic exchange \( J_{st}(R_{nm}) = \text{Tr} \left[ J_{st}(R_{nm}) \right] / 3 \) and the onsite anisotropy \( D_{s}^{zz} \) are relevant (see Table III in Appendix A1). In this limit, the vibrational and magnetic normal modes are decoupled, the latter being solutions of the secular equation:

\[
S \sum_{t} \left[ \delta_{st} I_s - \tilde{J}_{st}(q) \right] f^\dagger_t(q) = \omega_{\nu}(q) f^\dagger_s(q), \tag{7}
\]

where \( I_s = 2D_{s}^{zz} + \sum_{mt} J_{st}(R_{0m}) \), and \( S = \vert S_{ns} \vert \). The eigenvectors are normalized according to \( \sum_{t} f^{\sigma \nu}_t(q) f^{\nu \sigma}_s(q) = \delta_{\sigma \nu} \) and are directly related to the spin components in the \( xy \) plane via

\[
S_{ns}^+ = \frac{1}{N} \sum_{q\nu} e^{iq \cdot R_{ns}} \eta^\nu(q) f_s^{\nu \dagger}(q), \tag{8}
\]

with \( S_{ns}^+ = S_{ns}^x \pm i S_{ns}^y \), and \( \eta^\nu(q) \) is the amplitude of the \( \nu \)-th normal mode.

To lowest-order in \( u_{ns} \), the corrections to the Hamiltonian in Eq. (6) result in a linear coupling between the spin and lattice degrees of freedom (see Appendix B for a derivation)

\[
H_{sl} = -\sum_{q \mu \nu} \lambda_{\mu \nu}(q) \sqrt{\omega_{\nu}(q)} \xi_{\mu}(q) \eta^\nu(q) + \text{c.c.}, \tag{9}
\]

where \( \sqrt{\omega_{\nu}(q)} \) has been introduced so that \( \lambda_{\mu \nu}(q) \) has the dimension of a frequency.

The exchange interactions in Eq. (7), \( \tilde{J}_{st}(q) \), as well as the \( \lambda_{\mu \nu}(q) \) coupling constants in Eq. (9) corresponding to different phonon-magnon pairs, \( (\mu, \nu) \), can be estimated using CDFT, as explained in Appendix A2. In Fig. 4 we report the magnitude of the couplings between the acoustic magnon and the phonons in the energy range from 8 to 15 meV, thus estimated at the Dirac point, \( q = q_K \), in monolayer CrI\(_3\), using CDFT supercell calculations. The coupling constant with the optical magnon branch \( (\nu = 2) \) has been found to have the same absolute value as the one with the acoustic branch for all the phonon modes considered (see Appendix A2).

![FIG. 4. Absolute value of the magnon-phonon coupling constants, \( \lambda_{\mu \nu}(q) \), for different phonon modes (\( \mu = 9, \ldots, 16 \)) and for the acoustic magnon (\( \nu = 1 \)), computed at the Dirac point, \( q = q_K \), in monolayer CrI\(_3\), using CDFT supercell calculations. The coupling constant with the optical magnon branch (\( \nu = 2 \)) has been found to have the same absolute value as the one with the acoustic branch for all the phonon modes considered (see Appendix A2).](image)

B. Spin-lattice polaritons

The normal modes of Eq. (7) yield the frequencies and polarization patterns of the free spin waves in CrI\(_3\), \( i.e. \) of the magnetic excitations resulting from the neglect of the spin-lattice interactions embodied in Eq. (9). In order to quantify the impact of these on the spin-wave
where $\hat{a}_{\mu q}$ and $\hat{b}_{\nu q}$ are the annihilation operators of phonons and magnons, respectively, and $\hat{\lambda}_{\mu \nu}(q) = \lambda_{\mu \nu}(q) \sqrt{2\omega_{\nu}(q)}/\Omega_{\mu}(q)$. The phonon frequencies $\Omega_{\mu}(q)$ are interpolated from our DFT calculations, whereas the magnon frequencies $\omega_{\nu}(q)$ are derived from our spin model fitted to the TDDFpT dispersions. The $\lambda_{\mu \nu}(q)$ couplings are estimated at the Dirac point as detailed in the previous section and considered constant throughout the BZ, since the BZ portions in which the magnon and phonon branches cross in the proximity of the Dirac points. In the following we neglect the terms $\hat{a}_{\mu q}^\dagger \hat{b}_{\nu q}$ that do not conserve the number of quasiparticles and are thus expected to yield higher-order corrections with respect to the number-conserving terms.

The eigenmodes of the Hamiltonian in Eq. (11) are mixed magnon-phonon quasiparticles. As their nature is remnant of the mixed photon-photon or exciton-photon modes commonly known as polaritons, we dub them spin-lattice polaritons. In Fig. 5 we report the energy dispersions of spin-lattice polaritons obtained by diagonalizing the Hamiltonian in Eq. (11) in the number-conserving approximation. We consider two different sets of free-magnon frequencies, $\omega_{\nu}(q)$: in Figs. 5(a) and (b) the one obtained by using the exchange parameters fitted to our TDDFpT results, in Figs. 5(c) and (d) the one obtained by using the exchange parameters fitted to the INS data of Ref. [28]. The main polaritonic effect is due to the 13-th to 16-th phonon modes, which open a gap of roughly 1.5 meV, but lower-energy hybridization of smaller intensity are present as well. The net outcome is the suppression of the magnon character in an energy region of about 4 meV, which implies a concomitant suppression of the intensity in INS experiments; this compares well with the experimental width of the gap, of about 4 meV [28]. We remark that the exact location of this region in the BZ depends on the spin stiffness, which density functionals presently available are not able to predict with sufficient accuracy [36]. However, a renormalization of such stiffness to experimental data brings our theoretical prediction of both the energy and the location in the BZ at the K point of the gap in remarkable agreement with experiments, as illustrated in Figs. 5(c) and (d), and in the three dimensional plot of Fig. 6.

V. CONCLUSIONS

Our results show that SOC-induced magnetic anisotropies do not open a gap in the spin-wave spectrum of monolayer-CrI$_3$ at clamped nuclei. However, they are responsible for a sizeable magnon-phonon coupling, estimated to open hybridization gaps in a region of about 4 meV around the Dirac point. In the bulk case, the DM/Kitaev interactions have been shown to be too small to account for the observed gap [34], whereas strong electronic correlations have been reported to simply shift the Dirac point out of the high-symmetry lines [36], a scenario which has not been confirmed by the
constant-energy cut analysis of INS measurements [28]. Based on these premises, and considering that CrI₃ is a van der Waals crystal with feeble inter-layer interaction, we deem therefore magnon-phonon hybridization to be the most likely mechanism to explain the gap observed by INS measurements.

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Appendix A: Computational details

All the calculations were performed using the QUANTUM ESPRESSO™ distribution [55], a plane-wave+ pseudopotential suite of computer codes, using the LSDA exchange-correlation functional and including SOC self-consistently. We have used the norm-conserving pseudopotentials (PPs) from the PseudoDojo library [61] and by generating PPs with the atomic code using the configurations from v0.3.1 of the Pslibrary [62].

For the ground-state calculations, the Kohn-Sham wavefunctions and potentials are expanded in plane waves up to a kinetic-energy cutoff of 80 Ry and 320 Ry, respectively. Brillouin zone was sampled using a uni-

form Γ-centered 8 × 8 × 1 k points mesh for the hexagonal unit cell; uniform meshes of the same density have been adopted for calculations with supercells. The CrI₃ 2D crystal structure was obtained by extracting one layer from the trigonal bulk structure and by optimizing atomic positions and the in-plane lattice constant. The optimized in-plane lattice constant is 12.98 Bohr. The DFT calculations correctly yield a ferromagnetic (FM) ordering and a magnetic anisotropy with the out-of-plane directions as the easy axis.

Phonon dispersions were obtained using DFpT by computing the dielectric response, the Born effective charges, and the dynamical matrices on a Γ-centered 4 × 4 × 1 q points mesh; these results were then used to compute the matrix of interatomic force constants (IFC) in real space; and this latter has then been used to compute phonon energies and displacements at arbitrary wavevectors. The electrostatic long-range part of the IFC was computed by taking into account the artifacts produced by the nonphysical periodicity in the out-of-plane direction [63].

All the calculations of the magnon dispersions have been performed using the soon-to-be-released turboMagnon component of the QUANTUM ESPRESSO™ suite of codes [55], implementing the Liouville-Lanczos approach to TDDFpT in the adiabatic LSDA and including SOC self-consistently. This approach does not require the computation of unoccupied KS states and allows one to evaluate the spin susceptibility matrix in Eq. (2), χ(q,q;ω), using three Lanczos chains per wavenumber (one per Cartesian component of the perturbing magnetic field), encompassing the entire spectrum (i.e., independently of the frequency). With respect to what is detailed in Ref. [38], the algorithm has been upgraded by implementing the pseudo-hermitian symmetry along the lines of Ref. [64], yielding converged spectra with ∼ 15000 Lanczos steps, each one of which has roughly the cost of two Hamiltonian builds in a conventional static DFpT calculation. A Lorentzian smearing function with a broadening parameter of 1.4 meV has been used in the post-processing calculations. For the TDDFpT calculations, the kinetic energy cutoff is set to 60 and 240 Ry for the wavefunctions and potentials, respectively. The same k points mesh as for ground-state calculations has been used.

1. Exchange interactions

The estimate of the exchange interaction parameters of the Cr localized spins has been done using the total energy differences between the FM ground state and three antiferromagnetic (AF) patterns that can be contained in the supercell in Fig. 7. The four different magnetic arrangements are reported in Table II together with the the values of the Cr local spins.

To estimate the exchange parameters from the total energy differences we need to express the total energy
of each magnetic configuration as a function of the local spins. We start from the Heisenberg Hamiltonian, Eq. (6), and write the total energy per cell as:

$$E_{tot} = E_0 - \frac{1}{2} \sum_{i,j}^{\text{cell}} \sum_{j \neq i}^{\text{all}} J_{ij} S_i S_j,$$

(A1)

where $E_0$ is an adjustable energy term, independent from the spin interaction, $J_{ij}$ are the exchange interaction parameters between spins $S_i$ and $S_j$ of sites $i$ and $j$, respectively. $\sum_{\text{cell}}$ indicates a sum over the spins contained in one unit cell, and $\sum_{\text{all}}$ may in principle range infinitely over the lattice. Assuming that the exchange interactions vanish over the third neighbours, we replace the coupling constants $J_{ij}$ with $J_1$, $J_2$, and $J_3$ for the couplings between first, second, and third nearest neighbours, respectively.

The free parameters in Eq. (A1) now reduce to 4: $E_0$, $J_1$, $J_2$, $J_3$ to be fitted with the energy differences. Using the neighbours list in Table I and the spin and energy values of Table II we write the following linear system:

$$2E_0 - 27J_1 - 54J_2 - 27J_3 = 2E_{FM},$$
$$2E_0 + 27J_1 - 54J_2 + 27J_3 = 2E_{AFM},$$
$$2E_0 + 9J_1 + 18J_2 - 27J_3 = 2E_{AF2Y},$$
$$2E_0 - 9J_1 + 18J_2 + 27J_3 = 2E_{AF2Y}.$$  

(A2)

The resulting parameters, reported in Table III, are in close agreement with those obtained by, e.g. Besbes et al. [65] who used the magnetic force theorem. Together with the value obtained for the onsite magnetic anisotropy $D_s^{zz}$ of 0.30 meV, these exchange parameters reproduce closely the magnon dispersion of Fig. 1 in the LSDA (with SOC) case.
tor \(-q_K\) on the other the constrained energies remain unchanged when the magnetic configuration is inverted either by switching sublattices or by inverting the sign of the amplitude \(\theta\).

2. Spin-lattice coupling constants

The magnon-phonon coupling coefficient \(\lambda_{\mu\nu}(q)\) can be computed by means of supercell calculations, by distorting the lattice along a given phonon mode \(\mu q\):

\[
u_{ns} = \xi^\mu(q) \left[ e^\mu(q)e^{iqR_n} + e^\mu(-q)e^{-iqR_n} \right],
\]

and then by projecting the magnetization pattern on the spin-wave normal modes at the lattice distortion includes Fourier components of \(\pm q\), as shown in Eq. (A3). The resulting magnon-phonon coefficients \(|\lambda_{\mu\nu}(q = q_K)|\) are reported in Fig. 4, showing a stark increase in the coupling intensity roughly in correspondence of the increase in the phonon vDOS, pointing towards a relevant effect of the magnon-phonon coupling in proximity of the Dirac point.

Appendix B: Derivation of the spin-lattice Hamiltonian

In this appendix we present a derivation of the coupling term of Eq. (9) in our spin-lattice Hamiltonian. Starting from Eq. (6)

\[
H_{sp} = -\frac{1}{2} \sum'_{i \neq j} S_i \cdot J_{ij} \cdot S_j - \sum'_{i} S_i \cdot D_i \cdot S_i,
\]

where \(i \equiv ns\) and \(j \equiv mt\), we perform an expansion around the ferromagnetic state with the magnetization polarized along \(z\):

\[
S^z_i \approx S - \frac{1}{2S} S^+_i S^-_i.
\]

Recalling that \(S^\pm_i = S^z_i \pm iS^y_i\), one obtains

\[
H_{sp} = E_{FM} - S \sum_i' \left( a_i S^+_i + c.c. \right) - \sum_i' \left( b_i S^+_i S^+_i + c.c. \right) - \frac{1}{2} \sum_i' \left( d_i S^+_i S^+_i + c.c. \right) - \frac{1}{2} \sum_{ij} \left( e_{ij} S^+_i S^-_j + c.c. \right) + \mathcal{O}\left( (S^+_i S^-_j)^2 \right),
\]

(B3)
with \( E_{FM} = -S^2 \sum_i \left( D_{ij}^{zz} + \frac{1}{2} \sum_j' \mathcal{J}_{ij}^{zz} \right) \) being the energy of the ferromagnetic state and

\[
a_i = D_{ij}^{zz} - i D_{ij}^{yz} + \frac{1}{2} \sum_j' \left( \mathcal{J}_{ij}^{zz} - i \mathcal{J}_{ij}^{yz} \right),
\]

\[
b_i = \frac{1}{4} \left( D_{ij}^{xx} - D_{ij}^{yy} \right) - \frac{i}{2} D_{ij}^{xy},
\]

\[
c_i = \frac{1}{2} \left( D_{ij}^{xx} + D_{ij}^{yy} \right) - D_{ij}^{zz} - \frac{1}{2} \sum_j' \mathcal{J}_{ij},
\]

\[
d_{ij} = \frac{1}{4} \left( \mathcal{J}_{ij}^{xx} - \mathcal{J}_{ij}^{yy} \right) - \frac{i}{4} \left( \mathcal{J}_{ij}^{xy} + \mathcal{J}_{ij}^{yx} \right),
\]

\[
e_{ij} = \frac{1}{4} \left( \mathcal{J}_{ij}^{xx} + \mathcal{J}_{ij}^{yy} \right) + \frac{i}{4} \left( \mathcal{J}_{ij}^{xy} - \mathcal{J}_{ij}^{yx} \right).
\]

From Eq. (B3) one can see that an anisotropy-induced linear coupling between the spin and lattice degrees of freedom emerges from variations of the \( a_i \) coefficient with respect to the lattice displacements. Coming back to an explicit lattice notation

\[
a_{ns} \approx a_{ns} \left| \frac{\partial a_{ns}}{\partial R_{mt}} \right| \cdot u_{mt} = \sum_{mt} a^\prime_{st} (R_{mn} + \tau_t) \cdot u_{mt},
\]

with \( \tau_t \) denoting the position of the \( t \)-th spin inside a unit cell. In the second line we imposed translational invariance and assumed that \( a_{ns} = 0 \). Substituting this last term back into Eq. (B3) and expanding \( u_{mt} \) and \( S_{ns}^\dagger \) on their normal modes according to Eqs. (5) and (8), one obtains the spin-lattice term of Eq. (9), with:

\[
\lambda_{\mu\nu}(q) \sqrt{\omega_\nu(q)} \equiv \sum_s \sum_t f_s^{\mu*}(q) a^\prime_{st}(q) \cdot e^*_i(q).
\]

FIG. 8. Average changes in the angle \( \theta \) (in degrees) between the \( z \) axis and the spin momentum on Cr atoms, which are induced due to the displacement of Cr atoms along the \( \mu \)-th normal phonon mode \( (\mu = 7, \ldots, 18) \) with the amplitude \( \xi_{\mu}(q_K) \) [see Eq. (5)], and with and without SOC. On each panel, the phonon frequency of the corresponding \( \mu \)-th phonon mode at the Dirac point \( \Omega_{\mu}(q_K) \) in meV] is indicated. The abscissa of all panels is the normalized phonon coordinate, \( \xi_{\mu}(q_K) \) [see Eq. (5)], in units Å \( \times \) √AMU.

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