The surprising effect of electron correlations on exchange interactions and spin excitations in magnetic 2D van der Waals materials

Li Qin Ke\textsuperscript{1} and Mikhail I. Katsnelson\textsuperscript{2}
\textsuperscript{1} Ames Laboratory, U.S. Department of Energy, Ames, Iowa 50011, USA
\textsuperscript{2} Institute for Molecules and Materials, Radboud University, Heijendaalseweg 135, 6525 AJ Nijmegen, The Netherlands
(Dated: July 30, 2020)

Despite serious effort, the nature of the magnetic interactions and role of electron-correlation effects in magnetic two-dimensional (2D) van der Waals materials, remains elusive. Using CrI\textsubscript{3} as a model system, we show that the calculated electronic structure including nonlocal electron correlations yields spin excitations consistent with inelastic neutron scattering measurements. Remarkably, this approach identifies a novel correlation-enhanced interlayer super-supersuperexchange, which rotates the magnon Dirac lines off, and introduces a gap along, the high-symmetry Γ-K-M path. Our discovery provides a different perspective on the gap opening mechanism observed in CrI\textsubscript{3}, which was previously associated with spin-orbit coupling through the Dzyaloshinskii-Moriya interaction or Kitaev interaction. Our observation elucidates new concepts to describe spin ordering and spin dynamics in magnetic van der Waals materials and demonstrates the necessity of explicit treatment of electron correlation in the broad family of 2D magnetic materials.

The intimate interplay between correlated electrons, lattice, and magnetism can result in a rich variety of interesting and important physical phenomena \[1\]. In two-dimensional (2D) materials with open d- or f-shells, additional quantum confinement caused by the reduced dimensionality suppresses the screening \[2, 3\] thus may further enhance the electron correlation. The recent advancement of magnetic 2D van der Waals (vdW) materials adds a new magnetic functionality to the already vast appeal of the 2D materials family \[4\]–\[7\]. Their magnetism is very sensitive to, and can be controlled by, pressure \[8\], stacking arrangement \[9\], and external magnetic \[10\] and electric \[10,12\] fields. Such unprecedented tunability offers novel opportunities to design and construct dramatically new energy-efficient spin-based devices. Even in their bulk form, the reduced coordination number in quasi 2D lattices constrains the electron hopping, thereby increasing the role of the Coulomb interaction. It is pivotal to explicitly address how the electron correlations affect the magnetic interactions and reveal how the excitations in these confined systems might be understood, controlled and exploited.

Despite considerable attention, accurate \textit{ab initio} descriptions of spin excitations and a comprehensive understanding of magnetic interactions in magnetic 2D vdW materials (m2Dv) are still lacking. Some complexity is imparted by the role of spin-orbit coupling. For example, the presence of the Dzyaloshinskii-Moriya interaction (DMI), which is essential for materials to host topological magnons and other topological objects such as skyrmions \[13,14\], remains puzzling. Specifically, 2D honeycomb ferromagnets can be viewed as the magnetic analog of graphene \[15,16\]. Earlier theoretical works \[17,18\] demonstrated that introducing a next-nearest-neighbor DMI interaction, which breaks the inversion symmetry of 2D honeycomb lattice, can induce a spin gap at the Dirac points and realize the topological magnons. Recent inelastic neutron scattering (INS) experiments have shown a spinwave (SW) gap opening along the high-symmetry lines in pristine CrI\textsubscript{3} \[16\], implying a sizable DMI or Kitaev interaction \[19\] may indeed exist in CrI\textsubscript{3}. In contrast, \textit{ab initio} investigations show that DMI \[20,22\] or Kitaev interaction \[22\] is negligible in pristine CrI\textsubscript{3}. To help elucidate their magnetic nature, \textit{ab initio} investigations of magnetic excitations in these materials should address the electron-correlation effects beyond density functional theory (DFT). Besides the nonlocal on-site correlation, the off-site correlation could also be critical as it directly affects the relative positions of cation-3d and anion-p bands and thus the (super-)supersuperexchange coupling \[23\].

In this work, using the most studied m2Dv—CrI\textsubscript{3}—as a prototype, we identify the role of electron correlations on the magnetic interactions and excitations in m2Dv. The central quantity that characterizes the spin excitations—the dynamic transverse spin susceptibility (DTSS)—is calculated and directly compared with the SW spectra measured by INS \[16,24\]. We demonstrate that the explicit treatment of electron correlations is required to accurately describe the magnetic interactions, especially the interlayer interactions, in m2Dv. Furthermore, we made the remarkable discovery that a sizable magnon gap opens along the high-symmetry line in CrI\textsubscript{3} even without DMI. Instead, this gap is caused by a correlation-enhanced interlayer super-supersuperexchange coupling.

\textbf{Methods.}—Starting from a self-consistent \textit{ab initio} band structure, we first calculate the bare transverse spin susceptibility $\chi_0(r,r',\mathbf{q},\omega)$ using a linear response method \[25,26\]. Then the full transverse susceptibility $\chi$ is calculated, within the random phase approximation (RPA), as $\chi = \chi_0 + \chi_0 I \chi$, where $I$ is the exchange-correlation kernel. Two-particle quantities $\chi_0, \chi,$ and

\textsuperscript{*} Corresponding author: liqinke@ameslab.gov
I are functions of coordinates r and r’ within the unit cell. Since magnetic moments and excitations are nearly completely confined within the Cr sites, we project χ₀ onto the local spin densities of Cr pairs. This projection discretizes χ₀(r, r’; q, ω) into a matrix χ₀(i, j; q, ω), where i and j index the Cr sites in the unit cell. Such discretization allows us to 1) determine the kernel I using a sum rule [25]; 2) map χ⁻¹ into the Heisenberg model to extract pair exchange parameters; and 3) greatly reduce the computational effort. As for the electronic structures, we employ the quasiparticle self-consistent GW (QSGW) method [28, 29], wherein nonlocal electron correlations, both on-site and long-range off-site, are explicitly calculated. The widely used DFT+U methods [30], which provide a simplistic on-site correlation correction, are also employed for comparison. Further details of QSGW method and implementation [28, 29] and applications on CrI₃ [23] can be found in the Supplemental Material [33].

No DTSS calculations have yet been reported for CrI₃, or any other m2Dv. By far, except for a few studies [22, 31, 32] using the magnetic force theorem (MFT) [36], most of the theoretical investigations of the magnetic interactions in m2Dv are based on the total-energy-mapping method, often employed with DFT+U. However, the accuracy and applicability of such an approach in CrI₃ are not clear. Specifically, using the energy-mapping method, DFT overestimates the exchange couplings by 50% [16, 67]; the additional Cr site correlations further increase coupling, only worsening the agreement with experiments. In principle, the MFT approach is more suitable to describe the small spin deviation from the ground state, such as the SW excitations; moreover, it also allows one to resolve coupling into orbital contributions and elucidate the underlying exchange mechanism [35]. However, the resulted values still vary and are inconclusive; even the opposite trend of exchange-coupling dependence on on-site Cr correlations have been obtained [22]. The discrepancy likely lies in the details of the constructions of TB Hamiltonian and Greens function [38], which are often assisted by WANNIER90 technique [39]. On the other hand, DTSS is challenging to compute in practice. Studies to date have been mostly limited to simple systems, likely due to its computationally-demanding nature and other complications. For example, the evaluation of kernel I is not explicit in the case beyond the mean-field scheme, and the Goldstone theorem, which ensures at q = 0 and ω=0, is often not guaranteed. In this work, we first calculate DTSS on a product basis [29, 40] without resorting to the WANNIER90 techniques. We use the sum rule to calculate kernel I and ensure the Goldstone magnon mode at q = 0 and ω=0. Finally, in this work, to study the detailed SW dispersion near the Dirac point, high-resolution SW spectra are needed. This is achieved by calculating the real-space bare susceptibility χ₀(R, ω) on a R-mesh first and then obtain the χ₀(q, ω) along the high-symmetry path by Fast Fourier transform. Overall, the application of the current approach to these materials is not only novel but also necessary, as we will demonstrate later.

CrI₃ crystallizes in either the low-temperature rhombohedral structure (R-CrI₃) or high-temperature monoclinic structure (M-CrI₃) [41]. A honeycomb Cr monolayer is sandwiched between two I layers; then, the blocks of I-Cr-I triple layers are stacked along the z direction, held together by a weak vdW force. M-CrI₃ has a slightly distorted honeycomb Cr lattice and, more importantly, a different stacking arrangement, resulting in the A-type antiferromagnetic (AFM) ordering [9, 42, 44]. The sensitivity of interlayer Cr coupling on stacking arrangement reflects the changes of superexchange pathways across the vdW gap. A thorough understanding requires an explicit treatment of interlayer exchanges, instead of using a single effective interlayer exchange parameter as is often employed to describe the system. Thus, the exchange couplings and SWs in R-CrI₃ need to be considered in the context of rhombohedral symmetry. In this work, we first focus on the ferromagnetic (FM) R-CrI₃ and then discuss stacking effects on magnetism using M-CrI₃.

Figure 1 shows the crystal structure of R-CrI₃ and the corresponding Brillouin zone (BZ). The 2D honeycomb Cr sublattice and corresponding BZ are also shown for better comparison with previous studies, in which the SWs are often discussed in the hexagonal notation. The
FIG. 2. Spin excitations calculated from \(\text{Im}[\chi_0(q, \omega)]\) and \(\text{Im}[\chi(q, \omega)]\) in R-CrI\(_3\). The special \(q\) points along the high symmetry path \(Z-\Gamma-M_2|M_3-\Gamma-L\) are denoted in Figs. 1(c) and 1(d). (a) \(\text{Im}[\chi_0(q, \omega)]\) calculated in DFT. (b) The density of states calculated in DFT. The horizontal dashed-and-dotted line indicates the top of valence band. (c)–(f) \(\text{Im}[\chi(q, \omega)]\) calculated in DFT (c), DFT+\(U\) (d), QSGW (e), and QSGW+\(U\) (f). The intensity of \(\text{Im}[\chi(q, \omega)]\) is shown in log scale. Experimental SW energies, adopted from INS work by Chen et al. [16, 24], are denoted by open circles.

rhombohedral primitive unit cell contains two formula units (f.u.), while the conventional hexagonal cell includes six. Correspondingly, the former has a larger BZ than the latter. As shown in Fig. 1(d), along the \(k_x\) direction, its BZ boundary \(M_2\) is three times further from \(\Gamma\) than that of the hexagonal structure, \(\Gamma-M_1\). Along the \(k_y\) direction, \(M_2\) (equivalent to \(M_3\)) is at the BZ boundary for both rhombohedral and hexagonal cells.

First, we calculate within DFT the bare and full transverse susceptibilities, \(\chi_0(q, \omega)\) and \(\chi(q, \omega)\), which characterize the single-particle Stoner excitations and collective SW excitations, respectively. The intensities of \(\text{Im}[\chi_0(q, \omega)]\) and \(\text{Im}[\chi(q, \omega)]\) along high-symmetry paths are shown in Fig. 2(a) and 2(c), respectively. The energy scale of the Stoner excitations, starting at \(\sim 1.7\) eV and peaking at \(\sim 3\) eV, are two orders of magnitude larger than SW excitations, leaving the latter with negligible damping. This means that we can safely use the physical picture of well-defined local magnetic moments on Cr atoms and map the low-lying spin dynamics onto a purely localized-spin Hamiltonian as will be described in detail below. The threshold energy of \(\sim 1.7\) eV corresponds to the gap size of the spin-flip transition from the top of majority-spin Cr states to the bottom of minority-spin Cr states, as shown in Fig. 2(b), while the peak energy corresponds to the spin splitting of the Cr-\(d\) states. The SW energies, defined by the peaks of \(\text{Im}[\chi(q, \omega)]\), are solely determined by the RPA poles of \(1-I\chi_0 = 0\). With two Cr atoms in the primitive cell, we find two poles for each \(q\), resulting in two magnon branches, as shown in Figs. 2(c)–2(f). Experimental SW energies extracted from previous INS work by Chen et al. [16, 24] are plotted to compare with the calculated SW spectra.

We now compare the DFT SW spectra with INS data. Along the \(\Gamma-M_2\) path, Fig. 2(c) shows that two SW branches cross near the point \(K\), before reaching the BZ boundary \(M_2\). For other directions, a gap exists between two magnon branches. This is consistent with previous studies [16] without considering DMI. Along the \(\Gamma-M_3\) path, the SW minimum occurs at \([3 0 0]\) (hexagonal notation), instead of \([1 0 0]\) (\(\Gamma_1\)-point), reflecting the symmetry of the rhombohedral structure. The maximum energy of the optical mode measured in INS is about \(20\) meV, while DFT gives \(30\) meV and overestimates it by \(50\%\). As shown in Fig. 2(c), DFT also overestimates the acoustic magnon energies, most severely (by a factor of \(\sim 4\)) along the confined \(z\) direction. The overestimation of interlayer coupling also affects the in-plane SW energies, because nearly all interlayer couplings have in-plane components in their connecting vectors. Hence, in bulk materials, an accurate description of interlayer exchanges is essential to describe the in-plane SW accurately.

Next, we investigate the effects of electronic correlations on magnetic interactions within DFT+\(U\) and QSGW. We found that increasing the \(U\) value increases the on-site Cr moment and lowers the energies of both magnon branches. Figure 2(d) shows the SW spectra for \(U = 3\) eV, a typical value used for CrI\(_3\). The SW energy at the \(Z\) point is decreased to \(5.8\) meV, about \(2/3\) of the DFT value, however, it is still about a fac-
The optical magnon is centered at around 24 meV, similar as in DFT+U, but recovers some dispersion and agrees better with INS experiments. Interestingly, the acoustic SW energies are reduced in comparison to DFT+U. The energy-mapping method gives the opposite trend of the linear response and SW energy on U, so DFT+U not only reduces the optical SW energies but also improves the description of spin excitations in these systems. Considering GW methods may underestimate the on-site correlations, they have been applied on top of DFT+U for various systems [45]. Here, we also apply QSGW on top of DFT+U (QSGW+U) [46] to roughly mimic the additional on-site correlations. Figure 2(c) shows the SW spectra calculated with $U = 1.36$ eV. Although the in-plane acoustic SW is still somewhat overestimated, the overall spectra compare well with experiments, suggesting that additional on-site correlations beyond QSGW may be needed to best describe electronic structures and SW in CrI$_3$. More rigorous and comprehensive frameworks, such as the dynamical mean-field theory (DFMT)+GW [47], can be valuable for future research.

To develop a quantitative understanding of how electron correlations affect magnetic interactions and excitations, we calculate the effective pair exchange parameters $J_{ij}$ for a Heisenberg model $H = -\sum_{i\neq j} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j$, where $\mathbf{e}_i$ is the unit vector of the atomic spin moment at site $i$. The $J_{ij}$ parameters are obtained from the inverse of susceptibility matrix, $[\chi(q, \omega = 0)]^{-1}$, with a subsequent Fourier transform [25, 48–50]. As was shown in [49] the exchange parameters determined in this way coincide with those calculated via magnetic force theorem. The exchange parameters between the first few neighbors, as depicted in Fig. 3(a), are listed in Table I. Exchange couplings beyond 1 Å are negligible. Using the linear SW theory, we recalculate the SW spectra with the extracted $J_{ij}$ parameters. Results agree well with those determined by the peaks of $\text{Im}[\chi(q, \omega)]$ (See Supplemental Material [33] for details).

We find that on-site correlations between Cr-$d$ states included in DFT+U have a stronger effect on decreasing the nearest-neighbor coupling, while the explicit non-local correlations in QSGW have a more substantial effect on the longer-range interlayer couplings. Within DFT, the calculated in-plane exchanges are similar to values [37] derived from total energy mapping and $\sim 50\%$ larger than those extracted from INS [16]. In comparison to DFT, DFT+U decreases $J_1$ by $\sim 19\%$ and the overall interlayer (FM) couplings $\sum J_2 \sim 27\%$, while QSGW decreases $J_1$ by $\sim 14\%$ and $\sum J_2 \sim 60\%$. Thus, QSGW not only reduces the optical SW energies but also significantly lowers the acoustic ones, especially along the interlayer direction. Overall, the long-range correlations in QSGW are important to describe the interlayer (super-)superexchanges in CrI$_3$.

Interestingly, regarding the dependence of exchange and SW energy on $U$ parameter, the energy-mapping method gives the opposite trend of the linear response method. Using the energy-mapping method, exchange
and SW energy increase with $U$, worsening the agreement between theory and INS measurements. This suggests that non-Heisenberg interactions, such as biquadratic exchange or multi-site exchange interactions, are essential in this system, especially when additional on-site correlations are taken into account. By analogy with transition-metal oxide systems, one can assume that the induced magnetic polarization on iodine ligands can play a decisive role in this non-Heisenberg behavior. Contrary to the total energy differences, the linear response method describes accurately the small spin deviations from the given (ground) state and thus the spinwave spectra. The corresponding exchange integrals thus depends on the initial equilibrium spin configuration in which they are calculated.

Remarkably, besides the improvement of SW energies, correlations beyond DFT also open up a gap along the $\Gamma-K-M$ path in both DFT+$U$ and QSGW, as shown in Figs. 3(d) and 4(f). Although the calculated gap size ($\sim 1.8$ meV in QSGW+$U$) is smaller than the experimental value of $\sim 4$ meV observed in INS, the existence of such a gap is unexpected in the absence of DMI. Indeed, the gap opening at Dirac $K$ points had been interpreted as the evidence of the presence of DMI in CrI$_3$. As we show later, this gap can be caused by the correlation-enhanced interlayer super-exchange $J_2$. If one only considers the Cr sublattice itself, which has a higher symmetry ($R3\bar{m}$), $J_{2o}$ and $J_2$ should be equivalent. The presence of I sublattice breaks the mirror and 2-fold rotational symmetry, lifting the degeneracy of $J_{2o}$ and $J_2$.

Thus, although $J_{2o}$ and $J_2$ connect similar Cr pairs with the same distance of 9.517 Å, their exchange paths are not the same, which allows for $J_2$ and $J_{2o}$ to adopt different values. However, up until now, all previous work has considered that $J_2 = J_{2o} = 0$, which indeed is supported, to a great extent, by DFT. Surprisingly, the inclusion of correlations results in a sizable AFM $J_2$. As shown in Table 1, $J_{2o}$ vanishes in all calculations, while AFM $J_2$ is negligible in DFT but becomes stronger in QSGW and DFT+$U$, reaching $J_2 = 10\% J_1$ in QSGW+$U$. As shown in Fig. 3(b), $J_2$ corresponds to a Cr-I-I-Cr super-exchange with a Cr-I-I angle of 159°, giving the major AFM contribution to interlayer couplings, whereas vanishing $J_{2o}$ has no obvious exchange path with intervening I anions.

The gap between acoustic and optical modes, at an arbitrary $q$ point, gives the energy difference between the in-phase and out-of-phase precessions of two Cr-spin sublattices and depends on the inter-sublattice couplings $2|B(q)|$. Within the considered exchange range, we have $B(q) = J_1(q) + J_3(q) + J_{0}(q) + J_{1+}(q) + J_{2o}(q) + J_2(q)$, where $J_1(q)$ is the corresponding Fourier component of $J_1$. Couplings $J_1(q)$, $J_3(q)$, and $J_{1+}(q)$ are real functions along the $\Gamma-K-M$ path and vanish at the $K$-point, resulting a bandcrossing at $K$ if other terms are ignored. The interlayer coupling $J_0$ is along the $z$ direction; $J_0(q) = 0$ is a real constant when $q$ is in the basal plane, shifting the bandcrossing along the $\Gamma-K$ path. In real space, the connecting vectors (in-plane components) of $J_{2o}$ and $J_2$ are rotated by $\pi/6$ with respect to those of $J_1$, $J_3$, and $J_{1+}$. Correspondingly, in reciprocal space, $\tilde{J}_{2o}(q)$ and $\tilde{J}_2(q)$ are complex functions along the $\Gamma-K$ path (See Fig. S4 in the Supplemental Material). With $J_{2o} = 0$, $J_2(q)$ itself results in a non-vanishing $|B(q)|$ and thus a gap along the $\Gamma-K$ path. However, if $J_{2o} = J_2$, then $(\tilde{J}_{2o}(q) + \tilde{J}_2(q))$ is a real function when $q$ is in the basal plane, shifting the magnon crossing as $J_0$ does along the $\Gamma-K-M$ path. Thus, the combination of vanishing $J_{2o}$ and correlation-enhanced $J_2$ will induce the magnon gap along the $\Gamma-K$ path. To illustrate, we calculate the SW spectra in a simple $J_1-J_{2o}-J_2$ model with $J_{2o} = J_2 = J_1/12$ and a $J_1-J_2$ model with $J_2 = J_1/6$, respectively. As shown in Fig. 3(c), the latter case opens a gap along the $\Gamma-K-M$ path.

However, unlike the global DMI-induced gap, the gap induced by the nonequivalence of the exchange interactions $J_{2o}$ and $J_2$ does not persist through the whole BZ because a solution of $|B(q)|$=0 can be found near the $K$-point. The Dirac nodal lines in SW spectra still form but do not cross the $\Gamma-K-M$ lines (at the $k_z = 0$ plane). Using the $J_{ij}$ parameters obtained within QSGW+$U$, we calculate, within the linear SW theory, the in-plane SW spectra at various $k_z$ planes. Figure 3(d) shows the helical Dirac nodal lines form around the edges of the hexagonal BZ; each line crosses only twice the face of the first BZ. It would be interesting to see whether future INS experiments can confirm the small displacement of the Dirac point off the $\Gamma-K$ line or at finite $k_z$. However, such measurement may be challenging due to the complexity from the modulation of dynamic structure factor close to the gap, the requirements of high instrumental resolution and good sample mosaic.

Finally, we demonstrate that explicit treatments of electron correlations can correctly describe the dependence of interlayer interaction on stacking order. M-CrI$_3$ has different stacking than R-CrI$_3$, which dramatically modifies the inter-layer super-superexchange paths and results in A-type AFM ordering in M-CrI$_3$. This intimate interplay between stacking order and magnetic ordering plays a crucial role in manipulating the magnetism in these materials. DFT total energy calculations predict the wrong FM ground state for M-CrI$_3$, while DFT+$U$ calculations have shown that AFM interlayer configurations can be stabilized in M-CrI$_3$, depending on the $U$ value. Within QSGW, we calculate $\chi(q,\omega)$ in M-CrI$_3$ starting from both the FM and the A-type AFM ground-state configurations. The corresponding SW spectra along the high symmetry paths are shown in Fig. 3(a) and 3(b), respectively. The acoustic SW calculated with FM configuration, as shown in Fig. 3(a), is negative along the $\Gamma-Z$ path (normal to the basal plane), suggesting the instability of the FM interlayer configuration in M-CrI$_3$. In contrast, the SW spectra calculated with the AFM configuration, as shown in Fig. 3(b), are positive through the whole BZ. Thus, by taking into
Reciprocal lattice vectors are plotted along \( \Gamma - Y - \Gamma - Z - \Gamma \) for the FM structure. High-symmetry \( q \) points \( X, Y, \) and \( Z \) are at BZ boundaries and denoted as \((0.5, 0.5, 0)\), \((-0.5, 0.5, 0)\), and \((0, 0, 0.5)\), respectively. (a) \( \chi(q, \omega) \) calculated within QSGW. Wavevectors \( q = h b_1 + k b_2 + \ell b_3 \) is denoted as \( q = (h, k, \ell) \) in reciprocal lattice units (r.l.u.). SW are plotted along \( X - \Gamma - Y - \Gamma - Z \) for the FM structure and \( X - \Gamma - Y - \Gamma - Z - \Gamma \) for the AFM structure. High-symmetry \( q \) points \( X, Y, \) and \( Z \) are at BZ boundaries and denoted as \((0.5, 0.5, 0)\), \((-0.5, 0.5, 0)\), and \((0, 0, 0.5)\), respectively. (a) \( \text{Im}[\chi(q, \omega)] \) calculated in FM configuration. (b) \( \text{Im}[\chi(q, \omega)] \) calculated in AFM configuration. (c) The primitive cell of AFM M-CrI\(_3\) structure, in which the lattice vector \( a_3 \) is doubled, in comparison to the FM structure. (d) The first BZ of M-CrI\(_3\). Reciprocal lattice vectors \( b_1 \) and \( b_2 \) are the same for both FM and AFM structures while \( b_3^{(\text{FM})} = 2b_3^{(\text{AFM})} \).

In conclusion, we have demonstrated that the spin interactions and excitations in CrI\(_3\) can be accurately described by considering the non-local electron correlation effects within the system. In particular, we demonstrate that accounting for such correlations, without including DMI, are consistent with the SW spectra observed in INS experiments, previously interpreted as supporting the existence of a large DMI or Kitaev interaction. The other option may be magnon-phonon interaction as was hypothesised in Ref. [22]. Elucidating the nature of magnetic interactions is required to explore their potential topological applications. To experimentally verify the true physical mechanism of the gap opening, future INS experiments may be used to search for the bandcrossings off the high-symmetry line in bulk CrI\(_3\). Identifying the existence of the magnon gap along the \( \Gamma - K \) path in monolayer CrI\(_3\), in which the interlayer exchange is absent, can also help illuminate the responsible interactions. Of course, INS studies of single-layer materials seem to be impossible, due to a small number of atoms, but other techniques such as electron energy loss spectroscopy can be in principle used. Our work suggests the necessity of explicit treatment of electron correlations to accurately describe the magnetism in the broad family of magnetic layer materials including magnetic topological vdW materials, where the interaction between magnetization and the topological surface state is essential.

ACKNOWLEDGMENTS

We are indebted to B. Harmon, R. J. McQueeney, B. Li, J. R. Morris, and M. van Schilfgaarde for fruitful discussions. We are also grateful to L. Chen and P. Dai for providing their INS data. This work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division, and Early Career Research Program. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358. This research used resources of the National Energy Research Scientific Computing Center (NERSC), a U.S. Department of Energy Office of Science User Facility operated under Contract No. DE-AC02-05CH11231. The work by MIK is supported by European Research Council via Synergy Grant 854843 - FASTCORR.

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