On the origin of magnetic anisotropy in two dimensional CrI₃

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Abstract

The observation of ferromagnetic order in a monolayer of CrI₃ has been recently reported, with a Curie temperature of 45 K and off-plane easy axis. Here we study the origin of magnetic anisotropy, a necessary ingredient to have magnetic order in two dimensions, combining two levels of modeling, density functional calculations and spin model Hamiltonians. We find two different contributions to the magnetic anisotropy of the material, favoring off-plane magnetization and opening a gap in the spin wave spectrum. First, ferromagnetic super-exchange across the ≈90° Cr–I–Cr bonds, are anisotropic, due to the spin–orbit interaction of the ligand I atoms. Second, a much smaller contribution that comes from the single ion anisotropy of the $S = 3/2$ Cr atom. Our results permit to establish the XXZ Hamiltonian, with a very small single ion anisotropy, as the adequate spin model for this system. Using spin wave theory we estimate the Curie temperature and we highlight the essential role played by the gap that magnetic anisotropy induces on the magnon spectrum.

1. Introduction

The recent reports of ferromagnetic order in two different two dimensional crystals [1, 2], Cr₂Ge₂Te₆ and CrI₃, together with the report of antiferromagnetic order [3, 4] in FePS₃, a few months earlier, mark the beginning of a new chapter in the remarkable field of two dimensional materials. These discoveries extend significantly the list of electronically ordered two dimensional crystals, that included already superconductors [5, 6], charge density waves materials [7] and ferroelectrics [8]. In addition, there is an increasing amount of computational studies predicting magnetic order in large variety of two dimensional materials, such as VS₂ and VSe₂ [9], K₃CuF₄ [10], and the family of MPX₃, with M the 3d transition metals and X a group VI atom [11]. The integration of magnetically ordered 2D crystals in Van der Waals heterostructures [12] opens a vast field of possibilities for new physical phenomena and new device concepts, and is already starting to be explored experimentally [13].

Mermin and Wagner demonstrated the absence of long range magnetic order in spin–rotational invariant systems with short range exchange interactions [14]. Therefore, the observation of long range magnetic order in two dimensional insulating materials stresses the importance of a quantitative microscopic understanding of magnetic anisotropy in these systems. The breaking of spin rotational invariance can be due to three mechanisms, dipolar interactions, single ion anisotropy and anisotropy of the exchange interactions. In the case of very strong single ion anisotropy, a description in terms of the Ising model could be possible, which automatically entails a magnetically ordered phase at finite temperature, as predicted by Onsager in his remarkable paper [15]. However, large single ion anisotropies are normally associated to partially unquenched orbital moment of the magnetic ion, which only happens for specific oxidation states and low symmetry crystal environments, most notably in surfaces [16] or for rare earth atoms [17].

CrI₃ is a layered transition metal compound known to order ferromagnetically, in bulk, at $T_c = 61$ K [18, 19]. Ferromagnetic order has been shown to persist in mechanically exfoliated monolayers of CrI₃, with a Curie temperature of $T_c = 45$ K, as determined by magneto-optical measurements [2]. In this work we model magnetic anisotropy in a monolayer of CrI₃. Since dipolar interactions favor in-plane anisotropy, we focus on the study of both single ion anisotropy and exchange anisotropies. To do that, we first model the system with relativistic all electron density functional theory (DFT) calculations that include spin–orbit interactions, essential to account for magnetic anisotropy. Our calculations permit to build an effective spin model with three energy scales, the isotropic and...
anisotropic Cr–Cr exchange couplings, $J$ the anisotropic exchange $\lambda$, and the single ion anisotropy $D$. As we show below, $J$ and $\lambda$ are non-zero, whereas the single ion anisotropy $D$ is negligible.

Both experimental results [2, 19] and DFT calculations [20, 21] show that CrI$_3$ is an semiconducting material with a band-gap of 1.2 eV [18]. In a single layer of CrI$_3$, the plane of Cr atoms form a honeycomb lattice and is sandwiched between two atomic planes of I. The Cr ions are surrounded by 6 first neighbor I atoms arranged in a corner sharing octahedra. In an ionic picture, the oxidation state of Cr in this compound is $+3$, with 3 electrons occupying an orbit $3d^{3}$ in the $t_{2g}$ manifold, with 3 electrons occupying the $t_{2g}$ manifold [22]. Thus, we expect that Cr$^{3+}$ ions in this environment have $S = 3/2$, with 3 electrons occupying the $t_{2g}$ manifold, complying with first Hund rule (see figure 1(c)). The lack of orbital degeneracy results in an orbital singlet [22] with a quenched orbital moment. This picture is consistent with the observed [19] saturation magnetization of bulk CrI$_3$ that yields a magnetic moment of $\approx 3 \mu_B$ per Cr atom, that can be explained with $S = 3/2$ and $L = 0$.

Single ion magnetic anisotropy is originated by the interplay of spin–orbit coupling and the crystal field. In magnetic ions with a finite orbital moment, magnetic anisotropy scales like $E_{\text{MAE}} \propto \lambda (\hat{L} \cdot \langle \hat{S} \rangle)$, where $\lambda$ is the magnetic ion atomic spin–orbit coupling. However, when the orbital moment is quenched ($\langle \hat{L} \rangle = 0$), this lowest order non-zero contribution arises from quantum fluctuations of the orbital moment, and is given by $E_{\text{MAE}} \propto \frac{\lambda}{\Delta}$, where $\Delta$ is the energy separation with the crystal field excited states of the ion. Given that $\lambda \approx 10$ meV for Cr [23], and $\Delta$ is in the range of 500 meV, single ion anisotropy energies are very often way below 1 meV. In a purely octahedral environment this quadratic contribution would actually vanish [22, 24], and the magnetic anisotropy energy would scale like $E_{\text{MAE}} \propto \frac{\lambda}{\Delta}$, resulting in an extremely small single ion anisotropy. Based on these considerations, single ion anisotropy of Cr$^{3+}$ in CrI$_3$ should arise from the distortion of the octahedral environment.

Magnetic interactions between magnetic ions separated by non-magnetic ligands arise via the super-exchange mechanism proposed by Anderson [25]. This involves the virtual excitation of excited states where charge is transferred, during a Heisenberg time, from the ligand to the magnetic cations. This virtual processes reduce the total energy of the system and depend on the relative spin orientation of the magnetic atoms. The sign of this exchange interaction depends both on the angle $\theta$ formed by the two chemical bonds connecting the ligand and the magnetic atoms and on the filling of the $d$ levels of the cations. A set of rules to predict the sign of the interactions was proposed, independently, by Goodenough [26] and Kanamori [27]. In particular, ferromagnetic interactions are maximal when $\theta = 90^\circ$. For CrI$_3$, the angle $\theta \approx 93^\circ$, which accounts [28] for the ferromagnetic interactions. As long as spin–orbit interactions are neglected, these exchange interactions are always spin rotational invariant and can be described with a Heisenberg coupling $J_S \hat{S}_i \cdot \hat{S}_j$.

The possibility of magnetic anisotropy in the super-exchange interactions in magnetic insulators was proposed early on by Moriya [29]. In his seminar work, he considered the anisotropic interactions originated by spin–orbit coupling in the magnetic ions. He found two types of addition to the Heisenberg coupling, the first are the Dzyaloshinski–Moriya (DM) term or antisymmetric exchange, $J_D = \langle \hat{S}_i \times \hat{S}_j \rangle$, postulated by Dzyaloshinski [30]. The second is the anisotropic symmetric exchange, $\lambda S_i^z S_j^z$.

In the case of exchange mediated by an anion, the DM vector can be written as [31] $\vec{D}_{ij} = \vec{r}_i \times \vec{r}_j$, where $\vec{r}_i, \vec{r}_j$ link the anion with the two magnetic atoms. The DM favors non-collinear ground states. However, this term is absent in the CrI$_3$ crystal, since the two paths mediated by iodine contribute to with a DM vector with opposite sign that yield a net zero contribution. In contrast, the anisotropic symmetric exchange term is allowed by symmetry and, as we show below, it is definitely important in CrI$_3$. The symmetric and

![Figure 1](image-url). (a) Crystalline structure of CrI$_3$, showing the honeycomb arrangement of the chromium atoms. Every chromium atom has an octahedral iodine environment, where different octahedra are linked by an iodine forming an angle close to 90 degrees. (b) The octahedral environment splits the $d$ levels in the $e_g$ and $t_{2g}$ manifolds. (c) First Hund rule favors the $S = 3/2$ state, with 3 fully polarized electrons in the $t_{2g}$ manifold.
antisymmetric contributions to the anisotropic superexchange scale with $\lambda_I$ and $\lambda$, respectively [29], where $\lambda_I \simeq 0.6$ eV, is the atomic spin–orbit coupling of iodine [23].

2. Density functional methods

We perform density functional theory calculations with the pseudo-potential code quantum espresso [32] and the all-electron code Elk [33]. Monolayer structures were relaxed with quantum espresso, projector augmented wave (PAW) pseudopotentials [34, 35] and PBE exchange correlation functional [36] in the ferromagnetic configuration. With the relaxed structures, calculation with Elk are carried out using spin–orbit coupling in the non-collinear formalism, DFT + U with the Yukawa scheme [37] ($J = 0.7$ eV and $U = 2.7$ eV) in the fully localized limit and LDA exchange correlation functional [38]. We have verified that exchange energies with LDA or GGA, with or without DFT + U give qualitatively similar results.

The calculations of magnetic anisotropy require careful convergence of the total energy. We found that converging the total energy $10^{-3}$ eV yields stable results. We have used the feature of Elk that permits to tune the overall strength of spin–orbit interaction by a dimensionless constant scale factor, that we call $\alpha$. Thus, for $\alpha > 1$ the size of the spin–orbit coupling is increased above its actual value. In addition, we have introduced a modification in the source code of Elk in order to selectively turn on and off the spin–orbit coupling in the two different atoms independently, so that now we have two dimensionless scale factors, $\alpha_I$ and $\alpha_B$. As we discuss below, these two resources permit to to trace the origin of the magnetic anisotropy, as we discuss now.

3. Electronic properties of CrI$_3$

We now describe the most salient electronic properties of CrI$_3$, as described within our DFT calculations, in line with previous work [20, 39]. The calculations show that CrI$_3$ is a ferromagnetic semiconductor. The magnetic moment resides mostly in the Cr atoms, with a residual counterpolarized magnetization on the I atoms. The total magnetic moment in the unit cell is $6 \mu_B$, $3 \mu_B$ per Cr atom. Figure 2(a) shows the band structure, calculated with and without SOC. The bands undergo a rather large shift, in the range of 0.1 eV, when SOC is included. The size of this shift is a first indication that the spin–orbit interaction of iodine atoms plays an important role [40], as spin–orbit coupling in Cr is much smaller than 0.1 eV. Figures 2(b) and (c) shows the bands weighted over the projection on the $d$ orbitals of Cr (figure 2(b)) and the $p$ orbitals of I (figure 2(c)). It is apparent that the top of the valence band is formed mostly by spin unpolarized $p$ orbitals of the I atoms and the conduction band is formed by the minority spin $d$ orbitals of Cr. The majority spin $d$ orbitals, of the $t_{2g}$ manifold, are found 2 eV below the top of the valence bands. The shape of the magnetization field, not shown, clearly shows that the magnetic moment resides in orbitals with $t_{2g}$ symmetry, in line with previous results [39].

4. Magnetic anisotropy

We are now in position to discuss the main topic of this work, magnetic anisotropy. We have verified that the in-plane anisotropy is negligibly small. Therefore, in the following we focus on the off-plane anisotropy and we compute the quantity:

$$\Delta E_{\text{MAE}} = E_G(0) - E_G(90)$$

where $E_G(\theta)$ is the computed ground state energy as a function of the angle $\theta$ that forms the magnetic moment with the atomic planes. $E_{\text{MAE}} > 0$ describes an off-plane easy axis system. For the in-plane component, we take $M_z = 0$. In line with previous work [20], we obtain $\Delta E_{\text{MAE}} = 0.65$ meV. Thus, the calculation predicts that the system has an easy axis anisotropy, perpendicular to the atomic planes, in agreement with the experiments [2].

In the case $U = 0$, the weight of the $t_{2g}$ orbitals at the top of the valence band increases.

Figure 2. (a) Band structure of the system in the ferromagnetic state, without SOC (light blue) and with SOC (black). The large variation upon switching on spin–orbit coupling cannot be accounted by the energy scale of the spin–orbit coupling of chromium. Projection of the band structure over the chromium (b) and iodine (c) atoms. From panels (b) and (c) is inferred that the valence band shows a strong iodine character and that the conduction band is dominant from chromium, but with a non zero contribution from iodine. This chromium-iodine mixing is the responsible of the large variation of the conduction band upon switching on SOC, and is the ultimate responsible of the anisotropic exchange.
In order to study the origin of this magnetic anisotropy we compute how $E_{\text{MAE}}$ changes as we vary independently spin–orbit coupling in two atoms [41]. To do so, here we define the DFT Hamiltonian as

$$H_{\text{DFT}} = H_0 + \alpha_I H_{\text{SOC}}^I + \alpha_C H_{\text{SOC}}^C$$  \hspace{1cm} (2)$$

where $H_0$ is the non relativistic Hamiltonian, $H_C$ the relativistic Hamiltonian correction to chromium, and $H_I$ the relativistic Hamiltonian correction to iodine. We compute magnetic anisotropy energy from equation (1), keeping at the default value $\alpha_C = 1$ only one of the species, and ramping the other. The results are shown in figures 3(a) and (b) and permit to conclude that MAE arises predominantly from the spin–orbit coupling in iodine atoms. This suggests that anisotropic symmetric superexchange is the likely cause of magnetic anisotropy in this compound. This also seems to indicate that the local moments do not have a strong single ion anisotropy, and therefore they are not properly described as Ising spins.

### 4.1. Spin Hamiltonian

In order to validate these hypothesis, we now propose a model Hamiltonian for the spins of the Cr atoms in the honeycomb lattice:

$$H = -\sum_i D(S_i^z)^2 - \frac{J}{2} \sum_{i,j} S_i \cdot S_j + \frac{\lambda}{2} \sum_{i,j} S_i^z S_j^z$$  \hspace{1cm} (3)$$

where the sum over $i$ runs over the entire lattice of Cr atoms, and the sum over $i'$ runs over the 3 atoms, the first neighbors of atom $i$. The first term in the Hamiltonian describes the easy axis single ion anisotropy and we choose $z$ as the off–plane direction. The second term is the Heisenberg isotropic exchange and the final term is the anisotropic symmetric exchange. The sign convention is such that $J > 0$ favors ferromagnetic interactions and $D > 0$ favors off-plane easy axis. $\lambda = 0$ would imply a completely isotropic exchange interaction.

We first treat equation (3) in the classical approximation, and we describe the spins $S$ as dimensionless classical vectors of length $S$ in the sphere. We write the energy of the ground state for 4 possible ground states, depicted in figure 4(a): (I) ferromagnetic off-plane (FM, $z$), (II) antiferromagnetic off-plane (AF, $z$), (III) ferromagnetic in-plane (FM, $x$) and (IV) antiferromagnetic in-plane (AF, $x$). We denote the corresponding classical ground state energies as $E_{\text{FM},z}, E_{\text{AF},z}, E_{\text{FM},x}, E_{\text{AF},x}$. The spin model allows to write the energetics of the different configurations normalized per unit cell (2 Cr atoms) as

$$E_{\text{FM},z} = -2S^2D - 3S^2(J + \lambda)$$  \hspace{1cm} (4)$$

$$E_{\text{AF},z} = -2S^2D + 3S^2(J + \lambda)$$  \hspace{1cm} (5)$$

$$E_{\text{FM},x} = -3S^2J$$  \hspace{1cm} (6)$$

$$E_{\text{AF},x} = +3S^2J$$  \hspace{1cm} (7)$$

with $S = 3/2$ for CrI$_3$. In order to determine $J$, $D$ and $\lambda$, we use the ground state energies for these 4 configurations as obtained from our DFT calculations. In addition, we do this ramping the overall strength of the spin–orbit coupling, $\alpha = \alpha_C = \alpha_I$. For $\alpha = 1$ we obtain $J = 2.2$ meV, in line with the results by Zhang et al [20] Our results for $D$ and $\lambda$ are shown in figure 4(b).

It is apparent that the anisotropic symmetric exchange $\lambda$ is much bigger than the single ion anisotropy $D$, in particular for $\alpha = 1$. The precise value of $D$ was affected by numerical noise in the regime where both $J$ and $\lambda$ already reached convergence, being always $D$ at least 30 times smaller that the anisotropic exchange $\lambda$.  

![Figure 3](image-url)  

Figure 3. (a) Evolution of the magnetic anisotropy energy as a function of the spin–orbit coupling in iodine $\alpha_I$, keeping the spin–orbit coupling in Cr to the real value $\alpha_C = 1$ (b) Same, reverting the roles of I and Cr. These curves show the dominant contribution of iodine spin–orbit coupling to the MAE.
This yields a value of $D$ negligible with respect any other exchange energy scale. Thus, we have $\gg \lambda > JD$, which leads us to claim that the adequate spin model for CrI$_3$ is the XXZ model with negligible single ion anisotropy. This is the most important result of this work. We find $\lambda = 0.04 \pm 0.09 \text{ meV}$ for $\alpha = 1$. Thus, the flip-flop exchange is just 4 percent smaller than the Ising exchange $\lambda + J$, given by $\lambda = D + 3S(J + \lambda)$, where $\lambda$ is the spin–orbit coupling, with $D$ being the single ion anisotropy and $J$ the exchange interaction. Whereas the spin-flip part of exchange is actually responsible of the existence of dispersive spin wave excitations, the anisotropic term $\lambda$ opens up a gap in their spectrum, as we show below. This actually controls the transition from the ferromagnetic to the non-magnetic phase as the material is heated above $T_c$.

5. Spin wave theory

We now go beyond the classical approximation used in the previous section. To do that, we now treat the spins in Hamiltonian (3) as quantum mechanical $S = 3/2$ operators. We treat the Hamiltonian within the linear spin wave approximation. To do so, we use the so called Holstein–Primakoff representation [42] of the spin operators in terms of bosonic operators

$$S_i^+ = \sqrt{2S} \sqrt{1 - b_i^\dagger b_i} b_i$$

$$S_i^- = \sqrt{2S} b_i^\dagger \sqrt{1 - b_i^\dagger b_i}$$

$$S_i^z = S - b_i^\dagger b_i$$

with $b_i$ and $b_i^\dagger$ the bosonic annihilation and creation operator in site. The representation of the spin Hamiltonian (3) in terms of this bosonic operators leads a complicated non-linear Hamiltonian. The spin wave approximation consist in keeping only the quadratic terms in the bosonic operators $b$. This approximation is valid for a small occupation of the bosonic modes, i.e. when the magnetization is closed to $S_\parallel \sim S_\perp$, i.e. for small temperatures. In the spin wave approximation, the effective Hamiltonian for the spin excitations reads:

$$\mathcal{H}_{\text{spin waves}} = \sum_i (2DS + 3S(J + \lambda)) b_i^\dagger b_i - JS \sum_{\langle ij \rangle} b_i^\dagger b_j$$

where the sum over $i$ runs over the entire lattice and the sum over $j$ runs over the first neighbors of $i$. This Hamiltonian describes bosonic excitations moving in a honeycomb lattice, with an on-site energy $\epsilon_0 = 2DS + 3S(J + \lambda)$ and a hopping energy $JS$. Thus, the Bloch Hamiltonian for the honeycomb lattice reads

$$E^{\pm}(\vec{k}) = \epsilon_0 \pm JS \sqrt{f(\vec{k})}$$

where $\epsilon_0 = 3JS + 2SD + 3S\lambda$, $f(\vec{k}) = 1 + \epsilon_1^2 \hat{a}_1 + \epsilon_2^2 \hat{a}_2$ is the usual form factor for the honeycomb lattice, and $\hat{a}_{1,2}$ are the unit vectors of the triangular lattice. The resulting energy spectrum is

$$E^{\pm}(\vec{k}) \approx \Delta_0 + \rho k^2$$

where the spin wave gap is given by

$$\Delta_0 = 2DS + 3S\lambda$$

For CrI$_3$ we can take $D = 0$ and we have a spin wave gap $\Delta_0 = 3S\lambda = 0.4 \text{ meV}$. The so called spin stiffness is given by

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**Figure 4.** (a) Sketch and energetics of the different collinear magnetic configurations for two chromium atoms, showing in-plane and off-plane ferromagnetic and antiferromagnetic configurations. Comparison with DFT permits to extract $J, D$ and $\lambda$ (see text). (b) Evolution of the single ion anisotropy $D$ and anisotropic exchange $\lambda$ as a function of the spin–orbit coupling $\alpha$, where $\alpha = 1$ corresponds to the real value, as described at the DFT level.
\[ \rho = \frac{1}{4} JS \]  
\[ (16) \]

that yields for CrI\(_3\) a value \( \rho = 0.82 \text{ meV} \). The ratio \( \Delta \rho / \rho = 0.49 \) plays an important role in the following.

From equations (14) and (15) it is apparent that if the two terms that break spin rotational invariance in the original Hamiltonian (3), \( D \) and \( \lambda \), vanish, the spin wave spectrum becomes gapless. Therefore, in the spin wave spectrum, both the anisotropic exchange and the single ion anisotropy create a gap in the spin waves (see figure 5(a)), so that their effect on the spin wave dispersion is similar. This implies that simple inspection of the spin wave dispersion does not provide enough information to assess whether the correct model for a compound is single ion anisotropy or anisotropic exchange, and input from a microscopic first principles calculation is necessary. As we discuss now, the presence of their induced gap is essential to have magnetization at finite temperature.

### 5.1. Low temperature magnetization

Every magnon carries one unit of angular momentum. Therefore, in the linear spin wave framework, we can approximate the magnetization by

\[ M(T) = S - \delta M = S - \frac{1}{2(2\pi)^2} \int_\text{BZ} \frac{d^2k}{e^{\beta E(k)}} - \frac{1}{2} \]  
\[ (17) \]

where \( M(T) \) is the magnetization in units of \( \hbar \) per Cr atom as a function of the temperature, the factor 1/2 comes from having two Cr atoms in the unit cell and \( \beta^{-1} = k_B T \). Linear spin wave theory works well when the magnon density is small. In the following we use the fact that this integral is controlled by the low energy magnons, and we approximate \( E(k) \) by equation (14).

In addition, we replace the integral over the Brillouin zone by the integral over a circle of radius \( k_c \), chosen so that the density of magnons is properly normalized \( \frac{1}{2\pi} \int_0^{k_c} kdk = 2 \). Choosing this normalization includes the contribution from the high energy magnon branch in the high energy part of the dispersion. We first focus in the case \( \Delta_0 = 0 \), i.e. in the absence of anisotropic exchange, in that case the correction of the magnetization goes as

\[ \delta M = \frac{1}{4\pi} \int_0^{k_c} \frac{kdk}{\beta \rho k} \to \infty \]  
\[ (18) \]

This divergence signals the absence of order at finite temperature in the Heisenberg model in the gapless regime \( \Delta_0 = 0 \), consistent with the Mermin–Wagner theorem [14]. Therefore, the anisotropy gap is essential to protect the long range order in 2D.

We will move now to the case of finite spin wave gap \( \Delta_0 \neq 0 \). We now consider the very low temperature case, \( \beta \Delta_0 \gg 1 \). We can then approximate

\[ M(T) = M(T = 0) + \delta M = S - \frac{k_B T}{2\pi JS} e^{-\Delta_0 / \beta k_B T} \]  
\[ (19) \]

Thus, we expect that the magnetization will have a very weak temperature dependence for temperatures smaller than spin wave gap. According to our calculations \( \Delta_0 = 0.4 \text{ meV} \), so \( M(T) \) be almost maximal up to \( T = 5 \text{ K} \).

### 5.2. Estimate of \( T_c \)

We now provide a rough estimate of the Curie temperature, based on non linear spin wave theory. We use the initial expression for spin operators, and expand them retaining the up to fourth order in the bosonic operators

\[ S^z_i \approx \sqrt{2S} \left( 1 - \frac{b^\dagger b_i}{4S} \right) b_i \]  
\[ (20) \]

\[ S^z_i \approx \sqrt{2S} b_i \left( 1 - \frac{b^\dagger b_i}{4S} \right) \]  
\[ (21) \]

\[ S^z_i = S - b^\dagger b_i \]  
\[ (22) \]
At intermediate temperatures, there is a finite number of spin waves, that is accounted by the higher order terms in bosonic operators when substituting the previous expansion in the spin Hamiltonian. In that situation, the spin Hamiltonian contains four field operators and therefore is not exactly solvable. Thus, the effect of the spin wave population is described using a mean field approximation in the spin wave Hamiltonian by means of the substitution $b_i^\dagger b_j^\dagger b_j b_i \approx (b_i^\dagger b_j^\dagger b_j^\dagger b_j^\dagger) + C$. With the previous approximation it is straightforward to check that a finite population of spin waves is equivalent to a renormalization of the hopping energy and spin wave gap as $[44]$

$$ JS \rightarrow J(S - \langle b_i^\dagger b_j \rangle) = JM(T) \quad (23) $$

$$ \lambda S \rightarrow \lambda(S - \langle b_i^\dagger b_j \rangle) = \lambda M(T) \quad (24) $$

The previous substitutions lead to a selfconsistent equation for the magnetization as

$$ M = S - \frac{1}{2\pi(2\pi)^2} \int_{BZ} \frac{d^2k}{\beta} \sum_{\epsilon(k)} e^{i\beta\epsilon(k)/JS} = \frac{1}{2} \left[ 1 - \frac{\pi J S^2}{2 \log \frac{\Delta_0 + 2\pi J S}{\Delta_0}} \right] \quad (25) $$

where the integral extends over the first Brillouin zone. A qualitative behavior of the previous integral can be obtained approximating $E(k) = \Delta_0 + p k^2$ and $e^{i\beta\epsilon(k)} - 1 \approx \beta \epsilon(k)$. As equation (25) has no solution for $M = 0$, we define $T_c$ as the temperature at which the magnetization is depleted to $M = S/2$. This leads to the following equation:

$$ k_B T_c \approx \frac{2\pi \rho S}{\log \frac{\Delta_0 + 8\pi \rho}{\Delta_0}} = \frac{\pi J S^2}{2 \log \frac{\Delta_0 + 2\pi J S}{\Delta_0}} \quad (26) $$

A very similar result can be obtained using different spin representations $[45, 46]$. Equation (26), together with the numerical solution $[47]$ of equation (25) in figure 5(b), show several important results. First, $T_c$ is an increasing function of the spin wave gap $\Delta_0$ (see figure 5(c)). This is in line with the experimental results recently reported for CrGe$_2$Te$_6$ $[1]$, for which the major contribution to the spin wave gap comes from the Zeeman contribution, due to the very tiny intrinsic anisotropy, resulting in dramatic variations of $T_c$ as a function of the applied field. This is a feature specific of two dimensional magnets with dispersive spin waves. Second, $T_c$ is significantly smaller than the prediction coming from the Ising model. The exact solution for the Ising model in the honeycomb lattice $[48]$ yields $k_B T_c = 1.51 J$, where $J$ is the coupling between classical spins with $S = 1$. Using this result for CrI$_3$, we would have $k_B T_c = 1.51 (J + \lambda) S = 85$ K, that overshoots the experimental value 45 K.

On the other hand, using the prediction of $T_c$ obtained by the numerical solution of equation (25) shown in figure 5(c), we obtain a value of $k_B T_c = 3.5 \rho$, for $\Delta_0 = 0.49 \rho$, that gives $T_c = 33$ K, underestimating the the experimental value $[2]$ $T_c = 45$ K by 20%. Including the effect of the finite magnetic field would increase $\Delta_0$ and push the prediction upward. Inclusion of longer range coupling $[49–52]$ is also expected to increase the spin stiffness, yielding a larger estimate of the critical temperature. Furthermore, a more accurate treatment must consider the explicit spin wave density of states and a more careful treatment of fluctuations close to the critical point. The discrepancy highlights the limitations of the non-linear spin wave theory, and perhaps, also those of the DFT scheme to determine the energy scales of the Hamiltonian. Nevertheless, apart from the previous limitations, our approach highlights the role played by anisotropic exchange, as the ultimate mechanism responsible to controlling the divergence in equation (26).

6. Conclusions

We have studied the origin of magnetic anisotropy in two dimensional CrI$_3$, a recently discovered ferromagnetic two dimensional crystal with off-plane anisotropy. We have found that magnetic anisotropy in this system comes predominantly from the superexchange interaction, that gives rise to an anisotropic contribution to the conventional exchange interaction. The strength of the non Heisenberg correction is found to be controlled by the spin-orbit coupling of the intermediate iodine atom. The single ion anisotropy of the magnetic Cr atoms is found to give a negligible contribution to magnetic anisotropy. The suppression of the single ion anisotropy due to the octahedral environment, together with large spin-orbit coupling of iodine, make the anisotropic exchange the leading mechanism stabilizing the magnetic ordering in 2D CrI$_3$. Our calculations permit to conclude that the effective spin Hamiltonian for CrI$_3$ is a XXZ model. In turn, this implies that gapped spin waves are the essential elementary excitations that control the finite temperature properties of this new type of magnetic system. Given that spin waves in two dimensions are interesting on its own right, as they can exhibit thermal Hall effect and have topologically non-trivial phases $[53–58]$. As an example, one can consider inducing a Dzyaloshinskii-Moriya term in a CrI$_3$ monolayer by applying a perpendicular electric field, opening the possibility of a skyrmionic ground state whose magnonic Hamiltonian is topologically non-trivial and shows gapless edge magnonic excitations $[55]$. Another interesting playground would be the possibility of applying non uniform strain to the ferromagnetic monolayer, modulating the exchange constants and creating an artificial gauge field in the magnonic Hamiltonian $[59, 60]$. Therefore, the discovery of magnetic 2D crystals paves the way towards the exploration of these exciting phenomena.

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