Spin dynamics of antiferromagnetically coupled bilayers—the case of Cr$_2$TeO$_6$

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Abstract

Understanding the dynamics of interacting quantum spins has been one of the active areas of condensed matter physics research. Recently, extensive inelastic neutron scattering measurements have been carried out in an interesting class of systems, Cr$_2$(Te, W, Mo)O$_6$. These systems consist of bilayers of Cr$^{3+}$ spins ($S = 3/2$) with strong antiferromagnetic inter-bilayer coupling ($J$) and tuneable intra-bilayer coupling ($j$) from ferro (for W and Mo) to antiferro (for Te). In the limit when $J > |j|$, the system reduces to weakly interacting quantum spin-3/2 dimers. In this paper, we discuss the low-temperature magnetic properties of Cr$_2$TeO$_6$ systems where both intra-layer and inter-layer exchange couplings are antiferromagnetic, i.e., $J, j > 0$. Using linear spin-wave theory we obtain the magnon dispersion, sublattice magnetization, two-magnon density of states, and longitudinal spin–spin correlation function.

Keywords: magnetic spin systems, frustrated systems, quantum magnetism, quantum magnets

(Some figures may appear in colour only in the online journal)

1. Introduction

Quantum spin fluctuations (QSF) play an important role in the low temperature properties of quantum antiferromagnets (QAF), particularly in systems with low spin and low dimension [1–4]. For example, the ordered moment or sublattice magnetization ($M_s$) in a nearest-neighbor (NN) $D$-dimensional spin-1/2 Heisenberg quantum antiferromagnet (HQAF) is zero for $D = 1$ (no long range order), 0.3067$\mu_B$ for $D = 2$ [1, 5], and 0.423$\mu_B$ for $D = 3$ (isotropic couplings) [6], the latter two values obtained in a leading order approximation (to be discussed later in the paper). Thus QSF decrease with increasing $S$ and increasing $D$. The interplay of QSF and covalency induced reduction of $M_s$ in QAFs has also been a subject of great interest in the past, particularly in the parent compound of high $T_c$ superconductors (La$_2$CuO$_4$ where the Cu$^{2+}$ ions have $S = 1/2$, form a square lattice and interact with NN isotropic Heisenberg antiferromagnetic interaction) [7].

Recently Zhu et al [8] have studied the magnetic structure of an interesting class of layered magnetic systems containing Cr$_x$X$_{6-x}$, $X =$ Te, Mo, W where the Cr$^{3+}$ (spin-3/2) magnetic ions are arranged in bilayers. The inter-bilayer coupling is strongly antiferromagnetic due to the presence of Cr$^{3+}$–Cr$^{3+}$ dimers. The intra-bilayer couplings however can be either antiferromagnetic (AF) or ferromagnetic (F) depending on whether the system contains Te or W (also Mo) atom [9]. In fact the average intra-bilayer exchange can be tuned from one limit to the other in Cr$_2$W$_{1-x}$Te$_x$O$_6$ by changing $x$ from 0 to 1. Neutron powder diffraction (NPD) measurements have determined the ground state spin structure and the values of the sublattice magnetization. The Te compound consists of antiferromagnetic bilayers which are coupled antiferromagnetically (AF–AF). In contrast, the W and Mo analogs consist of ferromagnetic bilayers coupled antiferromagnetically (F–AF). In addition to the magnetic ordering the NPD measurements also give the values of sublattice magnetization $M_s$. The values of $M_s$ are reduced from their atomic spin value 3.0$\mu_B$ for Cr$^{3+}$ ($M_s \approx g\mu_B S$ assuming $g = 2$ and quenched orbital angular momentum. This reduction can be due to covalency where the Cr $d$ orbitals hybridize with O $p$ orbitals and due to QSF [7]. Electronic structure calculations reveal information about the reduction due to covalency whereas QSF-caused reduction can be calculated using a quantum Heisenberg spin Hamiltonian, which is one of the issues we address in this paper.)


*Ab initio* electronic structure calculations using density functional theory (GGA and GGA+U) [10–12] correctly reproduced the magnetic ordering in these three compounds [8]. Since the orbital angular momentum is quenched for the Cr$^{3+}$ configuration (three electrons in the $t_{2g}$ orbitals; $S = 3/2$) the magnetic moment comes from the spin. The estimated exchange parameters (inter-bilayer or inter-dimer exchange $J$ and intra-bilayer NN exchange $j$) were reasonable in view of the limitations of GGA or GGA+U approximations [8]. However the calculated values of the sublattice magnetization of the limitations of GGA or GGA+U approximations [8].

In contrast, the experimental values are reduced to $\sim 2.3 \mu_B$ [8]. One possible reason for this reduction is QSF [1]. Such dramatic reduction in ordered moment has been seen in many quasi-two dimensional QAFs, a classic example being La$_2$CuO$_4$ which consists of antiferromagnetic 2D square lattice of $S = 1/2$, where QSF reduce the sublattice magnetization ($M_s$) by $\sim 40\%$ [1]. In the present systems the reduction should be smaller (at least by a factor of three) due to $S = 3/2$.

To visualize the magnetic ordering and exchange coupling in these systems we will consider the ground state spin ordering in Cr$_2$TeO$_6$ (see figure 1) [13]. One has two bilayers (perpendicular to the $z$-axis) in the tetragonal unit cell ($a,a,c$) and four Cr spins/unit cell. The experimental unit cell parameters are $a = 4.545$ Å and $c = 8.995$ Å for Cr$_2$TeO$_6$ and $a = 4.583$ Å and $c = 8.853$ Å for Cr$_2$WO$_6$ [8]. The distance between the inter-bilayer (NN) Cr atoms i.e. Cr1 and Cr3 or Cr2 and Cr4 is $\delta \sim 3.00$ Å $\approx c/3$, whereas the distance between intra-bilayer NN Cr atoms (Cr1 and Cr2 or Cr3 and Cr4) is $\sim 3.80$ Å. One bilayer contains Cr1 and Cr2 spins and the other contains Cr3 and Cr4 spins. The inter-bilayer coupling comes through Cr1–Cr3 and Cr2–Cr4 dimers, it is antiferromagnetic and its strength is denoted by $J$. The NN intra-bilayer coupling is between Cr3–Cr4 and Cr1–Cr2 and its strength is denoted by $j$. The magnitude of $j$ is considerably smaller than that of $J$ in these systems which can therefore regarded as weakly interacting quantum dimers. In Cr$_2$TeO$_6$ the intra-bilayer coupling is antiferromagnetic. In thermodynamic measurements the high temperature properties (for example peak in heat capacity) are determined primarily by the dimers i.e. the energy scale is set by the intra-dimer coupling strength $J$ whereas for the low temperature properties below the antiferromagnetic transition temperature the intra-dimer coupling $j$ is responsible for the long range order. Also it plays an important role in magnon dispersion and quantum spin fluctuations. Experimental values of the couplings estimated from high temperature susceptibility measurements are: $|J| = 2.9$ meV and $|j| = 0.4$ meV for Cr$_2$TeO$_6$ and $|J| = 3.8$ meV and $|j| = 0.12$ meV for Cr$_2$WO$_6$ [14].

The present systems are somewhat peculiar. The inter-bilayer coupling $J$ is AF and strong. The intra-bilayer coupling $j$ is small and can be either F or AF. If we put $j = 0$, then the system consists of non-interacting quantum spin spin-3/2 dimers and the ground state consists of a product of dimer singlet states and there is no long range order (LRO). If on the other hand, $J = 0$, the system consists of non-interacting bilayers. When $j$ is AF, the system is similar to the cuprates [7] described above but the QSF reduces $M_s$ by only 13% because $S = 3/2$. When $j$ is $F$, there is no QSF reduction of $M_s$. An important question is how $J$ and $j$ interfere with each other. To study this interesting problem we have calculated the magnetic excitations and QSF for the interacting quantum spin dimer problem using a Heisenberg quantum Hamiltonian.

In the current article we discuss the low temperature magnetic properties and the role of QSF in Cr$_2$TeO$_6$ systems using a three parameter $(J,j,j')$ spin-3/2 quantum Heisenberg Hamiltonian. Here $j'$ is the next nearest neighbor (NNN) intra-bilayer exchange coupling which is weakly ferromagnetic. Specifically, we obtain the magnon energy dispersion, sublattice magnetization, longitudinal spin–spin correlation function, and its powder-averaged intensity. In a forthcoming paper we will discuss the spin dynamics of Cr$_2$WO$_6$ and Cr$_2$MoO$_6$ systems, where the intra-bilayer coupling is ferromagnetic and inter-bilayer coupling is antiferromagnetic. The paper is organized as follows: we define the spin Hamiltonian in section 2 and discuss the formalism using linear spin-wave theory [5, 15] for the AF–AF bilayers (section 2). In section 3 we present the results for the magnon dispersion, sublattice magnetization, two-magnon density of states, longitudinal spin–spin correlation function, and its powder-average. We conclude our paper with a brief conclusion in section 4.

2. Formalism

It is well-known that quantum fluctuations play a significant role in the magnetic properties [1, 2] and phase diagram of the system at zero temperature [3–6, 15–18]. Here we investigate the role of quantum fluctuations on the stability of the Néel state by calculating the magnon spectrum and see how these excitations reduce the sublattice magnetization from its Néel state value. There are several analytical and numerical methods to study the low temperature properties of quantum magnets [3]. One such method is the spin-wave theory (SWT), which has proved to be a very effective to the study of quantum magnets described by the Heisenberg hamiltonian especially for dimensions $D \geq 2$ and large spin $S$ [1]. SWT provides accurate results for many physical quantities even for the difficult case of spin-1/2 quantum Heisenberg antiferromagnet in two dimensions. Our quantum bilayer spin system is three dimensional and the Cr$^{3+}$ ions have spin-3/2—thus SWT method is well suited for the present bilayer spin system. In the SWT formalism, we first express the fluctuations around the classical long range ordered antiferromagnetic ground state in terms of the bosonic operators using the Holstein–Primakoff (HP) representation [19]. The quadratic term in magnon Hamiltonian corresponds to the linear spin-wave theory (LSWT), whereas the higher-order terms e.g. quartic terms [15] represent interactions between magnons which we ignore in our current work.
represents spin of Cr$_2$ in the $n$th unit cell respectively through the intra-bilayer NN coupling. The factor of 1/2 in equation (1) implies that each bond is counted once. For example, $J_{jn}^i$ is the number of cells along the $n$-direction) and $i,j$ are nearest-neighbor (NN) sites within the same bilayer ($\langle i,j\rangle$) implies that each bond is counted once). For example, $S_{m}^{(4)A}$ represents spin of Cr1 at site $i$ in the $n$th unit cell whereas $S_{m+1}^{(2)B}$ represents spin of Cr2 in the $(n+1)$th unit cell.

The Heisenberg Hamiltonian of the system with antiferromagnetic intra- and inter-bilayer couplings we also add next-to-nearest neighbor (NNN) ferromagnetic coupling $j'$. The full Hamiltonian is then:

$$H = H_{\text{NNN}} + H_{\text{NN}}, \quad (2)$$

$$H_{\text{NNN}} = -\sum_{n=1}^{N_c} \sum_{\langle i,j \rangle} \left[ S_{m}^{(4)A} \cdot S_{jn}^{(1)A} + S_{m}^{(4)A} \cdot S_{jn}^{(4)A} + S_{m}^{(2)B} \cdot S_{jn}^{(2)B} + S_{m}^{(3)B} \cdot S_{jn}^{(3)B} \right]. \quad (3)$$

Above $\langle \langle i,j \rangle \rangle$ refer to NNN interaction (bond) between the spins, each bond is counted once, and $j' > 0$ (ferromagnetic interaction). This spin Hamiltonian is mapped onto an equivalent Hamiltonian of interacting bosons by expressing the spin operators in terms of bosonic creation and annihilation operators $a^\dagger$, $a$ for ‘up’ sites on sublattice A (and $b^\dagger$, $b$ for ‘down’ sites on sublattice B) using the Holstein–Primakoff representation [19]

$$S_{m}^{+A} \approx \sqrt{2S}a_{m}^\dagger, \quad S_{m}^{-A} \approx \sqrt{2S}a_{m}, \quad S_{m}^{0A} = S - a_{m}^\dagger a_{m},$$

$$S_{m}^{+B} \approx \sqrt{2S}b_{m}^\dagger, \quad S_{m}^{-B} \approx \sqrt{2S}b_{m}, \quad S_{m}^{0B} = -S + b_{m}^\dagger b_{m}. \quad (4)$$

Substituting equation (4) into equation (2) we expand the Hamiltonian perturbatively in powers of 1/$S$ up to the quadratic term as

$$\mathcal{H} = \mathcal{H}_d + \mathcal{H}_0 + \cdots, \quad (5)$$

where

$$\mathcal{H}_d = -2JNS^2 \left[ 4(1 + \eta') + \eta \right]. \quad (5a)$$

\textbf{2.1. Magnon dispersion}

As discussed in section 2 there are four chromium atoms in the tetragonal unit cell as shown in figure 1. Within each bilayer the spins of Cr1 and Cr3 are coupled antiferromagnetically with the spins of Cr2 and Cr4 respectively through the intra-bilayer NN coupling. On the other hand, the NN Cr1 and Cr3 spins belonging to different bilayers are coupled antiferromagnetically within the same unit cell whereas Cr2 spin is coupled antiferromagnetically to Cr4 spin in the unit cell below and Cr4 spin is coupled with Cr2 in the unit cell above.

In addition to the NN intra-layer and inter-layer couplings we also add next-to-nearest neighbor (NNN) ferromagnetic coupling $j'$. The full Hamiltonian is then:

$$H = H_{\text{NNN}} + H_{\text{NN}}, \quad (2)$$

with

$$H_{\text{NNN}} = -\sum_{n=1}^{N_c} \sum_{\langle i,j \rangle} \left[ S_{m}^{(4)A} \cdot S_{jn}^{(1)A} + S_{m}^{(4)A} \cdot S_{jn}^{(4)A} + S_{m}^{(2)B} \cdot S_{jn}^{(2)B} + S_{m}^{(3)B} \cdot S_{jn}^{(3)B} \right]. \quad (3)$$

Above $\langle \langle i,j \rangle \rangle$ refer to NNN interaction (bond) between the spins, each bond is counted once, and $j' > 0$ (ferromagnetic interaction). This spin Hamiltonian is mapped onto an equivalent Hamiltonian of interacting bosons by expressing the spin operators in terms of bosonic creation and annihilation operators $a^\dagger$, $a$ for ‘up’ sites on sublattice A (and $b^\dagger$, $b$ for ‘down’ sites on sublattice B) using the Holstein–Primakoff representation [19]

$$S_{m}^{+A} \approx \sqrt{2S}a_{m}^\dagger, \quad S_{m}^{-A} \approx \sqrt{2S}a_{m}, \quad S_{m}^{0A} = S - a_{m}^\dagger a_{m},$$

$$S_{m}^{+B} \approx \sqrt{2S}b_{m}^\dagger, \quad S_{m}^{-B} \approx \sqrt{2S}b_{m}, \quad S_{m}^{0B} = -S + b_{m}^\dagger b_{m}. \quad (4)$$

Substituting equation (4) into equation (2) we expand the Hamiltonian perturbatively in powers of 1/$S$ up to the quadratic term as

$$\mathcal{H} = \mathcal{H}_d + \mathcal{H}_0 + \cdots, \quad (5)$$

where

$$\mathcal{H}_d = -2JNS^2 \left[ 4(1 + \eta') + \eta \right]. \quad (5a)$$
\[ H_0 = \sum_{\alpha = a, b} \sum_{\mathbf{k}, \mathbf{p}} \left[ \epsilon^{(\alpha)}_\mathbf{k} \left( \mathbf{a}^{\dagger}_\mathbf{k} \mathbf{a}_\mathbf{p} + \mathbf{b}^{\dagger}_\mathbf{k} \mathbf{b}_\mathbf{p} \right) + \frac{\gamma k}{2} \left( \mathbf{a}^{\dagger}_\mathbf{k} \mathbf{a}_\mathbf{p} + \mathbf{b}^{\dagger}_\mathbf{k} \mathbf{b}_\mathbf{p} \right) \right] \]

where \( \gamma_k = \frac{4 \epsilon^{k,c}/2 \cos(k,a/2) \cos(k,a/2)}{4 + \eta + 1 + \gamma k} \)

For the second constant term in equation (13), the quantization point energy, which contributes to the classical ground state energy. The quasiparticle energy \( E^{(1,2)}_{\alpha, k} \) for both \( \alpha \) and \( \beta \) magnon branches are given by:

\[ E^{(1,2)}_{\alpha, k} = jS(4 + \eta) k_n^2 \omega^{(1,2)}_{\alpha, k} \]

Finally, we diagonalize the quadratic part \( H_0 \) by transforming the operators \( \mathbf{a}_\mathbf{k} \) and \( \mathbf{b}_\mathbf{k} \) to magnon operators \( \mathbf{a}_\mathbf{k} \) and \( \mathbf{b}_\mathbf{k} \) using the generalized Bogoliubov (BG) [20, 21] transformations:

\[ \mathbf{a}^{(1)}_k = \frac{1}{\sqrt{2}} \left[ \mathbf{c} \mathbf{a}^{(1)}_k + \mathbf{d}^{(1)}_k \right], \quad \mathbf{b}^{(2)}_k = \frac{1}{\sqrt{2}} \left[ \mathbf{c} \mathbf{b}^{(1)}_k + \mathbf{d}^{(2)}_k \right] \]
Here \( \mathbf{R}_i \) is the position vector of the \( i \)th unit cell and \( \tau_{\mu} \) are the positions of the four Cr-atoms in the unit cell. The positions of the Cr-atoms are respectively: Cr1: \( \tau_1 = (0, 0, c/2 - \delta/2) \), Cr2: \( \tau_2 = (a/2, a/2, \delta/2) \), Cr3: \( \tau_3 = (0, 0, c/2 + \delta/2) \), and Cr4: \( \tau_4 = (a/2, a/2, c - \delta/2) \) (see figure 1). Based on the experimental data \( c = 2a \) and \( \delta = c/3 \). The quantity measured in neutron-scattering experiment is the Fourier transform of the time-dependent spin-correlation function \( \mathcal{L}_s(\mathbf{k}, t) \),

\[
\mathcal{L}_s(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} \frac{dt}{2\pi} \mathcal{L}_s(\mathbf{k}, t)e^{-i\omega t}.
\]

The spins for each of the sublattices 1, 2, 3, 4 are defined in terms of the operators \( a \) and \( b \) as:

\[
S_m^{(1)} = S - a_m^{(1)} + a_m^{(1)}, \quad S_m^{(4)} = S - a_m^{(4)} + a_m^{(4)},
\]

\[
S_m^{(2)} = -S + b_m^{(2)} + b_m^{(2)}, \quad S_m^{(3)} = -S + b_m^{(3)} + b_m^{(3)},
\]

which after FT become:

\[
S_c^{(1)}(\mathbf{k}) = \sqrt{4N}\sum p q \delta(\mathbf{k} + \mathbf{p} - \mathbf{q})f_{ak}\alpha_{k}^{(1)}a_{q}^{(1)},
\]

\[
S_c^{(4)}(\mathbf{k}) = \sqrt{4N}\sum p q \delta(\mathbf{k} + \mathbf{p} - \mathbf{q})f_{ak}\alpha_{k}^{(4)}a_{q}^{(4)},
\]

\[
S_c^{(2)}(\mathbf{k}) = -\sqrt{4N}\sum p q \delta(\mathbf{k} + \mathbf{p} - \mathbf{q})f_{ak}b_{k}^{(2)}b_{q}^{(2)},
\]

\[
S_c^{(3)}(\mathbf{k}) = -\sqrt{4N}\sum p q \delta(\mathbf{k} + \mathbf{p} - \mathbf{q})f_{ak}b_{k}^{(3)}b_{q}^{(3)},
\]

where \( f_{ak} = e^{-i\mathbf{k}\mathbf{r}_a} \) takes into account the relative phases of the different magnetic atoms inside the unit cell. The total spin can now be written as:

\[
S_c(\mathbf{k}) = -\sqrt{4N}\sum p q \delta(\mathbf{k} + \mathbf{p} - \mathbf{q})\left[f_{ak}\alpha_{k}^{(1)}a_{q}^{(1)} + f_{ak}\alpha_{k}^{(4)}a_{q}^{(4)}\right]
\]

\[
- [f_{ak}b_{k}^{(2)}b_{q}^{(2)} + f_{ak}b_{k}^{(3)}b_{q}^{(3)}].
\]

Using BG transformations we express \( S_c(\mathbf{k}) \) in terms of the magnon operators \( \alpha \) and \( \beta \). The result is shown in the appendix.

There are 16 × 16 time-ordered Green’s functions (GFs) that arise from equation (17), of which only four contribute to LSSF. These four are defined as:

\[
\Pi_1(t) = -i\langle T\beta_{-p}^{(2)}(t)\alpha_{q}^{(1)}(t)\alpha_{p}^{(1)}(0)\beta_{-q}^{(1)}(0) \rangle,
\]

\[
\Pi_2(t) = -i\langle T\beta_{-p}^{(2)}(t)\alpha_{q}^{(2)}(t)\alpha_{p}^{(2)}(0)\beta_{-q}^{(2)}(0) \rangle,
\]

\[
\Pi_3(t) = -i\langle T\beta_{-p}^{(2)}(t)\alpha_{q}^{(1)}(t)\alpha_{p}^{(1)}(0)\beta_{-q}^{(1)}(0) \rangle,
\]

\[
\Pi_4(t) = -i\langle T\beta_{-p}^{(2)}(t)\alpha_{q}^{(2)}(t)\alpha_{p}^{(2)}(0)\beta_{-q}^{(2)}(0) \rangle.
\]

These GFs can be calculated easily in the leading order which does not include magnon–magnon interactions. The imaginary parts of these GFs are:

\[
-\frac{1}{\pi} \text{Im} \Pi_1^{(0)}(\omega) = \delta_{pq}\delta_{p}^{(1)}\delta_{q}^{(1)}\delta(\omega - \omega_{p}^{(1)} - \omega_{q}^{(1)}),
\]

\[
-\frac{1}{\pi} \text{Im} \Pi_2^{(0)}(\omega) = \delta_{pq}\delta_{p}^{(2)}\delta_{q}^{(2)}\delta(\omega - \omega_{p}^{(2)} - \omega_{q}^{(2)}),
\]

\[
-\frac{1}{\pi} \text{Im} \Pi_3^{(0)}(\omega) = \delta_{pq}\delta_{p}^{(1)}\delta_{q}^{(1)}\delta(\omega - \omega_{p}^{(2)} - \omega_{q}^{(1)}),
\]

\[
-\frac{1}{\pi} \text{Im} \Pi_4^{(0)}(\omega) = \delta_{pq}\delta_{p}^{(2)}\delta_{q}^{(2)}\delta(\omega - \omega_{p}^{(1)} - \omega_{q}^{(2)}).
\]

The imaginary parts correspond to the spectral densities of the correlation function \( \mathcal{L}_s(\mathbf{k}, \omega) \), which is:

\[
\mathcal{L}_s(\mathbf{k}, \omega) = \frac{1}{4N}\left[\|D_{kk+p}\|_{2}\right]^{2} + \sum_{p} \delta(\omega - \omega_{p}^{(1)} - \omega_{p}^{(1)})\left\|D_{kk+p}\right\|_{2}^{2} + \sum_{p} \delta(\omega - \omega_{p}^{(2)} - \omega_{p}^{(2)})\left\|D_{kk+p}\right\|_{2}^{2} + \sum_{p} \delta(\omega - \omega_{p}^{(1)} - \omega_{p}^{(2)})\left\|D_{kk+p}\right\|_{2}^{2},
\]

where the form factors \( D_{kk+p} \) are defined as:

\[
D_{kk+p}^{(2)} = \frac{1}{2}\left\langle f_{ak} + f_{ak}\right\rangle \left\langle C_{kk+p} + C_{kk+p}\right\rangle - \frac{1}{2}\left\langle f_{ak} + f_{ak}\right\rangle \left\langle C_{kk+p} + C_{kk+p}\right\rangle.
\]

\[
D_{kk+p}^{(3)} = \frac{1}{2}\left\langle f_{ak} + f_{ak}\right\rangle \left\langle C_{kk+p} + C_{kk+p}\right\rangle + \frac{1}{2}\left\langle f_{ak} + f_{ak}\right\rangle \left\langle C_{kk+p} + C_{kk+p}\right\rangle.
\]

\[
D_{kk+p}^{(4)} = \frac{1}{2}\left\langle f_{ak} + f_{ak}\right\rangle \left\langle C_{kk+p} + C_{kk+p}\right\rangle + \frac{1}{2}\left\langle f_{ak} + f_{ak}\right\rangle \left\langle C_{kk+p} + C_{kk+p}\right\rangle.
\]

\[
\mathcal{L}_s(\omega, \mathbf{q}) = \frac{1}{4\pi} \int_{0}^{\pi} \int_{0}^{2\pi} d\phi d\theta \sin \theta \mathcal{L}_s(\mathbf{k}, \omega).
\]
2.4. Two-magnon density of states (DOS)

The two-magnon density of states (DOS) for the four GFs are given as:

\[
\text{DOS}_{11}(k, \omega) = \sum_p \delta(\omega - \omega^{(1)}_p - \omega^{(1)}_{k+p}), \quad (28a)
\]

\[
\text{DOS}_{22}(k, \omega) = \sum_p \delta(\omega - \omega^{(2)}_p - \omega^{(2)}_{k+p}), \quad (28b)
\]

\[
\text{DOS}_{21}(k, \omega) = \sum_p \delta(\omega - \omega^{(2)}_p - \omega^{(1)}_{k+p}), \quad (28c)
\]

\[
\text{DOS}_{12}(k, \omega) = \sum_p \delta(\omega - \omega^{(1)}_p - \omega^{(2)}_{k+p}). \quad (28d)
\]

For \( \eta = 0 \), the \( \alpha \) and \( \beta \) branches are four-fold degenerate. In that case, \( \text{DOS}_{11}(k, \omega) = \text{DOS}_{22}(k, \omega) = \text{DOS}_{12}(k, \omega) = \text{DOS}_{21}(k, \omega) \). For \( \eta \neq 0 \) there are also symmetries between the four DOS’s. For example, with \( k_x = \pi/a, k_y = k_z = 0 \) or \( k_x = k_y = \pi/a, k_z = 0 \) or \( k_x = k_y = 0, k_z = \pi/c \), or \( k_x = k_y = k_z = \pi/c \), \( k_x = k_y = k_z = \pi/a = k_x = k_y = k_z = \pi/c \), \( \text{DOS}_{11} = \text{DOS}_{12} \) and \( \text{DOS}_{22} = \text{DOS}_{21} \).

3. Results: magnon dispersion, sublattice magnetization, and longitudinal spin–spin correlation function

3.1. Magnon energy dispersion

In figure 2 we show the magnon dispersions. Since the tetragonal unit cell contains four Cr spins (two up and two down) there will be four spin wave (SW) branches, two \( \alpha \) and two \( \beta \) branches, for each \( k \). In figure 2(a) we show the results for \( \eta = 0 \), when the two bilayers are decoupled. In this limit, branch 1 and branch 2 with frequencies \( \omega^{(1)}_k \) and \( \omega^{(2)}_k \) correspond to two bilayers and are degenerate. For \( \eta' = 0 \), the magnon is dispersionless (within LSWT) from \((\pi/a, 0, 0)\) to \((\pi/a, \pi/a, 0)\) to \((\pi/a, \pi/a, \pi/c)\). The absence of any \( k_z \) dependence is obvious as with \( \eta = 0 \) there is no coupling along the \( z \)-direction. However \((\pi/a, 0, 0)\) to \((\pi/a, \pi/a, 0)\) independence is peculiar to a \( \text{NN} \) 2D antiferromagnet. Introduction of magnon–magnon interaction or a nonzero \( \text{NN} \) exchange coupling \( \eta' \) brings in dispersion. For the known case with \( J = j' = 0 \) our system is a 2D antiferromagnet on a square lattice with only \( \text{NN} \) interaction \( j \). In that case for the region \((\pi, 0)\) to \((\pi/2, \pi/2)\) the dispersion is flat within...
linear spin wave theory and with $1/S$ corrections. However, with $1/S^2$ corrections, magnon energy at $(\pi/a, 0)$ is smaller than at $(\pi/2a, \pi/2a)$ (see [5, 17] for details). In our case if we include the 2nd order corrections we will find similar results. However, for our system $S = 3/2$, we expect the dip (if there is any) to be smaller. In the limit $J$ and $j$ are zero, $j'$ controls the magnon dispersion. The system consists of non-interacting 2D ferromagnetic sheets (square lattice with NN coupling $j'$) with dispersion $\omega_k = 4j'S[1 - (\cos(k_xa) + \cos(k_ya))/2]$, which for small $k$ takes the well-known result $\omega_k = 2j'S(ka)^2$.

In figure 2(b) we see the effect of introducing inter-bilayer AF coupling $\eta$ (for simplicity we chose $\eta' = 0$).
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Non-zero $\eta$ couples the intra-bilayer modes, leading to acoustic (Goldstone modes, $\omega_1^{(1)} \to 0$ as $k \to 0$) and optic modes ($\omega_2^{(2)} \to 4\sqrt{\eta}/(4+\eta)$ as $k \to 0$). The new $\alpha$ and $\beta$ modes are linear combinations of the old decoupled bilayer $\alpha$ and $\beta$ modes. The four-fold degenerate modes split into two modes along $(0,0,0)$ to $(\pi/a,0,0)$ and the zero frequency modes along $(0,0,0)$ to $(0,0,\pi/c)$ split into acoustic and optic modes. Interestingly, the modes along $(\pi/a,0,0)$ to $(\pi/a,\pi/a,0)$ to $(\pi/a,\pi/a,\pi/c)$ are dispersionless and four-fold degenerate. Finally, in figure 2(c), we show how the NNN ferromagnetic coupling introduces dispersion to these modes, but it does not remove the degeneracy.

3.2. Sublattice magnetization

The effect of QSF in quantum anti-ferromagnets in reducing the mean-field (or classical) value of the sublattice magnetization is well known [1]. This effect is strongest for small $S$ values and low dimensions. In our system, if we turn off the inter-bilayer AF exchange ($\eta = 0$), the system reduces to 2D quantum $S = 3/2$ system. In figure 3, we discuss the effect of QSF on the scaled magnetization $m = M_s/M_0$ where $M_0 = g\mu_B$. In figure 3(a), we see that for the decoupled bilayers ($\eta = 0$), QSF reduces $m$ from 1.5 (classical value) to 1.303 when $\eta = \eta' = 0$, a 13% reduction from the classical value. When one increases the strength of ferromagnetic NNN coupling the QSF effect is suppressed and the value increases towards its classical mean-field value. In figure 3(b) we show the effect of increasing $\eta'$ on the magnetization. Going back to figure 3(a) we find that for $\eta' = 0$, magnetization starts at 1.303 for $\eta = 0$ and increases to 1.406 at $\eta = 1.2$ and then decreases monotonically. The 2D QSF are suppressed as one introduces inter-bilayer coupling, even if it is antiferromagnetic; this is a 3D effect. But when $\eta$ increases further local dimer-related QSFs start to dominate and for large values of $\eta$ ($> \sim 6$), the magnetization is smaller than the 2D value [5, 15]. In general, we find that the effect of nonzero $\eta'$ is to suppress QSF effect on the magnetization.

3.3. Two-magnon density of states (DOS)

As we have discussed in section 2.3 the longitudinal spin-spin correlation function $L_s(k, \omega)$, which is directly probed in inelastic scattering measurements depends sensitively on the two-magnon DOS (see equations (25) and (28)). The latter were calculated for different $k$-values by numerically
evaluating equations (28a)–(28d). The sum over the internal three-dimensional momenta $\mathbf{p}$ is done on mesh grid of size $L \times L \times L$, where $L = 256$. A Gaussian function of width 0.075 (in units of energy) was used to broaden the $\delta$-function. As seen in equation (28), there are contributions from the two branches 1 and 2 in different combinations (11, 22, 21, 12). In figures 4(a)–(d), we present the two-magnon DOS($\mathbf{k}, \omega$) for $\eta = 0$. In the absence of inter-bilayer coupling the bilayer modes are degenerate. In addition, the $\alpha$ and $\beta$ modes are also degenerate due to symmetry. Thus we have a 4-fold degenerate magnon mode (see figure 2(a)) and in this case DOS$_{11}$($\mathbf{k}, \omega$) = DOS$_{22}$($\mathbf{k}, \omega$) = DOS$_{21}$($\mathbf{k}, \omega$) = DOS$_{12}$($\mathbf{k}, \omega$) = DOS($\mathbf{k}, \omega$)/4. The peak occurs at 8$\sqrt{S}$ whereas the peak in the DOS of one-magnon excitation is at 4$\sqrt{S}$. This factor of 2 is a result of linear spin wave (LSW) approximation. As seen in figure 4, DOS($\mathbf{k}, \omega$) is independent of $k_z$ as it should be. The single peak structure seen for $\mathbf{k} = 0$ (Γ-point) at 8$\sqrt{S}$ develops two and three peak structures as one goes away from the Γ-point. This is seen clearly in figure 4(d). At the Brillouin zone boundary ($k_x = k_y = \pi/\alpha$), the peak appears below the band edge, at $\sim 6.8\sqrt{S}$ and there is a saddle point at 8$\sqrt{S}$.

Next, we discuss the case when inter-bilayer coupling ($J$) is nonzero. Since in the Cr$_2$XO$_6$ systems, $J$ is much larger than the intra-bilayer coupling ($j$) we choose $\eta = 10$ and still keep $\eta' = 0$. For simplicity. In figures 5 and 6, we plot the ($\mathbf{k}, \omega$) dependence of DOS$_{11}$, DOS$_{22}$, DOS$_{21}$, and DOS$_{12}$. In figure 5, we choose $k_z = 0$ and study the ($k_x, k_y$) dependence. The dominant feature is a narrow peak at energy $\sim 19.5\sqrt{S}$, whereas the single magnon branches lie between 0 and 13$\sqrt{S}$ (figure 2(c)). The peak comes from the flat part of the single magnon dispersion near 9.75$\sqrt{S}$. Since DOS$_{11}$ comes only from the acoustic branch it extends below the peak. On the other hand, DOS$_{22}$ comes only from the optic branch, it extends above the peak. The mixed contributions DOS$_{12}$ and DOS$_{21}$ have strong $\mathbf{k}$-dependence. It is interesting to note that for $k_x = k_y = k_z = 0$ (figures 5(c1) and (d1)), DOS$_{21} = DOS_{12}$. This is because at the Γ-point, equations (28e) and (28d) are identical. On the other hand, for $k_x = k_y = \pi/\alpha, k_z = 0$ (figures 5(a3)–(d3)), DOS$_{11} = DOS_{12}$ and DOS$_{22} = DOS_{21}$. In figure 6, we show the effect of $k_z$ on all four DOS. In contrast to the $\eta = 0$ case, for $\eta = 10$ DOS depends on $k_z$. For example, when $k_z = \pi/\alpha$, all the four DOS differ from their corresponding structures when $k_z = k_y = k_x = 0$ (figures 6(a3)–(d3)). Instead they are identical to the spectra when $k_x = \pi/\alpha, k_y = k_z = 0$. Interestingly for $k_x = k_y = 0, k_z = \pi/\alpha$, and $k_z = \pi/\alpha$, $k_z = \pi/\alpha$, and $k_z = \pi/\alpha$.
Figure 7. Longitudinal spin–spin correlation, $L_s(k, \omega)$ for different values of $k$ is plotted for $\eta = 10, \eta' = 0$ in (a)–(i). Note that for $k_x = k_y = \pi/a, k_z = 0$, $L_s(k, \omega) = 0$ (c). A narrow peak is seen around $19.5jS$.

Figure 8. Longitudinal spin–spin correlation $L_s(k, \omega)$ and the sum of the density of states of four magnon branches are plotted for $\eta = 10, \eta' = 0$ and two different values of $k = 0$ (a)–(b) and $k_x = k_y = k_c = \pi$ (c)–(d). The plots display the effects of form factors in $L_s(k, \omega)$. 
DOS$_{11}$ = DOS$_{12}$ and DOS$_{22}$ = DOS$_{23}$. These symmetries are mentioned in section 2.4. But for $k_x = k_y = 0, k_z = \pi/2c$ (figures 6(a2)–(d2)) all DOS’s are different.

3.4. Longitudinal spin–spin correlation function (LSSF)

In figures 7(a)–(i), we show the $k$-dependence of LSSF $\langle \mathcal{L}_s(k, \omega) \rangle$. As seen in equation (25), contributions from different two-magnon excitations get weighted by the associated form factors $D_{ij}^\eta k, k+p$. This leads to different energy dependence of LSSF compared to that of the total two-magnon DOS. For example, as seen in figure 7(e), for $k_x = k_y = \pi/a, k_z = 0$, LSSF vanishes and becomes nonzero as we increase $k_z$. In figure 8 we show both $\mathcal{L}_s(k, \omega)$ and the sum of the four DOS$_{ij}(k, \omega)$ for $k = 0$ and $k_{xR} = k_{yR} = k_{zR} = \pi$. For both the $k$ values the effect of the form factors is very significant.

Finally, we plot the angular average of $\mathcal{L}_s(k, \omega)$ for different magnitudes of $k$ in figure 9. For these plots, equation (27) was numerically evaluated by summing over the angles $\theta, \phi$. For each $\omega$ about 270 million points were evaluated. This is what is observed in a inelastic neutron scattering experiment from a powder sample. The generic feature is a narrow peak seen at 19.5$jS$—the intensity scale of this peak grows with the magnitude of $k$.

4. Conclusions

In this paper we have discussed the magnon dispersion, two-magnon density of states, and longitudinal spin–spin correlation function in the leading order approximation, in systems described by coupled bilayers where both intra ($J$) and inter-bilayer ($J$) nearest neighbor (NN) couplings are antiferromagnetic. Although the particular spin system we have studied is Cr$_2$TeO$_6$, which contain Cr$^{3+}$ ions with spin-3/2, our formalism is general and can be applied to any spin-$S$.

We have also investigated how a small intra-bilayer NNN ferromagnetic coupling ($J'$) affects the above properties.

One of the interesting features of our calculation is the non-monotonic $\eta = J'/J$ dependence of the reduced sublattice magnetization $m_a = M_a/M_0$. In the classical limit (mean-field) $m_a = 1.5$ for $S = 3/2$. When $\eta = 0$, that is in the decoupled bilayer limit, quantum spin fluctuations (QSF) reduce $m_a$ to 1.303 but as we increase $\eta$, $m_a$ first increases and equals to 1.406 when $\eta \sim 1$ and then decreases and becomes smaller than the decoupled bilayer value when $\eta > 6$. For Cr$_2$TeO$_6$, $\eta$ is estimated to be $\sim 10$ (interacting quantum spin-dimer limit) and $m_a$ differs substantially from its classical value. The presence of nonzero $J'$, QSF effects are suppressed.

Due to the quasi 2D geometry and local (NN) inter bilayer AF coupling, magnon dispersion shows a flat region over a large part of the 2D Brillouin zone. This results in a sharp peak in the
one-magnon density of states (DOS) near $\sim 10 J S$ for $\eta = 10$, not at the band maximum which occurs at $\sim 12 J S$, but closer. The two-magnon DOS also shows sharp peaked structure for most of the values of the total momentum $k$. The two-magnon peak appears $\sim 1.4(4 + \eta) J S = 19.5 J S$. The longitudinal spin-spin correlation function, $L_s(k, \omega)$ function depends both on the two-magnon spectrum and the Bogoliubov amplitudes and phases. In fact, for certain $k$, $L_s(k, \omega)$ vanishes even if the two-magnon DOS does not. Experiments in single crystal samples should test the results of this theoretical predictions. Unfortunately, large single crystal samples of $Cr_2TeO_6$ are not available, most of the neutron experiments are done in powder samples. In this case, one measures the energy dependence of $L_s(k, \omega)$ averaged over the angular components of $k$, that is for different magnitudes of $|k| = Q$. Again one finds a peak structure for most of the values of $Q$, again at 19.5J$S$, but the main effect of increasing $Q$ is seen in the increase of the total intensity. These predictions can be checked experimentally.

In a subsequent paper, we will discuss the case when intrabilayer exchange is ferromagnetic (e.g. in $Cr_2WO_6$ and $Cr_2MoO_6$) and compare this with the present case. Here the QSF effects are absent in the limit $\eta = 0$, but start to increase as one increases $\eta$.

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Appendix. Total spin $S_z$ in terms of $\alpha$ and $\beta$ magnons

$$S_z(k) = \frac{1}{2} \frac{1}{\sqrt{4N}} \sum_{p,q} \delta(k + p - q) \times \left\{ \begin{array}{l} \{ f_{1k} + f_{1k} | C_{p1} C_{1q} - f_{1k} + f_{1k} | C_{p1} C_{1q} \} c_{p}^{(2) \dagger} c_{q}^{(1)} \\ + \{ f_{1k} - f_{1k} | C_{p2} C_{1q} - f_{1k} - f_{1k} | C_{p2} C_{1q} \} c_{p}^{(1) \dagger} c_{q}^{(2)} \\ + \{ f_{2k} - f_{2k} | C_{p1} C_{2q} - f_{2k} - f_{2k} | C_{p1} C_{2q} \} c_{p}^{(2) \dagger} c_{q}^{(1)} \\ - \{ f_{2k} + f_{2k} | C_{p2} C_{2q} - f_{2k} + f_{2k} | C_{p2} C_{2q} \} c_{p}^{(1) \dagger} c_{q}^{(2)} \\ \end{array} \right\}$$

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