Effects of bi-quadratic and interlayer exchange coupling on layered triangular lattice antiferromagnets

M. Li, M. L. Plumer, and G. Quirion  
Department of Physics and Physical Oceanography,  
Memorial University, St. Johns, Newfoundland, Canada A1B 3X7

The magnetic field evolution of ground spin states of the stacked planar triangular antiferromagnet with antiferromagnetic interlayer interaction $J_c$ is explored using a 3D classical Heisenberg model. A bi-quadratic coupling is also used to mimic the effect of spin fluctuations [1] which are known to stabilize the magnetization plateau. A single ion anisotropy is included and states with a magnetic field applied in the ab plane and along the c axis are determined. For $\mathbf{H} \parallel ab$-plane, an additional new state, in contrast to 2D model [1], is obtained with weak interlayer interaction, while the magnetization plateau vanishes at large $J_c$ and other new states with z components of spins emerge. For $\mathbf{H} \parallel c$-axis, an extra state, compared with 2D model, is obtained with a weak interlayer interaction. When $J_c$ is large enough, only the state corresponding to the Umbrella phase in 2D model exits.

I. INTRODUCTION

The two-dimensional (2D) geometrically frustrated triangular lattice antiferromagnets (TLAFs) have been widely studied in a variety of compounds over recent decades. These systems are known to display various exotic magnetic states due to the extensive degeneracy associated with magnetic frustration on a triangular lattice [2–4]. In particular, one of these exotic states, associated with magnetic frustration on a triangular lattice, is the magnetization plateau in many quasi-2D TLAFs with easy-plane anisotropy at a value of 1/3 of saturation of a magnetization plateau in many quasi-2D TLAFs [2–4]. In particular, one of these exotic states, associated with magnetic frustration on a triangular lattice.

We first describe the 3D Heisenberg model representing a two-layer triangular lattice in Sec. II. The magnetization is believed to be stabilized by thermal/quantum spin fluctuations [1, 17–21]. This conclusion is well supported by 2D quantum numerical studies [1, 17–20] and Monte Carlo simulation [21, 22] which successfully reproduce the observed phase sequence (120° state at zero field, Y state at low field, ud state at intermediate field, and V state at high field) for a magnetic field applied in the basal plane. However, a new phase between the ud state and the V state was recently proposed by D. Yamamoto et al. [23, 24] who took into consideration the interlayer exchange coupling. This finding could account for the magnetization anomaly observed near 3/5 $M_s$ in $\text{Ba}_3\text{CoSb}_2\text{O}_9$ with $\mathbf{H} \parallel ab$-plane [10], and it indicates the importance of the interlayer interaction, even when it is weak compared to the intralayer interaction. So far, the effect of the interlayer interaction in TLAFs has been limited to a few studies [23, 24], hence there is still a need to explore how the magnetic properties evolve as a function of the interlayer exchange coupling.

In this work, the effect of the antiferromagnetic interlayer interaction of the layered easy-plane TLAFs is explored using a 3D classical Heisenberg model with bi-quadratic exchange coupling ($\gamma$) between nearest-neighbour ions within the basal plane. The bi-quadratic coupling ($\gamma$) plays an important role as it mimics the effect of spin fluctuations [1, 25] which are known to stabilize the magnetization plateau (ud state). The single ion anisotropy ($D$) is also considered and the magnetic states with $\mathbf{H} \parallel a$-axis and c-axis are determined and compared with existing experimental magnetic phase diagrams.

Without the interlayer interaction ($J_c = 0$), the model accounts for the 120° state, Y state, ud state, and V state with $\mathbf{H} \parallel a$-axis as obtained from 2D Monte Carlo simulations [21]. Our minimal classical model allows us to study in detail how the magnetic properties of TLAFs, with easy-plane anisotropy ($D = 0.05$) and bi-quadratic coupling ($\gamma = -0.05$), evolve as a function of the interlayer coupling parameter ($J_c$). Moreover, consistent with Ref. [24] and in contrast to the 2D model [1], a new state between the ud and the V phase is observed for weak interlayer interaction. The results also show that the width of the magnetization plateau decreases as $J_c$ increases, and vanishes at $J_c \sim 0.1$. Within a small interlayer coupling range ($0.15 < J_c < 0.16$), two new states with a small alternating z-component of spins emerge. For $\mathbf{H} \parallel c$-axis, the Umbrella (U) state and $V_z$ state, observed in experiments [10–12], are reproduced without the interlayer interaction ($J_c = 0$), while a state between the U and $V_z$ states is obtained with weak interlayer interaction and disappears at a medium value of $J_c (\sim 0.06)$. When $J_c$ is large ($> 0.2$), only the state corresponding to the Umbrella phase exits.

The remainder of this paper is organized as follows. We first describe the 3D Heisenberg model representing a two-layer triangular lattice in Sec. II. The magnetization obtained for that effective model, along with the spin configurations associated with the different magnetic orders, are shown in Sec. III and Sec. IV. For $\mathbf{H} \parallel ab$-plane, the results with $0 \leq J_c \leq 0.21$ are presented in Sec. III, while the results for $\mathbf{H} \parallel c$-axis with $0 \leq J_c \leq 0.24$ are presented in Sec. IV. Finally, summary and discussion are presented in Sec. V.
II. MODEL: TWO-LAYER TRIANGULAR LATTICE

The two-layer equilateral triangular lattice system is shown in Fig. 1, where each magnetic layer is comprised of three sublattices with spins at triangle vertices. In this model, Eq. 1 represents the energy per plane where only the intralayer exchange coupling constant \((J)\) between the nearest neighbouring spins is taken into consideration. The Heisenberg Hamiltonian per layer can be written as

\[
E_\alpha = J \sum_{i \neq j} S_{i\alpha} \cdot S_{j\alpha} - \frac{1}{3} H \sum_i S_{i\alpha} + \gamma \sum_{i \neq j} (S_{i\alpha} \cdot S_{j\alpha})^2 + D \sum_i S_{i\alpha z}^2,
\]

where \(E_\alpha\) denotes the energy of layer \(\alpha\) (stacked along the \(c\)-axis), \(i\) denotes one of the three magnetic ions at a triangle vertex. The first term corresponds to the intralayer coupling energy \((J)\), the second term is Zeeman energy, the third term is the bi-quadratic coupling energy \((\gamma)\) which stabilizes the magnetization plateau, and the last term is the single ion anisotropy energy \((D)\). Furthermore, taking into consideration the interlayer nearest neighbouring exchange coupling \((J_c)\), the total Hamiltonian for two layers (6 spins) can be written as

\[
E = \frac{1}{2} \sum_\alpha E_\alpha + J_c \sum_{\alpha \neq \beta} \sum_i S_{i\alpha} \cdot S_{i\beta},
\]

with the magnetization per site given by

\[
m = \frac{1}{6} \sum_\alpha \sum_i S_{i\alpha}.
\]

Here the spins are written as 3D vectors described by the angles \(\phi_{i\alpha}\) and \(\theta_{i\alpha}\):

\[
S_{i\alpha} = (\cos \phi_{i\alpha} \sin \theta_{i\alpha}, \sin \phi_{i\alpha} \sin \theta_{i\alpha}, \cos \theta_{i\alpha}).
\]

Minimizing the Hamiltonian (Eq. 2) relative to \(\phi_{i\alpha}\) and \(\theta_{i\alpha}\), the magnetization and the spin configurations are obtained for different parameter and field values.

III. \(H \parallel ab\)-PLANE

We first present results obtained using the effective Hamiltonian Eq. 2 by setting \(J_c = 0\) with \(H \parallel x\)-axis.

In this work, the coefficient of the antiferromagnetic intralayer exchange coupling is set to \(J = 1\), while the effect of the antiferromagnetic interlayer coupling \((0 < J_c < 0.24)\) is explored. In order to account for the collinear spin configuration (uud state), the bi-quadratic coupling coefficient \(\gamma\) must be negative [1]. Here \(\gamma\) is set to -0.05 in order to obtain a magnetization plateau width consistent with some experimental results [10, 11]. Since the single ion anisotropy and the exchange anisotropy have the same effect in the case of easy-plane anisotropy with the field in the plane [26], we set \(D = 0.05\) close to the experimental value of the exchange anisotropy as determined for Ba3CoSb2O9 [10].

![FIG. 1. Two-layer triangular lattice with intralayer interaction \(J\) and interlayer interaction \(J_c\).](image)
where the $x$-axis is in the $ab$-plane. The magnetization curve $m_x$ is shown in Fig. 2, while the different spin configurations are presented in Fig. 3. For $H = 0$, the ground state corresponds to the 120° spin structure. The so-called Y state is stabilized at low fields, the magnetization plateau associated with the up-up-down (uud) state follows and then the V state before the magnetization saturation is obtained at high fields. The results presented in Fig. 2 and Fig. 3, obtained using the classical 2D effective Hamiltonian Eq. 1 are in good agreement with previous results based on 2D Monte Carlo simulations [21] and 2D quantum models [1, 17–20, 23, 24]. Considering that the effective Hamiltonian (Eq. 1) adequately describes the magnetic properties of 2D TLAF, it can be easily modified (Eq. 2) in order to explore the properties of 3D TLAF with easy-plane anisotropy.

In Fig. 4a, we present the magnetization curves $m_x$ for different values of the antiferromagnetic interlayer coupling coefficient $J_c$ while Fig. 4b shows the derivatives of the magnetizations in order to identify the critical fields. The spin configurations of the different states are shown in Fig. 5.

In the range $0 < J_c < 0.1$, compared with the 2D model magnetization (Fig. 2), one additional C (canted) phase is obtained between the plateau and the V phase, consistent with results published in Ref. [24]. The spin configurations for the quasi-2D Y state, uud state, C state and V state are sketched in Fig. 5. Furthermore, as shown in Fig. 6, the Y and V phases have a small none-zero $y$-component (perpendicular to the $x$-axis) of the magnetization which alternates from one plane to the next. This figure also clearly illustrates the first order character of the phase transition between the C and V states. Consequently, we conclude that interlayer interaction, even weak compared with the intralayer exchange interaction, stabilizes a new state and modifies the spin configurations relative to the 2D system.

For $J_c$ larger than 0.1, as shown in Fig. 4a, the magnetization plateau disappears. Therefore, in the range $0.1 < J_c < 0.14$, the system transforms directly from the Y state to the C state via a first-order transition indicated by a jump on the magnetization in Fig. 4a.

For $0.14 < J_c < 0.16$, two new states, W state and V', are obtained whose spin configurations are presented in Fig. 5. In the W state, each layer develops a small $z$-component of the magnetization which alternates from

![FIG. 4](image-url)  
FIG. 4. a) Magnetization process of the two-layer TLAFs with $\gamma = -0.05$, $D = 0.05$, and $H \parallel x$-axis. b) The first derivatives of the magnetizations assisting to identify phase transitions. The curves are shifted vertically for clarity.

![FIG. 5](image-url)  
FIG. 5. Spin configurations of TLAFs with interlayer interaction and $H \parallel x$-axis in different phases. Black solid arrows (A, B, C) and red dotted arrows ($A'$, $B'$, $C'$) represent spins at the sublattice vertices in different layers, respectively. (See Fig. 1)

![FIG. 6](image-url)  
FIG. 6. The $y$-component magnetization in each layer with $J_c = 0.03$, $\gamma = -0.05$, and $D = 0.05$. 

one layer to the next (see Fig. 7), while the configuration in the xy-plane forms a W shape and a Y shape in different layers, respectively. In the V′ state, while the spin configuration in the xy-plane is identical to that of the V phase, two spins in each layer have a small z-component in opposite directions, maintaining the z-component magnetization per plane to zero. Furthermore, the derivative of the magnetization (Fig. 4b) shows that Y→V′, Y→W, Y→C, and C→V correspond to first order phase transitions.

To explain the appearance a spin polarization normal to the ab-plane in W and V′ states, we compare the energy of the anisotropic term (Ea) and the interlayer interaction (Ec), which involve the z component of the spins (see Eq. 5).

\[
E_a = \frac{D}{2} (S_{A z}^2 + S_{A z}^2 + S_{B z}^2 + S_{B z}^2 + S_{C z}^2 + S_{C z}^2)
\]

\[
E_c = J_z (S_{A z} S_{A z} + S_{B z} S_{B z} + S_{C z} S_{C z})
\]

When \( J_z \) is zero or very small, the energy is minimized by having no z component. However, when \( J_z \) is large enough (\( \sim 0.14 \)), the lowest energy can be reduced by having anti-parallel z component nearest neighbour interlayer spins.

We present in Fig. 8 the \( H_z-J_z \) phase diagram for the two-layer TLAFs. The dashed and solid lines indicate first and second order phase transitions, respectively. As shown, the range of the magnetization plateau (ud state) decreases with increasing interlayer interaction and vanishes at \( J_z = 0.1 \). The C state is only obtained with a weak interlayer interaction and disappears at \( J_z = 0.146 \). When \( 0.14 < J_z < 0.16 \), the W and V′ states, which have a spin z component, are stabilized. For \( J_z > 0.16 \), only the V′ state exists between the Y and V states. Therefore, we can conclude that the interlayer interaction plays an important role in the magnetization process of easy-plane TLAFs.

IV. H \( || \gamma \)-AXIS

With \( H \parallel \gamma \parallel \hat{c} \), the magnetization curve \( m_z \) obtained from the one-layer model \( (J_z = 0) \) is shown in Fig. 9, while the different spin configurations are presented in Fig. 10. The Umbrella state (U state) and Vz state, associated with the observation of a first order phase transition in experiments [10–12], are stabilized at low fields and high fields, respectively. Furthermore, the Vz state is also characterized by a small none-zero magnetization perpendicular to the z-axis (shown in the inset of Fig. 9).

In Fig. 11, we present magnetization curves \( m_z \) for different values of the antiferromagnetic interlayer coupling coefficient \( J_z \). For a weak interlayer interaction \( (0 < J_z < 0.06) \), compared with the 2D model magneti-
zation (Fig. 9), one extra $C_z$ (canted) state is obtained between the $U$ state and the $V_z$ state. The spin configurations for the quasi-2D $U$ state, $V_z$ state and $C_z$ state are sketched in Fig. 12. Different from the 2D model, the two-layer $V_z$ state has no net $xy$-component magnetization perpendicular to the $z$-axis, while the $C_z$ state has a small transverse magnetization as shown in the inset of Fig. 11. In the range $0.06 < J_c < 0.2$, U and $V_z$ states are obtained and represented by the blue lines in Fig. 11. For $J_c > 0.2$, only the $U$ state exists before the magnetization saturation, indicated by the black lines. The resulting $H_z$-$J_c$ phase diagram for the two-layer TLAFs is presented in Fig. 13 with dashed and solid lines representing first and second order phase transitions, respectively.

**V. SUMMARY AND DISCUSSION**

The ground state magnetization processes of TLAFs are calculated for both $H$ in the $ab$-plane and $H || c$-axis using a two-layer classical Heisenberg model with the single ion anisotropy ($D$) and the bi-quadratic exchange coupling ($\gamma$). To study realistic 3D TLAFs, we explored the effect of the antiferromagnetic interlayer interaction ($J_c$). Results with $H || ab$-plane and $H || c$-axis show that the interlayer interaction plays a role for stabilizing the additional state (C state), consistent with Ref. [24]. This additional state could account for the magnetization anomaly observed near $3/5 M_s$ in Ba$_3$CoSb$_2$O$_9$ with $H$ in the $ab$-plane [10]. Other new states, not previously reported, are also observed with higher values of the interlayer interaction. The range of field, over which all states are stabilized, depends on the value of $J_c$. Moreover, the spin configurations of the $W$ and $V'$ states show the appearance of small $z$ components due to the interlayer interaction competing with the single ion anisotropy. Due
to this competition, the system exhibits complex magnetization processes, especially when $0.144 < J_c < 0.146$ (see Fig. 8). It should be noticed that the values of $J_c$ for obtaining the W and V's states are large compared with the small $J$, with which the magnetization plateau survives, but still about one order of magnitude smaller than the intralayer interaction $J$. Therefore, this model can still be considered to be quasi-2D, with the interlayer exchange coupling playing an important role. We believe that a detailed analysis of relevant experimental results on existing and yet to be discovered TLAFs may benefit from the results present here.

VI. ACKNOWLEDGMENTS

The authors wish to acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada (NSERC).