Magnon bands in twisted bilayer honeycomb quantum magnets

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We study the magnon bands of twisted bilayer honeycomb quantum magnets using linear spin wave theory. Although the interlayer coupling can be ferromagnetic or antiferromagnetic, we keep the intralayer one ferromagnetic to avoid possible frustration. For the interlayer ferromagnetic case, we find the magnon bands have similar features with the corresponding electronic energy spectrums. Although the linear dispersions near the Dirac points are preserved in the magnon bands of twisted bilayer magnets, their slopes are reduced with the decrease of the twist angles. On the other hand, the interlayer antiferromagnetic couplings generate quite different magnon spectra. The two single-layered magnon spectra are usually decoupled due to the opposite orientations of the spins in the two layers. We also develop a low-energy continuous theory for very small twist angles, which has been verified to fit well with the exact tight-binding calculations. Our results may be experimentally observed due to the rapid progress in two-dimensional magnetic materials.

Keywords: magnon bands, twisted bilayer, quantum magnets, linear spin wave theory

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1. Introduction

The recent discovery of the correlated insulator and unconventional superconductivity in twisted bilayer graphene has attracted intense interests.[1–8] By rotating the layers to a small angle, a misalignment induced in the bilayer system introduces a long-period moiré superlattice. Such a superlattice therefore modulates the electronic structure, and leads to nearly flat band at the magic angles. These bands become so narrow that the electron-electron correlations dominate over kinetic energy, giving rise to the above correlated quantum phases.[9] Since twisted bilayer graphene is relatively simple and highly tunable, it is anticipated that this system can serve as an ideal platform to investigate the strongly-correlated physics.

Moiré superlattices have also been employed to engineer flat bands in other two-dimensional (2D) materials, such as twisted double-bilayer graphene, trilayer graphene, and twisted bilayer of the transition metal dichalcogenide, where rich correlation phenomena have been revealed.[10–19] While research has been primarily centered on twisted electronic systems, there appear theoretical studies addressing the remarkable physical properties in twisted Kitaev bilayers,[20] and twisted optical lattices.[21,22] It is accepting that twisting is a simple and general approach to creating exotic quantum matter.

Recently rapid progress in 2D magnetic atomic crystals has been made.[23–27] Various monolayer and multilayer van der Waals (vdW) magnetic materials have been discovered, and different kinds of ferromagnetic (FM) and antiferromagnetic (AFM) orders have been observed.[28–36] The 2D vdW magnetic materials have a huge potential to create novel functional devices, and have important applications in the next-generation nanoelectronics.[37–42] Of course, they are also important in fundamental research. The physics can be described by a generalized Heisenberg spin Hamiltonian, and a broad range of parameter regimes can be realized in a rich variety of materials. All three spin Hamiltonians, i.e., Ising, XY, and Heisenberg models, can be recovered in specific limits.[43–46] Besides, external perturbations, such as gating, strain, etc., can further tune the range of model parameters.[47–51] These make the magnetic 2D materials into an ideal platform to examine the well-established theories[52–54] and explore new quantum phases.[55–57]
Motivated by these advances in the studies of the twisted 2D materials and 2D magnetic materials, we study how the twist alters the magnon bands in twisted bilayer honeycomb quantum magnets here. We keep the intralayer FM coupling, and study the magnon bands with interlayer FM and AFM couplings in AA(AB)-stacked and twisted bilayer quantum magnets. Our study is closely related to the rapid experimental progress in two-dimensional magnetic materials.

2. AA-stacked honeycomb magnets

Twisted bilayers are characterized by non-zero angle \( \theta \) between two honeycomb layers. Since the twist may be performed based on AA-stacked bilayer, we start from the spin-\( \frac{1}{2} \) Heisenberg model on this geometry, which can be written as

\[
H = -J \sum_{(ij),\ell} \mathbf{S}_{i,\ell} \cdot \mathbf{S}_{j,\ell} - J_\perp \sum_{(ij)} \mathbf{S}_{i,2} \cdot \mathbf{S}_{j,1},
\]

where \( \mathbf{S}_{i,\ell} = (S^x_{i,\ell}, S^y_{i,\ell}, S^z_{i,\ell}) \) is the spin-1/2 operator at site \( i \) in layer \( \ell = 1, 2 \); the summation runs over nearest-neighbor sites \( (ij) \); \( J, J_\perp \) is the intralayer (interlayer) coupling constant, and we first consider the FM case, i.e., \( J > 0 \) and \( J_\perp > 0 \).

Using Holstein–Primakoff (HP) transformation, the spin operators are expressed in term of bosonic creation and annihilation operators. In the FM case, the transformation in the linear spin-wave theory is defined as

\[
S^+ = \sqrt{2S} a_i, \quad S^- = -a_i^\dagger a_i.
\]

After ignoring a constant and four-operator terms, the resulting bosonic tight binding Hamiltonian becomes

\[
\mathcal{H}^{\text{AA}}_{\text{FM}} = -J \sum_{(ij),\ell} \frac{1}{2} (a_i^\dagger a_j + a_i^\dagger a_j a_i^\dagger a_j) \\
- J_\perp \sum_{i,\ell} \frac{1}{2} (a_i^\dagger a_i a_i^\dagger a_\ell + a_i^\dagger a_i a_i^\dagger a_\ell) \\
+ \frac{3J + J_\perp}{2} \sum_{i,\ell} a_i^\dagger a_i a_i^\dagger a_\ell.
\]

Performing a Fourier transformation, the above Hamiltonian writes as \( H = \sum \mathbf{k} \psi^\dagger \mathcal{H}^{\text{FM}}(\mathbf{k}) \psi \), where

\[
\psi_k = \{ a_{A,1}(k), a_{B,1}(k), a_{A,2}(k), a_{B,2}(k) \}^T
\]

is the basis, and

\[
\mathcal{H}^{\text{AA}}_{\text{FM}}(k) = \mathcal{H}^{\text{AA}}_{0}(k) + \frac{3J + J_\perp}{2},
\]

with

\[
\mathcal{H}^{\text{AA}}_0(k) = \begin{pmatrix}
0 & f(k) & -J_\perp/2 & 0 \\
-f^*(k) & 0 & 0 & -J_\perp/2 \\
-J_\perp/2 & 0 & 0 & f(k) \\
0 & -J_\perp/2 & f^*(k) & 0
\end{pmatrix},
\]

and \( f(k) = -(J/2)(1 + e^{-ik_1 a_1} + e^{-ik_2 a_2}) \) \( [a_1 = (\sqrt{3}/2, 0), a_2 = (\sqrt{3}/2, 3/2) \) the primitive vectors].

With a unitary transformation \( \psi_k = U(k) \phi_k \), the above Hamiltonian is directly diagonalized, and the spectrum contains four branches: \( (3J + J_\perp)/2 \pm J_\perp \pm |f(k)| \). The magnon band structure is shown in Fig. 1. Compared to that of single-layer honeycomb magnet, the Dirac points are shifted upward and downward by \( J_\perp/2 \), which is exactly the same as that of AA-stacked graphene except for an overall translation of \( 3J + J_\perp/2 \).

Now we turn to consider the case with interlayer AFM exchange, i.e., \( J_\perp < 0 \), while the intralayer coupling remains FM coupling. The following magnetic configuration is assumed: the spins in layer 1 (2) are aligned, and the spectrum contains four branches: \( (3J + J_\perp)/2 \pm J_\perp \pm |f(k)| \). The magnon band structure is shown in Fig. 1. Compared to that of single-layer honeycomb magnet, the Dirac points are shifted upward and downward by \( J_\perp/2 \), which is exactly the same as that of AA-stacked graphene except for an overall translation of \( 3J + J_\perp/2 \).

Fig. 1. (a) The magnon band structure of AA-stacked FM bilayer. (b) The magnon bands along the high symmetry points in the first Brillouin zone, which are shown in the inset. The parameter is \( J_\perp/J = 0.17 \).

The resulting bosonic tight binding Hamiltonian becomes

\[
\mathcal{H}^{\text{AA}}_{\text{AFM}} = -J \sum_{(ij),\ell} \frac{1}{2} (a_i^\dagger a_j + a_i^\dagger a_j a_i^\dagger a_j) \\
- J_\perp \sum_{i,\ell} \frac{1}{2} (a_{i,1} a_{i,2} + a_{i,1}^\dagger a_{i,2}^\dagger) \\
+ \frac{3J - J_\perp}{2} \sum_{i,\ell} a_i^\dagger a_i a_i^\dagger a_\ell.
\]

Under the basis

\[
\psi_k = \{ a_{A,1}(k), a_{B,1}(k), a_{A,2}^\dagger (-k), a_{B,2}^\dagger (-k) \}^T,
\]

\[
077505-2
\]
the above Hamiltonian in the momentum space can be written as
\[ \mathcal{H}_{\text{AFM}}^{\text{AA}}(k) = \mathcal{H}_{0}^{\text{AA}}(k) + \frac{3J - J_{\perp}}{2}, \quad (J_{\perp} < 0). \quad (8) \]

We use Bogoliubov transformation \( \psi_{k} = U(k)\phi_{k} \) to diagonalize the above Hamiltonian
\[ U(k)^{\dagger}\mathcal{H}_{\text{AFM}}^{\text{AA}}(k)U(k) = D, \]
with \( D \) is a diagonal matrix. The transformation satisfies \( U(k)^{\dagger}s_{z}U(k) = s_{z} \) to maintain the commutation relation of bosons. Then we have \( s_{z}\mathcal{H}_{\text{AFM}}^{\text{AA}}(k)U(k) = U(k)s_{z}D \), which means the Bogoliubov transformation \( U(k) \) is the eigenvector of \( s_{z}\mathcal{H}_{\text{AFM}}^{\text{AA}} \) with the eigenvalue \( s_{z}D \). Thus we obtain the magnon spectrum
\[ E_{k}^{\pm} = \sqrt{\left| f(k) \right|^{2} + \frac{3J - J_{\perp}}{2} - \left( \frac{J_{\perp}}{2} \right)^{2}}, \]
each of which is two-fold degenerate. For small \( J_{\perp} \) and near the Dirac points,
\[ E_{k}^{\pm} \sim \left| f(k) \right| + \frac{3J - J_{\perp}}{2} - \frac{J_{\perp}^{2}}{4(3J - J_{\perp})}, \]
and the Dirac point is slightly shifted downward, and the dispersion remains linear. As shown in Fig. 2, the magnon spectrum is almost the same as that of the single-layer FM case, suggesting the magnon excitations from different spin alignments are decoupled.

![Fig. 2. (a) The magnon band structure of AA-stacked FM bilayer with interlayer AFM exchange. (b) The magnon bands along the high symmetry points in the first Brillouin zone. The parameters are the same as those of Fig. 1.](image)

3. AB-stacked honeycomb magnets

Next we consider AB-stacked bilayer quantum magnets, which can be viewed as rotating AA-stacked bilayer by \( \theta = 60^\circ \). In the intralayer and interlayer FM case, the bosonic tight binding Hamiltonian is
\[ H_{\text{FM}}^{\text{AB}} = -J_{\perp} \sum_{i} \frac{1}{2} \left( a_{i,1}^{\dagger}a_{i,2} + a_{i,2}^{\dagger}a_{i,1} \right) + \frac{J}{2} \sum_{i} \left( a_{i,1}^{\dagger}a_{i,1} + a_{i,2}^{\dagger}a_{i,2} \right) \]
\[ -J \sum_{i,j,\ell} \frac{1}{2} \left( a_{i,1}^{\dagger}a_{j,\ell} + a_{i,\ell}^{\dagger}a_{j,1} \right) \]
\[ + \frac{3J}{2} \sum_{i,\ell} a_{i,\ell}^{\dagger}a_{i,\ell}. \quad (9) \]

In the momentum space, it is written as
\[ \mathcal{H}_{\text{FM}}^{\text{AB}}(k) = \mathcal{H}_{0}^{\text{AB}}(k) + \mathcal{D}_{\text{FM}}, \quad (10) \]
with
\[ \mathcal{H}_{0}^{\text{AB}}(k) = \begin{pmatrix} 0 & f(k) & 0 & 0 \\ f^{*}(k) & 0 & -J_{\perp}/2 & 0 \\ 0 & -J_{\perp}/2 & 0 & f(k) \\ 0 & 0 & f^{*}(k) & 0 \end{pmatrix}, \]
\[ \mathcal{D}_{\text{FM}} = \begin{pmatrix} 3J/2 & 0 & 0 & 0 \\ 0 & (3J + J_{\perp})/2 & 0 & 0 \\ 0 & 0 & (3J + J_{\perp})/2 & 0 \\ 0 & 0 & 0 & 3J/2 \end{pmatrix}. \]
The spectrum contains four branches:
\[ E_{1}^{\pm} = \frac{3J}{2} \pm \left| f(k) \right|, \]
\[ E_{2}^{\pm} = \frac{3J + J_{\perp}}{2} \pm \frac{\sqrt{J_{\perp}^{2} + 4\left| f(k) \right|^{2}}}{2}, \]
where \( \left| f(k) \right| \) is linear in \( k \) near the Dirac points \( K, K' \), and thus \( E_{1}^{\pm} \) cross at \( 3J/2 \) linearly [see Fig. 3(a)]. Meanwhile,
\[ E_{2}^{+} \sim \frac{3J}{2} + J_{\perp} + \frac{\left| f(k) \right|^{2}}{J_{\perp}}, \]
\[ E_{2}^{-} \sim \frac{3J}{2} - \frac{\left| f(k) \right|^{2}}{J_{\perp}}, \]
which are quadratic in \( k \) near the Dirac points.

![Fig. 3. The magnon bands along the high symmetric lines in the first Brillouin zone for AB-stacked bilayers with: (a) the interlayer FM coupling; (b) the interlayer AFM coupling. Insets of both figures show the enlarged plots of the magnon bands near the Dirac point.](image)
In the momentum space, we have
\[ \mathcal{H}_{\text{AFM}}(k) = \mathcal{H}_0^{\text{AB}}(k) + D_{\text{AFM}}, \]
with
\[ D_{\text{AFM}} = \begin{pmatrix} 3J/2 & 0 & 0 & 0 \\ 0 & (3J - J_\perp)/2 & 0 & 0 \\ 0 & 0 & (3J - J_\perp)/2 & 0 \\ 0 & 0 & 0 & 3J/2 \end{pmatrix}. \]

By diagonalizing \( s_z \mathcal{H}_{\text{AFM}}^{\text{AB}}(k) \), we obtain the magnon spectrum
\[ E_k^\pm = \sqrt{\left(\frac{3J}{2}\right)^2 + g(k)^2 + |f(k)|^2 - \left(\frac{3J}{2}\right) J_\perp}, \]
with
\[ g(k) = \sqrt{\frac{3J}{2} \left[ \left(\frac{3J}{2}\right) J_\perp^2 - 8J_\perp |f(k)|^2 + 16 \left(\frac{3J}{2}\right) |f(k)|^2 \right]} \).

Near the Dirac points,
\[ E^+ \sim \sqrt{\left(\frac{3J}{2}\right)^2 + \frac{4(3J/2) - J_\perp |f(k)|^2}{2(3J/2)J_\perp}}, \]
\[ E^- \sim \sqrt{\left(\frac{3J}{2}\right)^2 - \left(\frac{3J}{2}\right) J_\perp + \frac{[-4(3J/2) + 3J_\perp |f(k)|^2}{2J_\perp \sqrt{(3J/2)^2 - (3J/2)J_\perp}}}, \]
which are quadratic in \( k \). The spectrum is gapped on the Dirac points, with the gap size \( 3J/2 - \sqrt{(3J/2)^2 - (3J/2)J_\perp} \).

4. Twisted bilayer quantum magnets

By taking any vertical bond in AA-stacked bilayer as the axis, the twisted bilayers are generated by rotating one layer around the axis, while the other layer remains fixed. The periodicity is maintained for commensurate \( \theta \), when a certain site of one layer ends up exactly over a site of the other after the rotation. The Bravais lattice of the superstructure is hexagonal with the two unit vectors: \( L_1 = ma_1 + na_2 \) and \( L_2 = -na_1 + (m+n)a_2 \). Each supercell contains \( N = 4(m^2 + mn + n^2) \) sites.\(^{[58,59]} \) The twist angle writes as
\[ \theta = 2\sin^{-1}\frac{m-n}{2\sqrt{m^2 + n^2 + mn}}. \]

In the following, we consider the commensurate twist angles 21.8°, 6.01°, and 3.89°, which correspond to \( (m,n) = (2,1), (6,5), (9,8) \), respectively. The case of smaller angles have too large supercells to deal with using our computer, and thus instead we calculate the magnon spectrum using the low-energy continuous theory near the Dirac points.\(^{[64,65]} \)

The nearest-neighbor intralayer FM coupling are considered with the strength as the energy scale. The strength of interlayer coupling between sites \( r_i \) and \( r_j \) is given by \( J_{ij} = J_\perp e^{-(|r_i - r_j| - d_0)/\xi} \), where the parameters are set as \( J_\perp/J = -0.17 \), \( d_0 = 0.335 \) nm, and \( \xi = 0.0453 \) nm.

![Fig. 4. Magnon band structures of twisted bilayers with twist angles of 21.8°, 6.01°, and 3.89°: (a)-(c) the interlayer FM coupling; (d)–(f) the interlayer AFM coupling. The intralayer couplings are FM case in all figures. We also plot the magnon spectrums without interlayer coupling (dashed red lines). To compare the Dirac velocity, the Dirac points are moved to the same positions as those in twisted bilayers.](#)
the studied angles. In the interlayer FM cases, while
the slopes are almost unchanged for large twist angles
(θ = 21.8°, 6.01°), there is a clear reduction for small
angle θ = 3.89°. The behavior is very similar to that
of twisted bilayer graphene. It is expected that the
magnon bands becomes flattening as the angle is further
decreased. In contrast, the spectrum of the interlay AFM
case differs little from that of the single-layer quantum
FM case, further verifying the spin excitations of oppo-
site spin orientations do not couple with each other.

Now we begin to study the magnon spectrums of
small twist angles using the low-energy continuous the-
ory near the valley $K_\pm = (4\pi/3\sqrt{3}a) (±1,0)$, where $a$
is the lattice constant. Ignoring the overlap potential
from the z-component of the Heisenberg couplings, the
effective bosonic Hamiltonian consists of three terms[64–71]

$$H^{(±)}_{\text{eff}} = \sum_k \left[ \psi^*_b(k) h^{(±)}_{\theta/2} (k - K^{b}_+) \psi_b(k) + \psi^*_t(k) h^{(±)}_{-\theta/2} (k - K^{t}_+) \psi_t(k) + \sum_{j=1}^3 \left( \psi^*_t(k) T^{(±)}_j \psi_t(k + X^{(±)}_j) + \text{h.c.} \right) \right]$$

(13)

with $K^{t}_+ = R_{\theta/2} K_+$ and $K^{b}_+ = R_{-\theta/2} K_+$, where $R_{\theta}$
represents the momenta near the Dirac point $K_\pm$, which
writes as $k_0 + m G_1 + n G_2 + Y_\pm$ with $k_0$ in the moiré
first Brillouin zone and $Y_\pm = (K^{t}_+ + K^{b}_+)/2 \pm G_1/2$.
Here, $G_1 = K_\theta(1,0)$ and $G_2 = K_\theta(-1/2,\sqrt{3}/2)$ with
$K_\theta = [8\pi \sin(\theta/2)]/3a$ is the length of the basis vectors
in the moiré Brillouin zone. In practice, the integers $m,n$
are cut off to finite values, i.e., $m,n = -l,\ldots,l$, and
the magnon spectrums converge quickly as the cutoff value
$l$ increases. The first two terms describe the isolated FM
sheets of the top and bottom layers under the bases

$$\psi_b(k) = \begin{bmatrix} \phi_{A,b}(k) \\ \phi_{B,b}(k) \end{bmatrix}, \quad \psi_t(k) = \begin{bmatrix} \phi_{A,t}(k) \\ \phi_{B,t}(k) \end{bmatrix}.$$  (14)

The low-energy Hamiltonian for a layer rotated by an
angle θ is $h^{(±)}_b(k) = h_\text{Dirac}(R_{\theta} k) \cdot (±\sigma_x, \sigma_y)$, which is a Dirac
one. The last term in Eq. (13) describes hoppings be-
 tween layers with the hopping matrix

$$T^{(±)}_j = \omega \begin{bmatrix} 1 & e^{±2i\pi/3(j-1)} \\ e^{±2i\pi/3(j-1)} & 1 \end{bmatrix},$$  (15)

and $X^{(±)}_j = (0,0), X^{(±)}_2 = ±(G_1 + G_2)$, and $X^{(±)}_3 = ±G_2$. The magnon spectrum is calculated directly, and
the result for the $\theta = 3.89°$ case is shown in Fig. 5. We
find the interlayer coupling strength $\omega = 0.0439$ eV gives
the best match between the continuous theory and ex-
act tight-binding calculation. It is expected that $\omega$ will
change with the twisted angle $\theta$. However the deter-
mined strength at relatively large $\theta$ provides a good ap-
proximation for the cases of smaller angles, which are
very hard to access in the tight-binding method.

![Fig. 5. Comparison of the low-energy magnon bands between the continuous theory and exact tight-binding calculation with the FM interlayer coupling. Here the twist angle is $\theta = 3.89°$.](image)

5. Conclusions

The magnon bands of both the bilayer and twisted
bilayer honeycomb quantum magnets are studied using
linear spin wave theory. The interlayer ferromagnetic or-
der but two kinds of intralayer exchange couplings are
considered. When the two layers are ferromagnetically
coupled, the linear dispersions near the magnon Dirac
points are preserved, and their slopes are reduced with
the decrease of the twist angles. For the interlayer an-
tiferromagnetic case, we find that the two monolayer
magnon dispersions are usually decoupled due to the op-
posite orientations of the spins in the two layers. Fi-
nally a low-energy continuous theory is developed for
the small twist angles, and fits well with the exact tight-
binding calculations.

Experimentally there have revealed intrinsic magnetism
in various atomically thin crystals. Among them, CrX$_3$
($X = \text{Cl, Br, I}$) is a family of 2D honeycomb quan-
tum magnets. Specifically, intralayer ferromagnetism
and interlayer antiferromagnetism has been observed in
bilayer CrI$_3$. Besides, it has been proposed that the
interlayer exchange coupling can be tuned between AFM
and FM by changing the interlayer stacking order. Thus
based on these bilayer honeycomb quantum magn-
ets, our results may be experimentally observed using
the state of the art measurements, such as inelastic neutron scattering and magneto-Raman spectroscopy. [77]

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