MAGNETIC ORDER AND ITS LOSS ON FRUSTRATED HONEYCOMB MONOLAYERS AND BILAYERS:
An Illustrative Use of the Coupled Cluster Method

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INTRODUCTION

- Example: $J_1$–$J_2$–$J_3$ Model on a Honeycomb Monolayer
- The Coupled Cluster Method

RESULTS

- Results on the Honeycomb Monolayer
  - The spin-1/2 $J_1$–$J_2$–$J_3$ Heisenberg model
- Results on the Honeycomb Bilayer
  - The spin-1/2 $J_1$–$J_2$–$J_3$–$J_1^\perp$ Heisenberg model

SUMMARY

References

D.J.J. Farnell, R.F. Bishop, P.H.Y. Li et al., PRB 84, 012403 (2011)
P.H.Y. Li, R.F. Bishop et al., PRB 86, 144404 (2012)
R.F. Bishop, P.H.Y. Li et al., PRB 92, 224434 (2015)
1. **INTRODUCTION**
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3. **SUMMARY**
The Coupled Cluster Method

**Example:** $J_1 - J_2 - J_3$ Model on a Honeycomb Monolayer

$J_1 - J_2 - J_3$ model on the 2D honeycomb lattice (i.e., all bonds of Heisenberg type)

We’ll look at the case with $s = \frac{1}{2}$ spins (viz., the most quantum case)

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j + J_2 \sum_{\langle\langle i,k \rangle\rangle} \mathbf{s}_i \cdot \mathbf{s}_k + J_3 \sum_{\langle\langle\langle i,l \rangle\rangle\rangle} \mathbf{s}_i \cdot \mathbf{s}_l$$

(and set $J_1 \equiv 1$) where, on the honeycomb lattice:

- $\langle i, j \rangle$ bonds $J_1 \equiv \cdots$ all NN bonds
- $\langle\langle i, k \rangle\rangle$ bonds $J_2 \equiv \cdots$ all NNN bonds
- $\langle\langle\langle i, l \rangle\rangle\rangle$ bonds $J_3 \equiv \cdots$ all NNNN bonds

**NOTE:** The honeycomb lattice is bipartite but non-Bravais (– two sites per unit cell: A, B)
Limiting Cases

limiting bond cases
- $J_2 = J_3 = 0$: isotropic HAF on 2D honeycomb lattice
- $J_1 = J_3 = 0$: two uncoupled isotropic HAFs on 2D triangular lattice
- $J_1 = J_2 = 0$: four uncoupled isotropic HAFS on 2D honeycomb lattice

classical limit ($s \to \infty$)
- For $J_1 > 0$: ground-state (GS) phase diagram is complex, containing 6 different ordered phases -
  - Néel
  - Striped
  - Néel-II
  - Spiral-I
  - Spiral-II
  - Ferromagnetic
- For $J_1 < 0$: also 6 phases, related to those above by simple symmetries (i.e., $J_1 \iff -J_1$; $J_3 \iff -J_3$; $s_i^B \iff -s_i^B$)
Both the Striped and Néel-II regions actually have an infinitely degenerate family of non-coplanar ground states, from which the collinear states shown are selected by thermal or quantum fluctuations.

The most highly frustrated point at $J_2 / J_1 = \frac{1}{2}, J_3 / J_1 = \frac{1}{2}$ (i.e., a classical triple point) lies along the line $J_3 = J_2$. 

Classical $(s \to \infty)$ Phase Diagram $(J_1 > 0)$
Néel, Striped, Spiral-I, and Néel-II Model States

Example: $J_1 - J_2 - J_3$ Model on a Honeycomb Monolayer

The Coupled Cluster Method

Honeycomb Monolayers & Bilayers via the CCM

NMP17
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- Results on the Honeycomb Monolayer
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3 SUMMARY
Elements of the CCM

We use the **coupled cluster method** (CCM)

- **ground-state (GS) wavefunction:**
  \[ |\Psi\rangle = e^S |\Phi\rangle; \quad \langle \tilde{\Psi}| = \langle \Phi| \tilde{S} e^{-S}; \quad \langle \tilde{\Psi}|\Psi\rangle = \langle \Phi|\Psi\rangle = \langle \Phi|\Phi\rangle \equiv 1 \]

  \[ S = \sum_{I \neq 0} S_I C_I^+; \quad \tilde{S} = 1 + \sum_{I \neq 0} \tilde{S}_I C_I^- \]

- \( C_0^+ \equiv 1; \quad C_I^- \equiv (C_I^+)^\dagger; \quad C_I^- |\Phi\rangle = 0, \quad \forall I \neq 0 \)

- \( C_I^+ |\Phi\rangle \) are a complete set of wf’s; \([C_I^+, C_J^+] = 0\)

- choose model state \( |\Phi\rangle \) to be, e.g., a classical GS (i.e., Néel, Striped, Spiral-I, and Néel-II)

- choose spin axes on each site so that \( |\Phi\rangle = |\downarrow\downarrow \cdots \downarrow\rangle \) in these local axes

  \[ \Rightarrow C_I^+ \rightarrow s_{i_1}^+ s_{i_2}^+ \cdots s_{i_k}^+; \quad s_j^+ \equiv s_j^x + is_j^y \], in local axes
Elements of the CCM

- Each $s_i^+$ in $C_i^+$ can appear at most once for $s = \frac{1}{2}$, twice for $s = 1, \cdots$, and $2s$ times for general spin-$s$ case, on a given lattice site $i$.

- Solve for $\{S_I, \tilde{S}_I\}$ from GS Schrödinger eqs. for $|\Psi\rangle, \langle \tilde{\Psi}| \implies$ equivalently, minimize $\tilde{H} = \tilde{H}(S_I, \tilde{S}_I) \equiv \langle \Phi| \tilde{S}e^{-S}He^S|\Phi\rangle$ with respect to all parameters $\{S_I, \tilde{S}_I; \forall I \neq 0\}$

$$\rightarrow \frac{\delta \tilde{H}}{\delta \tilde{S}_I} = 0 \implies \langle \Phi| C_I^- e^{-S}He^S |\Phi\rangle = 0, \ \forall I \neq 0$$

- A coupled set of nonlinear equations for $\{S_I\}$

$$\implies E = \langle \Phi| e^{-S}He^S |\Phi\rangle = \langle \Phi| He^S |\Phi\rangle$$ (1)

$$\rightarrow \frac{\delta \tilde{H}}{\delta S_I} = 0 \implies \langle \Phi| \tilde{S}e^{-S}[H, C_I^+]e^S|\Phi\rangle = 0, \ \forall I \neq 0$$

$$\implies \langle \Phi| \tilde{S}(e^{-S}He^S - E)C_I^+ |\Phi\rangle = 0, \ \forall I \neq 0$$

- A coupled set of linear generalized eigenvalue equations for $\{\tilde{S}_I\}$ with $\{S_I\}$ as input.
Elements of the CCM

- Note that the nonlinear exponentiated terms only ever appear in the form of the similarity transform of the Hamiltonian: $e^{-S}He^S$

  $\Rightarrow$ use the nested commutator expansion

  $e^{-S}He^S = H + [H, S] + \frac{1}{2!}[[H, S], S] + \cdots$

  NOTE: This series will terminate exactly after the term bilinear in $S$ for our Heisenberg Hamiltonians $\Rightarrow$

- CCM satisfies the Goldstone linked cluster theorem and satisfies the Hellmann-Feynman theorem, for all truncations on complete set $\{I\}$

- We use the natural lattice geometry to define the approximation schemes and we retain all distinct fundamental configurations (fc) in the set $\{I\}$ with respect to space- and point-group symmetries of both the Hamiltonian and the model state $|\Phi\rangle$

- A similar CCM parametrization exists for excited states too
only approximation is to truncate set \( \{ I \} \)

- for \( s = \frac{1}{2} \) case we typically use the LSUB\( m \) scheme in which we retain all possible multispin-flip correlations over different locales on the lattice defined by \( m \) or fewer contiguous lattice sites
- for \( s \geq 1 \) cases we often use the alternative SUB\( n-m \) scheme in which we retain all multispin-flip correlations involving up to \( n \) spin flips spanning a range of no more than \( m \) adjacent (or contiguous) lattice sites. We then set \( m = n \) and employ the so-called SUB\( m-m \) scheme.

**NOTE:** LSUB\( m \equiv \) SUB\( 2s m-m \) for general spin-\( s \) case, (i.e., LSUB\( m \equiv \) SUB\( m-m \) only for \( s = \frac{1}{2} \) case)
Number of CCM Fundamental Configurations, $N_f$

For the spin-1/2 $J_1 - J_2 - J_3$ model on the honeycomb lattice:

<table>
<thead>
<tr>
<th>Method</th>
<th>$N_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Néel</td>
</tr>
<tr>
<td>LSUB4</td>
<td>5</td>
</tr>
<tr>
<td>LSUB6</td>
<td>40</td>
</tr>
<tr>
<td>LSUB8</td>
<td>427</td>
</tr>
<tr>
<td>LSUB10</td>
<td>6237</td>
</tr>
</tbody>
</table>

**NOTE:** To obtain a single data point (i.e., for given values of $J_2$ and $J_3$, with $J_1 = 1$) for the spiral-I phase at the LSUB10 level we typically require about 6 h computing time using 2000 processors simultaneously.
at each LSUB \( m \) or SUB \( m-m \) level the CCM operates at the \( N \to \infty \) limit from the outset

calculate \( E/N \) and magnetic order parameter (i.e., local average onsite magnetization) \( M \equiv -\frac{1}{N} \sum_{N} \langle \tilde{\Psi} | s_{i}^{z} | \Psi \rangle \) in the local rotated axes

extrapolate to the exact \( m \to \infty \) limit, using well-tested empirical scaling laws

\[
\begin{align*}
E/N &= a_0 + a_1 m^{-2} + a_2 m^{-4} \\
M &= b_0 + b_1 m^{-1} + b_2 m^{-2} \\
M &= b_0 + b_1 m^{-0.5} + b_2 m^{-1.5}
\end{align*}
\]

for unfrustrated models

for highly frustrated models
INTRODUCTION

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Results on the Honeycomb Bilayer
- The spin-1/2 $J_1-J_2-J_3-J_4^{\perp}$ Heisenberg model

SUMMARY
We have done a large study of this model

Results include:

- **The case when** $J_3 = J_2$ **for which we have investigated the full phase diagram for both signs of the bonds**

  References
  
  D.J.J. Farnell *et al.*, PRB *84*, 012403 (2011)

- **The case when** $J_3 = 0$ *(i.e., the* $J_1$–$J_2$ *model)*; $J_1 > 0, J_2 > 0$

  References
  

- **The full** $J_1$–$J_2$–$J_3$ **model**; $J_1 > 0, J_2 > 0, J_3 > 0$

  Reference
  
  P.H.Y. Li *et al.*, PRB *86*, 144404 (2012)
The classical \((s \rightarrow \infty)\) \(J_1-J_2-J_3\) model on the monolayer honeycomb lattice is most frustrated at the classical tricritical point \((J_2/J_1 = 1/2, J_3/J_1 = 1/2)\) at which three phases (Néel, striped and spiral-I) meet. Let us restrict ourselves initially, for illustrative reasons, to study the model along the line \(J_3 = J_2 = \alpha J_1\) for \(J_1 > 0\), at the point \(\alpha = 1/2\) there is a classical phase transition from a non-degenerate Néel phase to an infinitely degenerate family of GS phases (from which the striped phase is selected by quantum or thermal fluctuation). This region should be a fertile hunting-ground for novel phases for the \(s = \frac{1}{2}\) quantum case.
RESULTS I: Monolayer with $J_1 \equiv +1; J_3 = J_2$

- We study the case $J_1 \equiv +1; 0 \leq J_3 = J_2 \equiv \alpha J_1 \leq 1$
- Notice how we obtain (real) solutions, for a given model state, only for certain ranges of $\alpha \equiv J_2/J_1$, with termination points shown.
- The energy and magnetic order parameter results clearly show the existence of a GS phase intermediate between the Néel and striped phases.
- We can test for other orderings by measuring the response to a field operator $F \equiv \delta \hat{O}_F$ added to $H$, and calculating $e(\delta) \equiv E(\delta)/N$ for the perturbed Hamiltonian $H + F$. We then measure the response by the susceptibility:

$$\chi_F \equiv - \left[ \frac{\partial^2 e(\delta)}{\partial \delta^2} \right]_{\delta=0}$$
$s = \frac{1}{2} J_1 - J_2 - J_3$ Model with $J_3 = J_2$: GS Energy ($J_1 \equiv 1$) for the Néel and Striped States

DJJF, RFB, PHYL, JR, CEC / PRB 84, 012403 (2011)
$s = \frac{1}{2} J_1 - J_2 - J_3$ Model with $J_3 = J_2 (J_1 \equiv 1)$: Order Parameter for the Néel and Striped States

Let us now test for PVBC order in the intermediate regime.
\( s = \frac{1}{2} J_1 - J_2 - J_3 \) Model with \( J_3 = J_2 \): \( \frac{1}{\chi_p} \) versus \( J_2 \) (\( J_1 \equiv 1 \)) for the Néel and Striped States

Right: The perturbations (fields) \( F = \delta \hat{O}_p \) for the plaquette susceptibility \( \chi_p \). Thick (red) and thin (black) lines correspond respectively to strengthened and weakened NN exchange couplings, where \( \hat{O}_p = \sum_{\langle i,j \rangle} a_{ij} \mathbf{s}_i \cdot \mathbf{s}_j \), and the sum runs over all NN bonds, with \( a_{ij} = +1 \) and \( -1 \) for thick (red) and thin (black) lines respectively.

\( \text{LSUB} \to \infty \) uses: \( \chi_p^{-1}(m) = x_0 + x_1 m^{-2} + x_2 m^{-4} \) (to extrapolate \( \text{LSUB} m \))
Intermediate Discussion

The energy and order parameter results clearly show:

- Néel ordering persists for $\frac{J_2}{J_1} \equiv \alpha < \alpha_{c_1} \approx 0.47$
- Striped ordering exists only for $\alpha > \alpha_{c_2} \approx 0.60$
- PVBC ordering appears to exist for $\alpha_{c_1} < \alpha < \alpha_{c_2}$

compared to the direct classical phase transition between the Néel and striped AFM phases at $\alpha = 0.5$

These results are confirmed from calculations of

- $\Delta$, triplet spin gap
- $\rho_s$, spin stiffness coefficient
- $\chi$, zero-field, uniform transverse magnetic susceptibility

Reference

R.F. Bishop, P.H.Y. Li et al., PRB 92, 224434 (2015)

– and see Appendix for details
We can also investigate the case $J_1 \equiv -1$ to examine the other boundary of the striped AFM phase.

Finally, we can also investigate the case $J_1 \equiv 1$ but with $J_2 < 0$ to examine the other boundary of the Néel AFM phase.

The classical FM state is also an eigenstate of the quantum Hamiltonian. Its GS energy is given by

$$E_{\text{FM}}^{\text{cl}} = \frac{s^2}{N} \left( \frac{3}{2} J_1 + \frac{9}{2} J_2 \right)$$
\[ s = \frac{1}{2} \left( J_1 - J_2 - J_3 \right) \quad \text{Model with } J_3 = J_2: \text{GS energy} \]

\[ (J_1 = -1) \text{ vs } J_2 \text{ for the Striped and FM States} \]

**PHYL, RFB, DJJF, JR, CEC / PRB 85, 085115 (2012)**

**NOTE**: Curves with symbols refer to the case \( J_1 = +1 \), for comparison

There is clear evidence for either

- a direct first-order transition between the striped and FM phases at \( \alpha \approx -0.10 \), or
- an intervening phase in the very narrow range \( -0.12 \lesssim \alpha \lesssim -0.10 \)
  
  (c.f., the classical case of an intervening spiral phase in the larger range \( -\frac{1}{5} < \alpha < -\frac{1}{10} \))
$s = \frac{1}{2} J_1 - J_2 - J_3$ Model with $J_3 = J_2$: GS Energy $(J_1 \equiv 1; J_2 < 0)$ for the Néel and Striped States

There is clear evidence for a direct first-order phase transition between the Néel and FM phases at $\alpha = -1.17 \pm 0.01$ (c.f., the classical value $\alpha = -1$)
\[ s = \frac{1}{2} J_1 - J_2 - J_3 \] Model \((J_3 = J_2)\): Full Phase Diagram

RFB, PHYL / PRB 85, 155135 (2012)

(a) Classical \((s \to \infty)\)

- The transition from Néel to PVBC order is a continuous (and hence deconfined) one
- The transition from PVBC to striped order is a first-order one
- The transitions from striped and Néel AFM order to FM order are both first-order ones
$s = \frac{1}{2} J_1 - J_2 - J_3$ Model: Phase Diagram

$(J_1 \equiv 1; 0 \leq J_2 \leq 1, 0 \leq J_3 \leq 1)$

**NOTE:** c.f., the classical ($s \to \infty$) model has Néel, striped and spiral phases only, with phase boundaries shown by the light grey lines (dashed for continuous transitions and solid for first-order transition)
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3 SUMMARY
Model on the Honeycomb Bilayer Lattice

- $J_1 - J_2 - J_3 - J_{1\perp}$ model on the honeycomb bilayer lattice (i.e., all bonds of Heisenberg type) – now 4 sites per unit cell: $1_A, 2_A, 1_B, 2_B$ as shown

- We’ll look at the case with $s = \frac{1}{2}$ spins (viz., the most quantum case)

- $H = J_1 \sum_{\langle i, j \rangle, \alpha} \mathbf{s}_i,\alpha \cdot \mathbf{s}_j,\alpha + J_2 \sum_{\langle \langle i, k \rangle \rangle, \alpha} \mathbf{s}_i,\alpha \cdot \mathbf{s}_k,\alpha + J_3 \sum_{\langle \langle \langle i, l \rangle \rangle \rangle, \alpha} \mathbf{s}_i,\alpha \cdot \mathbf{s}_l,\alpha + J_{1\perp} \sum_i \mathbf{s}_i, A \cdot \mathbf{s}_i, B$

  (where $\alpha = A, B$ labels the two layers, and set $J_1 \equiv 1$)

- $- - - - = J_{1\perp}$: NN interlayer bond on both layers $\alpha = A, B$
We have investigated several special cases for this model.

Results include:

- **The case when** $J_3 = J_2 \equiv \alpha J_1 > 0$; $J_1 > 0$, $J_1^\perp \equiv \delta J_1 > 0$,
  for which we have investigated the stability of the Néel and striped phases in the $\alpha$–$\delta$ plane.

  Reference

- **The case when** $J_3 = 0$ (i.e., the $J_1$–$J_2$–$J_1^\perp$ model);
  $J_1 > 0$, $J_2 \equiv \kappa J_1 > 0$, $J_1^\perp \equiv \delta J_1 > 0$,
  for which we have investigated the stability of the Néel phase in the $\kappa$–$\delta$ plane.

  Reference
Limiting Cases

- limiting bond cases
  - $J_{1-} = 0$: two uncoupled honeycomb monolayers
  - $J_{1-} \rightarrow \infty$: with finite $J_1$, $J_2$, $J_3$; NN interlayer pairs form spin-singlet dimers $\Rightarrow$
    GS is a nonclassical interlayer dimer valence-bond crystal (IDVBC),

\[
\frac{E}{N} \xrightarrow{J_{1-} \rightarrow \infty} \frac{E_{IDVBC}}{N} = -\frac{1}{2} s(s + 1) J_{1-}
\]

($s = \text{spin quantum number}$)
We study the case $J_1 \equiv +1; 0 \leq J_3 = J_2 \equiv \alpha J_1 \leq 1; J_1^\perp \equiv \delta J_1 \geq 0$

As before for the monolayer we obtain real solutions, for a given model state (i.e., Néel or striped), only for certain regions in the $\alpha$–$\delta$ phase space

We have calculated $E/N, M$ as before

We have also calculated

- the triplet spin gap $\Delta$ (i.e., the excitation energy from the GS to the lowest-lying $s = 1$ excited state)
- the zero-field uniform transverse magnetic susceptibility, $\chi$ [i.e., put system in a transverse magnetic field $h$, in units where $g\mu_B/\hbar = 1$, and calculate $\chi(h) = -\frac{1}{N}d^2E/dh^2; \chi \equiv \chi(0)$]
$s = \frac{1}{2} J_1 - J_2 - J_3 - J_1^\perp$ Honeycomb Bilayer Model with $J_3 = J_2 (J_1 \equiv 1)$: Order Parameter for the Néel State

RFB, PHYL / unpublished (2017)

\[ \delta \equiv \frac{J_1^\perp}{J_1}; \quad \alpha \equiv \frac{J_3}{J_1} (= \frac{J_2}{J_1}) \]

**NOTE:** `LSUB\infty(i)` extrapolations are based on `LSUBm` data sets with

- $m = \{2, 6, 10\}$ for $i = 1$
- $m = \{4, 6, 8, 10\}$ for $i = 2$
Honeycomb Bilayer Model with $J_3 = J_2$ ($J_1 \equiv 1$): Order Parameter for the Striped State

\[ s = \frac{1}{2} J_1 - J_2 - J_3 - J_1^\perp \]

\[ \delta \equiv \frac{J_1^\perp}{J_1}; \quad \alpha \equiv \frac{J_3}{J_1} (= \frac{J_2}{J_1}) \]

NOTE: LSUB$\infty$(i) extrapolations are based on LSUB$m$ data sets with

- $m = \{2, 6, 10\}$ for $i = 1$
- $m = \{4, 6, 8, 10\}$ for $i = 2$
\[ s = \frac{1}{2} J_1 - J_2 - J_3 - J_1^\perp \]  
Honeycomb Bilayer Model with \( J_3 = J_2 \) \((J_1 \equiv 1)\): Extrapolated Order Parameter for the Néel and Striped States

\[
\delta \equiv \frac{J_1^\perp}{J_1}; \quad \alpha \equiv \frac{J_3}{J_1} (= \frac{J_2}{J_1})
\]

NOTE: \( \text{LSUB}_\infty \) extrapolations are based on \( \text{LSUB}m \) data sets with \( m = \{2, 6, 10\} \)
\( s = \frac{1}{2} J_1 - J_2 - J_3 - J_{1\perp} \) Model: Phase Diagram

\(( J_3 = J_2 \equiv \alpha J_1 > 0; J_{1\perp} \equiv \delta J_1 > 0; J_1 \equiv 1)\)

RFB, PHYL / unpublished (2017)

NOTE:
- \( \text{LSUB}\infty \) extrapolations are based on \( \text{LSUB}m \) data sets with \( m = \{2, 6, 10\} \)
- The red cross (\( \times \)) symbols and the green plus (\( + \)) symbols are points at which the extrapolated GS magnetic order parameter \( M \) for the Néel and striped phases vanishes, for specified values of \( \delta \) and \( \alpha \), respectively
Discussion

- Both the Néel and striped AFM phases exhibit reentrant regimes.
- The phase boundaries of the two quasiclassical AFM phases exhibit a prototypical avoided crossing behaviour.
- The paramagnetic regime is likely to contain a mixture (at least) of phases with IDVBC order and PVBC order in both layers separately.
In conclusion, we know of no more powerful nor more accurate method than the CCM for dealing with these strongly correlated and highly frustrated 2D spin-lattice models of quantum magnets, such as the honeycomb examples used here for an illustration.

By now, we have used the CCM for many other spin-lattice models. Some other typical examples are:

- the $J_1$–$J_2$ model on the Union Jack lattice
- the $J_1$–$J_2$ model on the checkerboard lattice
- other similar depleted $J_1$–$J_2$ models on the square lattice
- other models that interpolate between various lattices, e.g.,
  - (a) kagome-triangle; (b) kagome-square;
  - (c) square-triangle; (d) hexagon-square

There are now $\gtrsim 125$ papers using the CCM for spin lattices.
Acknowledgements

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THANK YOU FOR YOUR ATTENTION!
Some references for the CCM methodology and applications

\[ s = \frac{1}{2} J_1 - J_2 - J_3 \]

Model with \( J_3 = J_2 \equiv \alpha J_1 \) \((J_1 > 0)\):

**Triplet Spin Gap**


\[ \Delta (m) = d_0 + d_1 m^{-1} + d_2 m^{-2} \quad \text{(to extrapolate LSUBm)} \]
$s = \frac{1}{2} J_1 - J_2 - J_3$ Model with $J_3 = J_2 \equiv \alpha J_1$ ($J_1 > 0$):

Spin Stiffness Coefficient

RFB, PHYL, OG, JR, CEC / PRB 92, 224434 (2015)

- Impose a twist $\theta$ per unit length ($d \equiv$ honeycomb lattice spacing) to a quasiclassical state

\[
\frac{E(\theta)}{N} = \frac{E(\theta=0)}{N} + \frac{1}{2} \rho_s \theta^2 + O(\theta^4)
\]

$\rho_s = \text{spin stiffness coefficient}$

- LSUB$\infty$ uses: $\rho_s(m) = s_0 + s_1 m^{-1} + s_2 m^{-2}$ (to extrapolate LSUB$m$)
\[ s = \frac{1}{2} J_1 - J_2 - J_3 \] Model with \( J_3 = J_2 \equiv \alpha J_1 \) (\( J_1 > 0 \)):

Zero-Field Transverse Magnetic Susceptibility

RFB, PHYL, OG, JR, CEC / PRB 92, 224434 (2015)

- Put \( z_s \)-aligned system in a transverse magnetic field \( \mathbf{h} = h \hat{x} \) (in units where \( g\mu_B/\hbar = 1 \)): \( H \rightarrow H(h) = H(0) - h \sum_i s_i^x \)

\[
\frac{E(h)}{N} = \frac{E(h=0)}{N} - \frac{1}{2} \chi h^2 + O(h^4)
\]

\( \chi \) = zero-field, uniform, transverse magnetic susceptibility

- \( \text{LSUB}\infty(1) \) uses: \( \chi(m) = x_0 + s_1 m^{-1} + x_2 m^{-2} \) (to extrapolate \( \text{LSUB}m \))
- \( \text{LSUB}\infty(2) \) uses: \( \chi(m) = \bar{x}_0 + \bar{x}_1 m^{-\nu} \) (to extrapolate \( \text{LSUB}m \))
\( s = \frac{1}{2} J_1 - J_2 - J_3 \) Model with \( J_3 = J_2 \equiv \alpha J_1 \ (J_1 > 0) \):

**Discussion**

- The extrapolated curves for \( \Delta \) show clear evidence of a **gapped state** between the Néel and striped phases (i.e., consistent with our previous identification of a PVBC intermediate state)

- Points where \( \rho_s \to 0 \) are clear signals of a magnetic phase losing its stability

- Points where \( \chi \to 0 \) are clear signals of the opening up of a **gapped state** (c.f., the classical transition from Néel to striped)

- Each of the curves for \( \Delta, \rho_s \) and \( \chi \) yields corresponding QCPs to those found from the previous curves for \( M \)