Spin-wave excitation spectra and spectral weights
in square lattice antiferromagnets

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Using a recently developed method for calculating series expansions of the excitation spectra of quantum lattice models, we obtain the spin-wave spectra for square lattice, $S = 1/2$ Heisenberg-Ising antiferromagnets. The calculated spin-wave spectrum for the Heisenberg model is close to but noticeably different from a uniformly renormalized classical (large-$S$) spectrum with the renormalization for the spin-wave velocity of approximately 1.18. The relative weights of the single-magnon and multi-magnon contributions to neutron scattering spectra are obtained for wavevectors throughout the Brillouin zone.

Thermodynamic properties and excitation spectra of two-dimensional quantum antiferromagnets have attracted much attention, especially because of their potential relevance to high temperature superconductivity in the cuprate perovskites. Methods based on high-order series expansions about the Ising model have proven to be very successful in accurate calculations of the thermodynamic properties of two-dimensional antiferromagnets. One major limitation of these methods has been their inability to deal directly with dynamical properties or excitation spectra. These quantities have so far been studied within the series approach, via frequency moments and single mode approximations. The reliability of using only a few moments to reproduce spectral lineshapes has come into question, for example, in the case of the two-magnon Raman spectra. Currently efforts are underway to use a large number of frequency moments to obtain the dynamical properties by numerical analytical continuation.

Recently one of us has shown that a series expansion method can be used to directly calculate excited state properties of quantum many-body systems. Here, we apply this method to calculate the energies and spectral weights of the elementary excitations in square lattice, $S = 1/2$ Heisenberg-Ising magnets by expansion around the Ising limit to order $(J_\perp/J_x)^{10}$. The spin-wave dispersion is to construct an effective Hamiltonian for the states which are the natural, perturbatively conserved multi-magnon excitations even though the relative weight of such excitations vanishes near $(0,0)$ and $(\pi, \pi)$.

The Heisenberg-Ising Hamiltonian under consideration is defined by

$$\mathcal{H} = J_z \sum_{\langle i,j \rangle} S_i^z S_j^z + \alpha(S_i^x S_j^z + S_i^y S_j^y),$$

(1)

where the sum runs over nearest-neighbor pairs on a square lattice for which the lattice constant is the unit length ($a \equiv 1$), and $\alpha = J_\perp/J_x$. In the Ising limit, $\alpha = 0$, there are two degenerate ground states and the single-magnon excitations are single spin-flips with respect to the Néel states. In this limit the excitations are purely local, or, alternatively, one could say that the magnon energies are degenerate over the entire band. For $\alpha \neq 0$, the single-magnon states evolve into a set of states with a nonzero dispersion. The key to calculating the spin-wave dispersion is to construct an effective Hamiltonian for the states which are the natural, perturbatively constructed extensions of the single spin-flip states at finite $\alpha$. The effective Hamiltonian is then readily diagonalized by Fourier transformation.

Because the spins on the two sublattices are oriented in opposite directions, the single spin-flip states naturally divide into two sets, those corresponding to $S^z = +1$ and those with $S^z = -1$. The effective Hamiltonian, which conserves $S^z$, thus connects only the basis states with spin-flips on the same sublattice: this ensures that the spin-wave spectrum is degenerate between wavevectors...
(q_x, q_y) and (π − q_x, π − q_y). The full effective Hamiltonian
to order α^10 is presented in Table I.

For α ≠ 1 there is a gap in the spectrum, the minimum
being at (0, 0) and (π, π). The expansion for the gap is
\[ 2 - (5/3)α^2 + 0.31712963α^4 - 0.41923376α^6 
+ 0.27099699α^8 - 0.38943351α^{10} + \cdots \]
which agrees completely with the “mass gap” calculated
by Zheng et al. General arguments tell us that the gap
must close in the Heisenberg limit, α → 1. Moreover, we
expect that for small q = |q| the spectrum has the form
\( c(q) \approx [A(α) + B(α) q^2]^{1/2} \), where A(α) 0 as α → 1.
The spin-wave velocity in the Heisenberg limit is given by
B(1)^{1/2}. In order to calculate the spin-wave velocity, we
expand c(q) in powers of q, c(q) = C(α) + D(α) q^2 + \cdots,
and identify C = A^{1/2} and D = B/2A^{1/2}. Thus the
square of the spin-wave velocity for the Heisenberg model
is given by the α → 1 limit of the series
\[ 2C(α)D(α) = 4α^2 - 2.3055560α^4 + 2.410512α^6 
- 3.064695α^8 + 4.100549α^{10} + \cdots. \]

Since we expect only a weak, energy-like singularity
in this series at α = 1, we can sum it by Padé approxi-
mants. The near-diagonal approximants [2/2], [2/3] and
[3/2] give estimates of 2.779, 2.785 and 2.785 respectively,
which leads to values for the spin-wave velocity c/Ja of
1.667, 1.669 and 1.669. In the large-S limit c/Ja = √2,
so the quantum renormalization of the spin-wave veloc-
ity is Z_κ ≈ 1.18. This number is in excellent agreement
with the high-order spin-wave calculation and previous
indirect estimates using the hydrodynamic relation
c^2 = ρ_σ/χ_⊥, where ρ_σ is the spin-stiffness and χ_⊥
the uniform transverse susceptibility.

Away from the gapless points (0, 0) and (π, π) the
spin-wave spectrum for the Heisenberg model can be es-
timated by direct Padé approximants. Along the line
q_x = q_y, the spectrum, within our numerical uncertain-
ties, is uniformly renormalized with respect to the clas-
sical (large-S) spectrum. However, along the line q_x = 0
appreciable differences appear. In particular, the excita-
tion energy at (π, 0) appears to be a shallow local mini-
num along this line, and is lower than that at (π/2, π/2)
by about 7%. Note that at the classical and 1/S lev-
els the spin-wave spectrum is degenerate at (π, 0) and
(π/2, π/2). A plot of the dispersion relation along se-
lected directions is shown in Fig. I. A hint of the devi-
tions from classical spin-wave theory which we have
found near (π, 0) can be seen in the results of the projec-
tor quantum Monte Carlo calculations of Chen et al., even
though those authors emphasize the similarity of the
S = 1/2 spin-wave spectrum and the uniformly renormal-
ized classical spectrum. The first calculation of the spec-
trum by spin-wave expansion to order 1/S^2 by Igarashi
and Watabe also suggested that (π, 0) would be a local
minimum for the spin-wave spectrum; however, that mini-
num is no longer present in the more recent spin-wave
calculations by Igarashi.

We now turn to the spin-wave spectral weights. The
magnetic neutron scattering cross sections is proportional
to the dynamic structure factor, given by the expression
\[ \tilde{S}(q, ω) = \int dt e^{-iωt} \sum_r e^{iqr} \langle S_x(0, 0)S_x(r, t) \rangle + \langle S_y(0, 0)S_y(r, t) \rangle. \]

We consider the T = 0 limit, where the angular brackets
refer to ground state expectation values. In general, we
expect \( \tilde{S}(q, ω) \) to consist of a sum of two parts,
\[ \tilde{S}(q, ω) = A(q)δ(ω − c(q)) + B(q, ω). \]

Here c(q) is the spin-wave dispersion and A(q) is the
residue or spectral weight associated with the spin-waves.
B(q, ω) is associated with multi-magnon excitations,
which are present when a single spin-flip in an anti-
eromagnet cannot be exactly represented as a superpo-
sition of single-magnon states. Integrating \( \tilde{S}(q, ω) \) over
all frequencies yields the equal-time correlation function
\[ \tilde{S}(q) = \sum_r e^{iqr} \langle S_x(0, 0)S_x(r, 0) \rangle + \langle S_y(0, 0)S_y(r, 0) \rangle. \]

To determine the residue A(q), we need to restrict the
intermediate states that arise in the calculation of the ex-
pectation values to be single magnon states, which gives
the expression
\[ A(q) = \sum_r e^{iqr} \langle S_x(0, 0)P S_x(r, 0) \rangle + \langle S_y(0, 0)P S_y(r, 0) \rangle, \]

where P is the projection onto the manifold of single-
magnon states. In the cluster expansions, each single-
magnon state evolves with the coupling α, and is of the
form
\[ |ψ_i⟩ = |i⟩ + \sum_n C_{i,n}(α)|n⟩, \]

where |i⟩ is a single-magnon state in the Ising limit and
|n⟩ represents basis states (eigenstates in the Ising limit)
which are not degenerate with the single-magnon states.
However, the states |ψ_i⟩ for different i are not orthogonal
to each other when α ≠ 0. Thus in order to construct the
projection operator, we need to define the overlap matrix
\( g_{i,j} = ⟨ψ_i|ψ_j⟩ \). Then the projection operator onto
the single-magnon subspace is given by the expression
\[ P = \sum_{i,j} g_{i,j}^{-1} |ψ_i⟩ ⟨ψ_j|. \]

The expansion coefficients for the residues in real space
as a function of the vector distance are given in Table I.
The coefficients for the transverse structure factor are
given in Ref. 5.
We can now estimate the multi-magnon contribution to neutron scattering by simply subtracting $A(q)$ from the equal-time correlation $\hat{S}(q)$. To get to the Heisenberg limit a series extrapolation is needed. Since the series for the multi-magnon weights are reduced by two terms (the first two being zero) compared to the parent series, one might suspect it would be better to extrapolate the series for the total cross section and the single-magnon contribution and take differences; we have used both methods to estimate the multi-magnon weights, carrying out the extrapolations by direct Padé approximants. In Fig. 2 results are presented for both the multi-magnon spectral weight as well as the ratio of the multi-magnon weight to the total spectral weight, along several lines in the Brillouin zone. We see that the multi-magnon contribution is particularly large near $(\pi, 0, 0)$ (where, unfortunately, the extrapolation uncertainties are largest as well) and amounts to roughly a quarter of the total spectral weight. The spin-wave calculations of Igarashi and Watabe take differences; we have used both methods to look for the multi-magnon excitations.

Recently, Stringari has developed general bounds and sum rules for single-magnon and multi-magnon spectra at special wavevectors. Near $q = (0, 0, 0)$ the single magnon spectral weight vanishes linearly while the multi-magnon weight vanishes quadratically; and near $q = (\pi, \pi, \pi)$, the single-magnon spectral weight diverges as $|q - q|^{-1}$ while the multi-magnon weight goes to a constant. Our results are consistent with all of these requirements.

In summary, series expansions and extrapolations have been carried out for dynamic properties of the $S = 1/2$ square lattice Heisenberg-Ising model. Our numerical results indicate that the Heisenberg model spin-wave spectrum is close to but noticeably different from a uniformly renormalized classical spectrum. In addition, the single-magnon and multi-magnon spectral weights have been estimated throughout the Brillouin zone. In light of our numerical results, it would be interesting to examine the inelastic neutron scattering data on the antiferromagnetic parent compounds of the cuprate superconductors to look for the multi-magnon excitations.

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FIG. 1. Spin-wave spectrum for the Heisenberg model, in units of $J$, along three lines in reciprocal space. The solid circles with error bars are the results from the series expansions; the solid line is the classical spin-wave spectrum multiplied by an overall factor $Z_c = 1.18$.

FIG. 2. Heisenberg model multi-magnon spectral weight (crosses, narrowest error bars) and the ratio of the multi-magnon spectral weight to the total spectral weight as determined by extrapolations for the multi-magnon weight series (solid squares, widest error bars) and by the difference of extrapolations for the total weight and the single-magnon weight (diamonds, intermediate width error bars).

TABLE I. Effective Hamiltonian for the Heisenberg-Ising model elementary excitations in real space, up to an overall factor of four. The dispersion in reciprocal space is found by summing all of the given real-space series with a factor $(1/4)[\cos(q_x r_x + q_y r_y) + \cos(q_x r_x - q_y r_y) + \cos(q_x r_y + q_y r_x) + \cos(q_x r_y - q_y r_x)]$, and then dividing by four.

| $r$     | Series                                      |
|---------|---------------------------------------------|
| (0,0)   | $8 - 0.6666662e^2 + 0.664352e^6 - 0.291737/e^8 + 0.201076/e^8 - 0.177466/e^{10}$ |
| (1,1)   | $-4e^2 + 1.222222e^4 - 0.541756e^6 + 0.513359/e^8 - 0.430699/e^{10}$ |
| (2,0)   | $-2e^2 + 0.111111e^4 - 0.290209/e^6 + 0.255912/e^8 - 0.331992/e^{10}$ |
| (2,2)   | $-0.291667/e^4 + 0.071979/e^6 + 0.060564/e^8 - 0.133137/e^{10}$ |
| (3,1)   | $-0.388889/e^4 - 0.177120/e^6 + 0.143711/e^8 - 0.220432/e^{10}$ |
| (3,3)   | $-0.072627/e^6 + 0.02003/e^8 - 0.029703/e^{10}$ |
\begin{equation}
(4.0) \quad -0.048611a^4 - 0.074359a^6 + 0.021355a^8 - 0.055154a^{10}
\end{equation}

\begin{equation}
(4.2) \quad -0.108941a^6 - 0.013755a^8 - 0.050071a^{10}
\end{equation}

\begin{equation}
(4.4) \quad -0.016444a^8 - 0.010852a^{10}
\end{equation}

\begin{equation}
(5.1) \quad -0.043576a^6 - 0.030875a^8 - 0.034415a^{10}
\end{equation}

\begin{equation}
(5.3) \quad -0.026311a^8 - 0.021359a^{10}
\end{equation}

\begin{equation}
(5.5) \quad -0.005482a^{10}
\end{equation}

\begin{equation}
(6.0) \quad -0.003631a^6 - 0.009438a^8 - 0.011028a^{10}
\end{equation}

\begin{equation}
(6.2) \quad -0.013155a^8 - 0.017717a^{10}
\end{equation}

\begin{equation}
(6.4) \quad -0.009137a^{10}
\end{equation}

\begin{equation}
(7.1) \quad -0.003759a^8 - 0.009967a^{10}
\end{equation}

\begin{equation}
(7.3) \quad -0.005221a^{10}
\end{equation}

\begin{equation}
(8.0) \quad -0.000235a^8 - 0.001596a^{10}
\end{equation}

\begin{equation}
(8.2) \quad -0.001958a^{10}
\end{equation}

\begin{equation}
(9.1) \quad -0.000435a^{10}
\end{equation}

\begin{equation}
(10.0) \quad -0.000220a^{10}
\end{equation}

\begin{table}[h]
\centering
\caption{Single-magnon spectral weight series in real space. To evaluate the residue $A(q)$ carry out the sum described in the preceding table caption.}
\begin{tabular}{|c|c|}
\hline
\textbf{r} & \textbf{Series} \\
\hline
(0,0) & $0.5 - 0.041667a^2 + 0.011685a^4 - 0.00642a^6 + 0.024677a^8$ \\
\hline
(1,0) & $-0.666667a + 0.110185a^3 - 0.130751a^5 + 0.117059a^7 - 0.153306a^9$ \\
\hline
(1,1) & $0.194444a^2 + 0.084876a^4 - 0.035514a^6 + 0.092202a^8$ \\
\hline
(2,0) & $0.97222a^2 + 0.072647a^4 - 0.015601a^6 + 0.057647a^8$ \\
\hline
(2,1) & $-0.216667a^3 - 0.096930a^5 + 0.055191a^7 - 0.151490a^9$ \\
\hline
(2,2) & $0.065365a^4 + 0.008393a^6 + 0.030857a^8 - 0.045770a^{10}$ \\
\hline
(3,0) & $-0.036111a^3 - 0.049374a^5 + 0.006892a^7 - 0.045770a^9$ \\
\hline
(3,1) & $0.087153a^4 + 0.034750a^6 + 0.054776a^8$ \\
\hline
(3,2) & $-0.074005a^5 - 0.018980a^7 - 0.049672a^9$ \\
\hline
(3,3) & $0.018793a^6 + 0.015430a^8$ \\
\hline
(4,0) & $0.010894a^4 + 0.018817a^6 + 0.018378a^8$ \\
\hline
(4,1) & $-0.037003a^5 - 0.028024a^7 - 0.036099a^9$ \\
\hline
(4,2) & $0.028190a^6 + 0.029517a^8$ \\
\hline
(4,3) & $-0.023254a^7 - 0.021256a^9$ \\
\hline
(4,4) & $0.007614a^8$ \\
\hline
(5,0) & $-0.003700a^5 - 0.009765a^7 - 0.011880a^9$ \\
\hline
(5,1) & $0.011276a^6 + 0.021890a^8$ \\
\hline
(5,2) & $-0.013953a^7 - 0.018825a^9$ \\
\hline
(5,3) & $0.012182a^{10}$ \\
\hline
(5,4) & $-0.009245a^9$ \\
\hline
(6,0) & $0.000940a^6 + 0.004772a^8$ \\
\hline
(6,1) & $-0.004651a^7 - 0.011938a^9$ \\
\hline
(6,2) & $0.006091a^8$ \\
\hline
(6,3) & $-0.006284a^9$ \\
\hline
(7,0) & $-0.000323a^7 - 0.002181a^9$ \\
\hline
(7,1) & $0.001740a^{10}$ \\
\hline
(7,2) & $-0.002693a^9$ \\
\hline
(8,0) & $0.000109a^8$ \\
\hline
(8,1) & $-0.000673a^9$ \\
\hline
(9,0) & $-0.000037a^9$ \\
\hline
\end{tabular}
\end{table}
