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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 multiple-magnon contributions to neutron-scattering spectra are obtained for wave vectors 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.[1] 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.[2,3] 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[4] and single mode approximations.[5] The reliability of using only a few moments to reproduce spectral line shapes has come into question, for example, in the case of the two-magnon Raman spectra.[6] Currently efforts are underway to use a large number of frequency moments to obtain the dynamical properties by numerical analytical continuation.[7]

Recently one of us[8] 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 $(J_\perp/J_z)^{10}$.

Let us briefly summarize the results. The spin-wave spectrum has an energy-gap for $J_\perp<J_z$, which vanishes as the Heisenberg limit ($J_\perp=J_z$) is reached. Using series extrapolation methods to estimate the dispersion in the Heisenberg limit, we find a spin-wave velocity which agrees with $1/S$ expansions and other previous estimates. Along the line $q_x=q_y$, the dispersion is nearly uniformly renormalized with respect to its classical value. Measurable deviations are found in other directions. In particular, along the line $q_x+q_y=\pi$ the spin-wave energy is maximized at $(\pi/2,\pi/2)$ and it exceeds its value at $(\pi,0)$ by about 7%.

Another quantity that has been calculated by this method is the single-magnon contribution to neutron scattering, that is, the coefficient associated with the δ function in the dynamic structure factor. When compared with the equal-time transverse correlation function, it yields the relative weight of multiple-magnon and single-magnon contributions to the neutron-scattering spectra at different wave vectors. We find that over substantial parts of the Brillouin zone approximately 20% of the total spectral weight is associated with the multiple-magnon excitations even though the relative weight of such excitations vanishes near (0,0) and (π,π).

The Heisenberg-Ising Hamiltonian under consideration is defined by

$$\mathcal{H}=J_z\sum_{\langle i,j\rangle} S_i^zS_j^z+\alpha(S_i^xS_j^x+S_i^yS_j^y), \tag{1}$$

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 $\alpha=J_\perp/J_z$. 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 α. 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 wave vectors (q_x,q_y) and $(\pi-q_x,\pi-q_y)$. The full effective Hamiltonian to order α^{10} is presented in Table I.

For $\alpha\neq 1$ there is a gap in the spectrum, the minimum being at (0,0) and (π,π). The expansion for the gap is

$$2-(5/3)\alpha^2+0.31712963\alpha^4-0.41923376\alpha^6 +0.27099699\alpha^8-0.38943351\alpha^{10}+\cdots, \tag{2}$$

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 $(1/4)[\cos(q_xr_x+q_yr_y)+\cos(q_xr_x-q_yr_y)+\cos(q_xr_y+q_yr_x)+\cos(q_xr_y-q_yr_x)]$, and then dividing by 4.

r	Series
(0,0)	$8-0.666666\alpha^2+0.664352\alpha^4-0.292737\alpha^6+0.201076\alpha^8-0.177446\alpha^{10}$
(1,1)	$-4\alpha^2+1.222222\alpha^4-0.541756\alpha^6+0.513359\alpha^8-0.430609\alpha^{10}$
(2,0)	$-2\alpha^2+0.111111\alpha^4-0.290209\alpha^6+0.255912\alpha^8-0.331992\alpha^{10}$
(2,2)	$-0.291667\alpha^4-0.071979\alpha^6+0.060564\alpha^8-0.133137\alpha^{10}$
(3,1)	$-0.388889\alpha^4-0.177120\alpha^6+0.143711\alpha^8-0.220432\alpha^{10}$
(3,3)	$-0.072627\alpha^6+0.002003\alpha^8-0.029703\alpha^{10}$
(4,0)	$-0.048611\alpha^4-0.074359\alpha^6+0.021335\alpha^8-0.055154\alpha^{10}$
(4,2)	$-0.108941\alpha^6-0.013755\alpha^8-0.050071\alpha^{10}$
(4,4)	$-0.016444\alpha^8-0.010852\alpha^{10}$
(5,1)	$-0.043576\alpha^6-0.030875\alpha^8-0.034415\alpha^{10}$
(5,3)	$-0.026311\alpha^8-0.021359\alpha^{10}$
(5,5)	$-0.005482\alpha^{10}$
(6,0)	$-0.003631\alpha^6-0.009438\alpha^8-0.011028\alpha^{10}$
(6,2)	$-0.013155\alpha^8-0.017717\alpha^{10}$
(6,4)	$-0.009137\alpha^{10}$
(7,1)	$-0.003759\alpha^8-0.009967\alpha^{10}$
(7,3)	$-0.005221\alpha^{10}$
(8,0)	$-0.000235\alpha^8-0.001596\alpha^{10}$
(8,2)	$-0.001958\alpha^{10}$
(9,1)	$-0.000435\alpha^{10}$
(10,0)	$-0.000022\alpha^{10}$

which agrees completely with the "mass gap" calculated by Zheng *et al.*[3] General arguments tell us that the gap must close in the Heisenberg limit, $\alpha=1$. Moreover, we expect that for small $q=|\mathbf{q}|$ the spectrum has the form $\epsilon(\mathbf{q})\approx[A(\alpha)+B(\alpha)q^2]^{1/2}$, where $A(\alpha)\to 0$ as $\alpha\to 1$. 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 $\epsilon(\mathbf{q})$ in powers of q, $\epsilon(q)=C(\alpha)+D(\alpha)q^2+\cdots$, 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 $\alpha\to 1$ limit of the series

$$2C(\alpha)D(\alpha)=4\alpha^2-2.305555\alpha^4+2.410512\alpha^6-3.064895\alpha^8+4.100549\alpha^{10}+\cdots. \quad (3)$$

Since we expect only a weak, energylike singularity in this series at $\alpha=1$, we can sum it by Padé approximants. The near-diagonal approximants [2/2], [2/3], and [3/2] give estimates of 2.779, 2.785, and 2.785, respectively, which lead to values for the spin-wave velocity c/Ja of 1.667, 1.669, and 1.669. In the large-S limit $c/Ja=\sqrt{2}$, so the quantum renormalization of the spin-wave velocity is $Z_c\approx 1.18$. This number is in excellent agreement with high-order spin-wave calculations[9,10] and previous indirect estimates[11] using the hydrodynamic relation $c^2=\rho_s/\chi_\perp$, where ρ_s is the spin stiffness and $\chi_\perp$ the uniform transverse susceptibility.

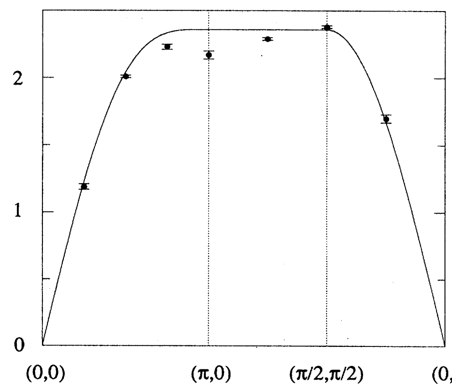

FIG. 1. Spin-wave spectrum for the Heisenberg model, in units of J, along three lines in reciprocal space. The solid circles with error bars are the results from the series expansions; the solid line is the classical spin-wave spectrum multiplied by an overall factor $Z_c=1.18$.

Away from the gapless points (0,0) and (π,π) the spin-wave spectrum for the Heisenberg model can be estimated by direct Padé approximants. Along the line $q_x=q_y$, the spectrum, within our numerical uncertainties, is uniformly renormalized with respect to the classical (large-S) spectrum. However, along the line $q_x=0$ appreciable differences appear. In particular, the excitation energy at $(\pi,0)$ appears to be a shallow local minimum along this line, and is lower than that at $(\pi/2,\pi/2)$ by about 7%. Note that at the classical and $1/S$ levels the spin-wave spectrum is degenerate at $(\pi,0)$ and $(\pi/2,\pi/2)$. A plot of the dispersion relation along selected directions is shown in Fig. 1. A hint of the deviations from classical spin-wave theory which we have found near $(\pi,0)$ can be seen in the results of the projector quantum Monte Carlo calculations of Chen *et al.*,[12] The first calculation of the spectrum via spin-wave expansion to order $1/S^2$ by Igarashi and Watabe[13] using the Holstein-Primakoff transformation also suggested that $(\pi,0)$ would be a local minimum for the spin-wave spectrum; but that minimum is no longer present in the more recent spin-wave calculations by Igarashi.[9] The spin-wave calculation by Canali *et al.*[10] based on the Dyson-Maleev transformation does yield results qualitatively similar to ours, with $\epsilon(\pi/2,\pi/2)/\epsilon(\pi,0)-1>0$, albeit with a value closer to 0.02 than 0.07. However, the two sets of results have a noteworthy discrepancy: those of Canali *et al.* indicate $\epsilon(\pi/2,\pi/2)$ should lie above the uniformly renormalized classical spectrum, rather than $\epsilon(\pi,0)$ lying below it, as we find.

We now turn to the spin-wave spectral weights. The magnetic neutron-scattering cross section is proportional to the dynamic structure factor, given by the expression

$$\hat{S}(\mathbf{q},\omega)=\int dt\, e^{-i\omega t}\sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}}\langle S^x(\mathbf{0},0)S^x(\mathbf{r},t)+S^y(\mathbf{0},0)S^y(\mathbf{r},t)\rangle. \quad (4)$$

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TABLE II. Single-magnon spectral weight series in real space. To evaluate the residue $A(\mathbf{q})$ carry out the sum described in the preceding table caption.

r	Series
(0,0)	$0.5-0.041667\alpha^2+0.011685\alpha^4-0.030642\alpha^6+0.024677\alpha^8$
(1,0)	$-0.666667\alpha+0.110185\alpha^3-0.130751\alpha^5+0.117059\alpha^7-0.153300\alpha^9$
(1,1)	$0.194444\alpha^2+0.084876\alpha^4-0.035514\alpha^6+0.092202\alpha^8$
(2,0)	$0.097222\alpha^2+0.072647\alpha^4-0.015601\alpha^6+0.057647\alpha^8$
(2,1)	$-0.216667\alpha^3-0.096930\alpha^5+0.055191\alpha^7-0.151490\alpha^9$
(2,2)	$0.065365\alpha^4+0.008393\alpha^6+0.030857\alpha^8$
(3,0)	$-0.036111\alpha^3-0.049374\alpha^5+0.006892\alpha^7-0.045770\alpha^9$
(3,1)	$0.087153\alpha^4+0.034750\alpha^6+0.054776\alpha^8$
(3,2)	$-0.074005\alpha^5-0.018980\alpha^7-0.049672\alpha^9$
(3,3)	$0.018793\alpha^6+0.015430\alpha^8$
(4,0)	$0.010894\alpha^4+0.018817\alpha^6+0.018378\alpha^8$
(4,1)	$-0.037003\alpha^5-0.028024\alpha^7-0.036609\alpha^9$
(4,2)	$0.028190\alpha^6+0.029517\alpha^8$
(4,3)	$-0.023254\alpha^7-0.021256\alpha^9$
(4,4)	$0.007614\alpha^8$
(5,0)	$-0.003700\alpha^5-0.009765\alpha^7-0.011880\alpha^9$
(5,1)	$0.011276\alpha^6+0.021890\alpha^8$
(5,2)	$-0.013953\alpha^7-0.018825\alpha^9$
(5,3)	$0.012182\alpha^8$
(5,4)	$-0.009425\alpha^9$
(6,0)	$0.000940\alpha^6+0.004772\alpha^8$
(6,1)	$-0.004651\alpha^7-0.011938\alpha^9$
(6,2)	$0.006091\alpha^8$
(6,3)	$-0.006284\alpha^9$
(7,0)	$-0.000332\alpha^7-0.002181\alpha^9$
(7,1)	$0.001740\alpha^8$
(7,2)	$-0.002693\alpha^9$
(8,0)	$0.000109\alpha^8$
(8,1)	$-0.000673\alpha^9$
(9,0)	$-0.000037\alpha^9$

We consider the $T=0$ limit, where the angular brackets refer to ground-state expectation values. In general, we expect $\hat{S}(\mathbf{q},\omega)$ to consist of a sum of two parts,

$$\hat{S}(\mathbf{q},\omega)=A(\mathbf{q})\,\delta[\omega-\epsilon(\mathbf{q})]+B(\mathbf{q},\omega). \tag{5}$$

Here $\epsilon(\mathbf{q})$ is the spin-wave dispersion and $A(\mathbf{q})$ is the residue or spectral weight associated with the spin-waves. $B(\mathbf{q},\omega)$ is associated with multiple-magnon excitations, which are present because a single spin flip in an antiferromagnet cannot be exactly represented as a superposition of single-magnon states. Integrating $\hat{S}(\mathbf{q},\omega)$ over all frequencies yields the equal-time correlation function

$$\hat{S}(\mathbf{q})=\sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}}\langle S^x(\mathbf{0},0)S^x(\mathbf{r},0)+S^y(\mathbf{0},0)S^y(\mathbf{r},0)\rangle. \tag{6}$$

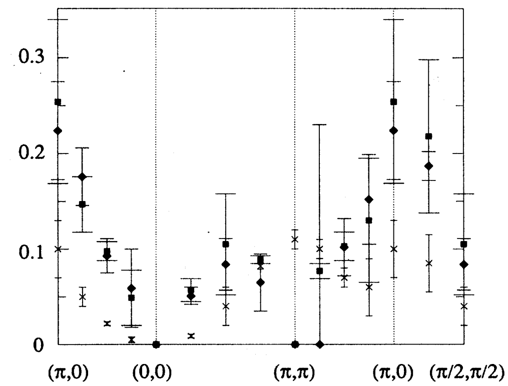

FIG. 2. Heisenberg model multiple-magnon spectral weight (crosses, narrowest error bars) and the ratio of the multiple-magnon spectral weight to the total spectral weight as determined by extrapolations for the multiple-magnon weight series (solid squares, widest error bars) and by the difference of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

To determine the residue $A(\mathbf{q})$, we need to restrict the intermediate states that arise in the calculation of the expectation values to be single-magnon states, which gives the expression

$$A(\mathbf{q})=\sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}}\langle S^x(\mathbf{0},0)\mathscr{P}S^x(\mathbf{r},0)+S^y(\mathbf{0},0)\mathscr{P}S^y(\mathbf{r},0)\rangle, \tag{7}$$

where $\mathscr{P}$ 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

$$|\psi_i\rangle=|i\rangle+\sum_n C_{i,n}(\alpha)|n\rangle, \tag{8}$$

where $|i\rangle$ is a single-magnon state in the Ising limit and $|n\rangle$ represents basis states (eigenstates in the Ising limit) which are not degenerate with the single-magnon states. However, the states $|\psi_i\rangle$ for different i are not orthogonal to each other when $\alpha\neq 0$. Thus in order to construct the projection operator, we need to define the overlap matrix $g_{i,j}=\langle\psi_i|\psi_j\rangle$. Then the projection operator onto the single-magnon subspace is given by the expression[14]

$$\mathscr{P}=\sum_{i,j} g_{i,j}^{-1}|\psi_i\rangle\langle\psi_j|. \tag{9}$$

The expansion coefficients for the residues in real space as a function of the vector distance are given in Table II. The coefficients for the transverse structure factor are given in Ref. 5.

We can now estimate the multiple-magnon contribution to neutron scattering by simply subtracting $A(\mathbf{q})$ from the equal-time correlation $\hat{S}(\mathbf{q})$. To get to the Heisenberg limit a series extrapolation is needed. Since the series for the multiple-magnon weights is 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 multiple-magnon weights, carrying out the extrapolations by direct Padé approximants. In Fig. 2 results are presented for both the multiple-magnon spectral weight as well as the ratio of the multiple-magnon weight to the total spectral weight, along several lines in the Brillouin zone. We see that the multiple-magnon contribution is particularly large near $(\pi,0)$ (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[13] yield roughly twice as much spectral weight in the multiple-magnon excitations as we find by expansions about the Heisenberg-Ising model; however, given Igarashi's later remarks[9] about the incorrect treatment of umklapp processes in that work, we do not view the discrepancy as significant. Furthermore, the spin-wave calculation of Canali and Wallin[15] yields multiple-magnon weights consistent with ours.

Recently, Stringari[16] has developed general bounds and sum rules for single-magnon and multiple-magnon spectra at special wave vectors. Near $\mathbf{q}=(0,0)$ the single-magnon spectral weight vanishes linearly while the multiple-magnon weight vanishes quadratically; and near $\mathbf{q}=(\pi,\pi)$, the single-magnon spectral weight diverges as $|(\pi,\pi)-\mathbf{q}|^{-1}$ while the multiple-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 multiple-magnon spectral weights have been estimated throughout the Brillouin zone. In light of our numerical results, which largely confirm the deviations from linear spin-wave theory found in the higher-order calculations by Canali *et al.*,[10,15] it would be interesting to examine the inelastic-neutron-scattering data on the antiferromagnetic parent compounds of the cuprate superconductors to look for the multiple-magnon excitations and variations in the spin-wave energies along the line $q_x+q_y=\pi$.

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