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Quantum renormalizations in the spin-1 Heisenberg antiferromagnet on the square lattice

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The renormalized parameters, characterizing the static and dynamic low-temperature properties of the spin-1 Heisenberg antiferromagnet on the square lattice, are estimated by expansions around the Ising limit. The calculations include the renormalization of the perpendicular susceptibility ($\chi_\perp$) and the frequency moments of the inelastic light scattering spectra from spin-pair excitations. The results are used to assess the quantitative accuracy of the spin-wave calculations of Oguchi for $\chi_\perp$ and the two-magnon calculation of Parkinson for the light scattering spectra. They are found to be in agreement with experiments on K_2NiF_4.

Recent experiments on light scattering[1] and neutron scattering[2] from "high-T_c" materials have provided dramatic evidence for enhanced quantum fluctuations at low-temperatures in two-dimensional magnetic systems. The statics and dynamics of the neutron scattering experiments have been interpreted[3-5] in terms of a "renormalized classical theory" for two-dimensional Heisenberg antiferromagnets (2D HAFM). Quantum renormalization of the various parameters entering such a description, for the spin-$\frac{1}{2}$ Heisenberg antiferromagnet appropriate to the CuO_2 materials, have been calculated in a controlled and systematic manner by expansions around the Ising limit.[6] The shape of the light scattering spectra from spin-pair excitations have also been shown[7] to be in agreement with those of $S=\frac{1}{2}$ 2D HAFM once quantum renormalizations are taken into account.

Similar experiments were performed many years ago[8,9] on K_2NiF_4 and were interpreted without considering any quantum renormalizations.[10] Since K_2NiF_4 is a 2D HAFM with $S=1$, one expects quantum fluctuations to be weaker in this case but not absent. The purpose of this paper is to present a systematic calculation of the renormalized parameters for the $S=1$ case and compare them with the light and neutron scattering experiments. We find that the primary effect of quantum fluctuations is to renormalize the spin-wave spectrum to higher energies. Furthermore, similar to the $S=\frac{1}{2}$ case, the long-wavelength and short-wavelength dispersion relations are renormalized roughly proportionately.[7] Since the exchange constant deduced from the light scattering and neutron scattering experiments depend on precisely these renormalizations, it is not surprising that the unrenormalized theory works well with an effective value for the exchange constant.

For the basic thermodynamic parameters of the spin-one model, we estimate its ground-state energy E_0, the sublattice magnetization M^+, and the perpendicular susceptibility $\chi_\perp$. M^+ and E_0 have been studied previously by series expansion[11] and finite-size diagonalization[12] studies. Since it is $\chi_\perp$ which deviates the most from the order $1/S$ spin-wave value[13] in the case of $S=\frac{1}{2}$, we expect it to set the accuracy of the spin-wave estimates for the $S=1$ case also. We find that our estimate ($\chi_\perp J=0.095 \pm 0.002$) differs from the spin-wave theory ($\chi_\perp J=0.0905$) by less than 5%.

In addition, we calculate the first two frequency moments of the light scattering spectra from spin-pair excitations. The first moment (ρ_1) gives a measure of the peak position and can be used to extract the exchange constant J from the experiments.[7] The second cumulant[14] (M_2) gives a measure of the width of the peak. In particular, the ratio (R) of M_2 to ρ_1 gives us the peak width relative to its position in a parameter-free manner and thus is a characteristic of 2D HAFM with nearest-neighbor exchanges. We find that for K_2NiF_4 the values for the exchange constant J deduced from light scattering and neutron scattering experiments agree to within a percent. The ratio R, which is much more uncertain in theory, agrees with experiments within 20%.

The spin-one Heisenberg-Ising Hamiltonian is given by

$$\mathcal{H}=J\sum_{\langle i,j\rangle} S_i^z S_j^z + J_{xy}\sum_{\langle i,j\rangle}(S_i^x S_j^x + S_i^y S_j^y) + H\sum_i S_i^x\,, \tag{1}$$

where the sum $\langle i,j\rangle$ runs over all nearest-neighbor pairs of spins, and H is a uniform perpendicular field. The sublattice magnetization is defined by the relation

$$M^+=\langle S_0^z\rangle\,, \tag{2}$$

while the susceptibility is defined through the ground-state energy per spin, $E(H)$, via the relation

$$E(H)=E_0-\tfrac{1}{2}\chi_\perp H^2+\cdots\,. \tag{3}$$

The expansion in powers of $x(=J_{xy}/J)$ are developed by the method of Singh, Gelfand, and Huse.[15] The expansions are (quoting six significant digits)

$$M^+=1-0.0816326x^2-0.0269591x^4-0.0136997x^6-0.00880047x^8-0.00625520x^{10}+\cdots\,, \tag{4}$$

$$2E_0/J=-4-0.571428x^2-0.0504597x^4-0.0144762x^6-0.00656238x^8-0.00358690x^{10}+\cdots\,, \tag{5}$$

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and

$$\tfrac{1}{2}\chi_\perp J = \tfrac{1}{8} - 0.142857x + 0.144643x^2 - 0.149916x^3 + 0.150672x^4 - 0.153095x^5 + 0.153437x^6 - 0.154932x^7 + \cdots . \quad (6)$$

The series are analyzed in a manner analogous to the $S=\frac{1}{2}$ case.[6] The subtleties of the expansion techniques and the details of the analysis will be discussed elsewhere.[16] We estimate, at the Heisenberg point ($x=1$),

$$E_0/J = -2.327 \pm 0.001 , \quad (7)$$

$$M^+ = 0.81 \pm 0.01 , \quad (8)$$

and

$$\chi_\perp J = 0.095 \pm 0.002 . \quad (9)$$

We now discuss the calculation of the light scattering spectra. Light scattering from spin-pair excitations in an insulating antiferromagnet can be described by an effective Hamiltonian[10]

$$H_R = \sum_{\langle i,j \rangle} (\mathbf{E}_{\rm inc} \cdot \hat{\boldsymbol{\sigma}}_{ij})(\mathbf{E}_{\rm sc} \cdot \hat{\boldsymbol{\sigma}}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j , \quad (10)$$

where $\mathbf{E}_{\rm inc}$ and $\mathbf{E}_{\rm sc}$ are the incident and scattered electric field intensities and $\hat{\boldsymbol{\sigma}}_{ij}$ is a unit vector connecting spin sites i and j. In the absence of any resonant scattering one expects only the nearest-neighbor terms in the sum to be significant.[7,8,10] This leads to scattering in the B_{1g} geometry only.[7] The scattering Hamiltonian for this geometry is

$$H_R = A \sum_i \mathbf{S}_i \cdot (\mathbf{S}_{i+\hat{\mathbf{x}}} - \mathbf{S}_{i+\hat{\mathbf{y}}}) . \quad (11)$$

Here the sum runs over all sites of the lattice, A is an undetermined constant that gives the overall magnitude of the Raman cross section, and $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are unit vectors along the x and y directions. The scattered intensity at $T=0$ is given by

$$I(\omega) = \sum_k \delta[\omega - (E_k - E_0)] \, | \langle 0 | H_R | k \rangle |^2 , \quad (12)$$

where $|k\rangle$ represent the eigenstates of the Heisenberg Hamiltonian and E_k are the corresponding energies. $|0\rangle$ represents the ground state. The frequency moments can be expressed in terms of the ground-state expectation values of multiple spin operators[7]

$$\begin{aligned} I_T &= \langle 0 | H_R^2 | 0 \rangle , \\ \rho_1 &= \langle 0 | H_R [H, H_R] | 0 \rangle / I_T , \\ \rho_2 &= -\langle 0 | [H, H_R]^2 | 0 \rangle / I_T \cdots . \end{aligned} \quad (13)$$

Series expansions for these moments are

$$\begin{aligned} 2\rho_1/J = {} & 14 + 0x + 0.530612x^2 - 0.141138x^3 \\ & + 0.0338370x^4 + 0.0171678x^5 + \cdots , \end{aligned} \quad (14)$$

$$\begin{aligned} 4\rho_2/J^2 = {} & 196 + 0x + 17.4694x^2 - 5.33453x^3 \\ & + 3.32992x^4 + 0.626767x^5 + \cdots . \end{aligned} \quad (15)$$

Since the terms in the series for ρ_1 and ρ_2 decrease rapidly in magnitude, they can be estimated quite accurately. However, the second cumulant (M_2) defined by

$$(M_2)^2 = \rho_2 - \rho_1^2 \quad (16)$$

cannot be properly estimated this way as ρ_2 nearly equals ρ_1^2. Hence, we construct the series expansion for $(M_2)^2$ and extrapolate that to get M_2. We obtain

$$\begin{aligned} 4(M_2)^2/J^2 = {} & 2.61224x^2 - 1.38267x^3 \\ & + 2.10093x^4 + 0.29585x^5 + \cdots . \end{aligned} \quad (17)$$

By adding up terms in the series as well as using Padé approximants we estimate

$$\rho_1/J = 7.22 \pm 0.02, \quad \rho_2/J^2 = 53.0 \pm 0.3 , \quad (18)$$

and

$$R \equiv M_2/\rho_1 = 0.12 \pm 0.03 .$$

Notice that because the series for M_2 does not decrease rapidly in magnitude, the uncertainties in R are large.

Let us now compare these results with the two-magnon calculation of Parkinson[10] and the experiments on K_2NiF_4.[8] The first moment derived from Parkinson's formula is $\rho_1 = 6.63J$. This should be corrected for the renormalization of the spin-wave spectrum, due to quantum fluctuations. Oguchi's[13] order $1/S$ calculation provides a k-independent multiplicative renormalization of the spin-wave spectrum by an amount $Z_c = 1.08$. Taking this into account, the two-magnon spectra should have a first moment of $7.16J$. This differs from Eq. (18) by less than a percent, suggesting that the renormalization is indeed k independent. Hence, the earlier agreement between the values for the exchange constants for K_2NiF_4 deduced from neutron scattering and light scattering experiments using unrenormalized theory remains valid with the renormalized parameters as well. From Fig. 1 in Ref. 8 we obtain $\rho_1 \approx 515$ cm^{-1}, leading to $J \approx 71$ cm^{-1} for K_2NiF_4.

The ratio R, which is not affected by the renormalization of the spin-wave dispersion, equals 0.09 in the two-magnon calculation, whereas it is 0.12 ± 0.03 for the series estimate. Because the line is very narrow for the $S=1$ case (compared to $S=\frac{1}{2}$) the uncertainty in the width computed from the expression $\rho_2 - \rho_1^2$ is large. Even though the series is not accurate enough, it appears that the ratio R is underestimated in the two-magnon calculation on the order of 30%. Let us remember that the latter calculation has a sharp cutoff for spin-pair excitations at energy $8J$. There is no such sharp cutoff in a fully quantum-mechanical system, as the excitation of a pair of spins about the Heisenberg ground state involves a mixture of 2,4,6,... magnon states. The experiments[8] on K_2NiF_4 have a line shape in excellent agreement with Parkinson's calculations and the ratio R roughly equals 0.1 for Fig. 1 of Ref. 8. This suggests that the two-magnon part of the spectra is accurately described by Parkinson's calculation. The difference between the two theoretical results arises from scattering beyond the classical cutoff energy primarily due to the admixture of four-magnon states in the spin-pair excitation spectrum. This scattering is weak enough to leave the first moment unaffected, but because the line is quite narrow, contributes significantly to the second cumulant. This explains why the experimental cumulant, obtained by truncating the spectra at an energy around $8J$, is also lower than the series estimate. In fact, since the width of the experimental line shape is about a tenth of its peak position, roughly 1% integrated intensity around twice the peak position can easily account for a 30% difference. Such a weak scattering, especially if spread over a certain energy range, may be difficult to distinguish from the background. It may, nevertheless, prove useful to redo the experiments at higher energies to see if there is some observable scattering there.

We now compare the role of quantum fluctuations in the spin-$\frac{1}{2}$ and spin-1 systems. In both cases the spin-wave spectrum appears to be renormalized upwards in a wave-vector-independent manner. The order $1/S$ spin-wave calculation for the susceptibility differs from the series estimates by less than a few percent for the $S=1$ case, whereas for $S=\frac{1}{2}$ there is about a 15% difference.[6] As for the spin-pair excitation spectra, the width relative to the peak position is enhanced due to quantum fluctuations in both cases, mostly due to the admixture of higher-magnon states. The change is about 30% for $S=1$, and about a factor of 2 for $S=\frac{1}{2}$. It is worth noting that in Parkinson's calculations[10] the spin-half case stands separate from all higher spins. This is because the line shape, which changes gradually as a function of the parameter S for large S, is altered substantially between $S=1$ and $S=\frac{1}{2}$. For $S=\frac{1}{2}$, the peak position is greatly shifted from the classical cutoff value, and the asymmetry of the line shape changes sign. (The half width at half maximum, which increases gradually as S is reduced from infinity to one, begins to decrease between $S=1$ and $S=\frac{1}{2}$. Also, the third cumulant, which is negative for $S\geq 1$, becomes positive for $S=\frac{1}{2}$.) This may be an indication that even the purely two-magnon line shape is less reliable in the spin-wave calculation for $S=\frac{1}{2}$.

An additional feature of the Raman scattering in La_2CuO_4 is the observed peaks in A_{1g} and B_{2g} geometries. This was explained[7] by assuming that light could overturn diagonal next-neighbor (DNN) pairs of spins in addition to the nearest-neighbor ones. It was shown that such a DNN term did not give rise to any scattering in the classical limit. With quantum fluctuations it gave rise to scattering in A_{1g} and B_{2g} geometries. However, it did not affect the B_{1g} cross section at all. These would remain true for a spin-1 system as well. The amplitude for such a DNN spin-pair excitation was found to be comparable to the nearest-neighbor one in La_2CuO_4. However, in the case of K_2NiF_4, there is no reason to expect a significant amplitude for DNN spin-pair excitations. This is because in K_2NiF_4 there are no optically active states of the system near the incident laser energies. Hence, one expects the amplitude for a given spin-pair excitation to be proportional, within an order of magnitude, to the corresponding exchange constants in the Hamiltonian. Since the electronic overlaps for nearest neighbors are orders of magnitude larger than further-neighbor ones in K_2NiF_4, the nearest-neighbor spin-pair excitations should dominate the scattering process, and the selection rules giving negligible scattering in A_{1g} and B_{2g} geometries would continue to hold.

Before we conclude, we would like to make a few remarks comparing the present calculational technique with the others. While the Ising expansions do not provide full information on the spectral line shape (as compared with the spin-wave calculation) they represent systematic and controlled calculations of the parameters of the model. The series obtained are convergent and hence the accuracy can be improved by going to higher orders. Thus, they can be used to judge the quantitative validity of other uncontrolled (such as spin wave) calculations or to fit parameters in a renormalization-group-type theory (such as the Chakravarty, Halperin, and Nelson theory for 2D HAFM). Furthermore, unlike 1D, where finite-size studies are very effective, it is unlikely that in higher dimensions one can study a system of comparable size by exact diagonalization methods. The series presented in this paper include contributions from spin-spin correlations with spins distant ten sites apart along any axis. In contrast, exact diagonalization even for a 4×4 system represents a major achievement.[11] The method of series expansions works by dividing the larger problem into many small pieces.[15]

Our calculations presented here illustrate the role of quantum fluctuations in the $S=1$ system in two dimensions. The primary effect of these fluctuations is to renormalize the spin-wave spectrum, in a manner roughly independent of wave vector. Other properties such as the perpendicular susceptibility and frequency moments of the inelastic light scattering spectra are also renormalized. The spin-wave calculations are quantitatively accurate to a few percent when corrections of order $1/S$ are taken into account. A recent reanalysis of the correlation length data from the neutron scattering experiments[17] is found to be in excellent agreement with the spin-wave theory, but the data is probably not accurate enough to disentangle 5% effects. It would be interesting to see if light scattering is observable beyond the classical cutoff frequency.

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