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APPLICATION OF THE THERMAL BOSON EXPANSION TO THE HEISENBERG ANTIFERROMAGNET MnF_2

M. BRAJCZEWSKA, C. FIOLHAIS and J. DA PROVIDÊNCIA
Departamento de Física da Universidade de Coimbra, 3000 Coimbra, Portugal

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We apply the thermal boson expansion which has been presented in refs. [1–3] to the Heisenberg isotropic antiferromagnet, submitted to an external field. Considering only spin waves with wave vectors up to a certain cut-off, the magnetization and the principal susceptibilities are calculated. The results are compared with the experimental data on MnF_2.

1. Introduction

Let us consider a Heisenberg antiferromagnet made up of $2N$ spins, the interactions being restricted to the nearest neighbours.

We take only lattices which may be decomposed into two interpenetrating sublattices, denoted by j (sublattice a, for the spin up sublattice) and l (sublattice b, for the spin down sublattice), such that the nearest neighbours of an atom in one sublattice all belong to the other sublattice.

The simplest form of the Hamiltonian of an ideal antiferromagnet, under an external magnetic field, is

$$\mathscr{H} = J \sum_{jl} S_j \cdot S_l + Hg\mu_B \Big(\sum_j S_j^z + \sum_l S_l^z \Big) \tag{1.1}$$

where $J > 0$ is the exchange integral between nearest neighbours, H is the external magnetic field directed along the z-axis, g is the Landé factor and μ_B the Bohr magneton. For the sake of simplicity, anisotropic interactions are not taken into account, although they may be important for real magnetic materials (MnF_2, for instance, shows a small amount of anisotropy [7]).

We apply to this model the thermal boson expansion which has been introduced in ref. [1] and applied to the ferromagnetic case in refs. [2, 3]. We wish to calculate the magnetization and the principal susceptibilities $\chi_\parallel$ and $\chi_\perp$. In order to obtain the perpendicular susceptibility $\chi_\perp$, the second term of (1.1) is replaced by

$$H_x g\mu_B \Big(\sum_j S_j^x + \sum_l S_l^x \Big) . \tag{1.2}$$

The interest in studying antiferromagnets is well known:

1) The ground state, on which the spin wave excitations are built, is not an eigenstate of the Hamiltonian, as it is in the ferromagnetic Heisenberg model. The sublattice magnetization shows a zero-point spin deviation.

2) There is a kinematic interaction between spin waves even at low temperatures.

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compare them with the experimental data available on MnF_2.
The conclusions appear in section 4.

2. Thermal boson expansion

The arguments follow closely the treatment of the ferromagnetic problem [1, 2], the main difference being that there are two sublattices, each one requiring a separate boson expansion.

The Fourier transforms of $\mathbf{S}_j$ and $\mathbf{S}_l$ are

$$\begin{aligned} S^{\pm}_{K\mathrm{a}} &= \frac{1}{\sqrt{N}} \sum_j \mathrm{e}^{\pm \mathrm{i}KR_j} S^{\pm}_j ,\\ S^{z}_{K\mathrm{a}} &= \frac{1}{\sqrt{N}} \sum_j \mathrm{e}^{\mathrm{i}KR_j} S^{z}_j ,\\ S^{\pm}_{K\mathrm{b}} &= \frac{1}{\sqrt{N}} \sum_l \mathrm{e}^{\pm \mathrm{i}KR_l} S^{\pm}_l ,\\ S^{z}_{K\mathrm{b}} &= \frac{1}{\sqrt{N}} \sum_l \mathrm{e}^{\mathrm{i}KR_l} S^{z}_l , \end{aligned} \tag{2.1}$$

where the sums for $S^{\pm}_{K\mathrm{a}}$ and $S^{z}_{K\mathrm{a}}$ extend over the N atoms j of the sublattice a, while the sums for $S^{\pm}_{K\mathrm{b}}$, $S^{z}_{K\mathrm{b}}$ extend over the atoms l of the sublattice b.

The commutation relations for the spin operators $S^{\pm}_{K\mathrm{a}}$ are given by

$$[S^{+}_{K\mathrm{a}}, S^{-}_{K'\mathrm{a}}] = \frac{2}{\sqrt{N}} S^{z}_{K-K'\mathrm{a}}, \quad [S^{z}_{K\mathrm{a}}, S^{\pm}_{K'\mathrm{a}}] = \pm \frac{1}{\sqrt{N}} S^{\pm}_{K'\pm K\mathrm{a}} . \tag{2.2}$$

The same relations hold for $S^{\pm}_{K\mathrm{b}}$, while the commutators between spin operators of different sublattices vanish.

Let us construct a boson mapping of the spin operators. The essential idea of the thermal boson expansion consists in preserving the commutation relations (the Lie algebra of the spin operators reproduced by the boson images) and all mean-field expectations values (the mean field is described by an independent particle density matrix).

Two different kinds of boson operators (B_K, B^{+}_K and C_K, C^{+}_K) are needed, one set for each sublattice

$$[B_K, B^{+}_{K'}] = \delta_{KK'}, \ [C_K, C^{+}_{K'}] = \delta_{KK'} . \tag{2.3}$$

The mean-field density matrix is written as an exponential of a one-body operator

$$\mathscr{D}_0 = A\, \mathrm{e}^{\alpha[\Sigma_j S^z_j - \Sigma_l S^z_l]} , \tag{2.4}$$

where α is a variational parameter and A a normalization constant.

Using the technique of ref. [2], we obtain the following boson expansions (up to the first order)

$$
\begin{aligned}
(S_j^+)_B &= \frac{1}{\sqrt{N}} \sum_K e^{-iKR_j} \sqrt{2SX}\, B_K + \cdots ,\\
(S_j^-)_B &= \frac{1}{\sqrt{N}} \sum_K e^{iKR_j} \sqrt{2SX}\, B_K^+ + \cdots ,\\
(S_j^z)_B &= SX - \frac{1}{N} \sum_{KK'} e^{-i(K-K')R_j} B_K^+ B_{K'} ,\\
(S_l^+)_B &= \frac{1}{\sqrt{N}} \sum_K e^{-iKR_l} \sqrt{2SX}\, C_K^+ + \cdots ,\\
(S_l^-)_B &= \frac{1}{\sqrt{N}} \sum_K e^{iKR_l} \sqrt{2SX}\, C_K + \cdots ,\\
(S_l^z)_B &= -SX + \frac{1}{N} \sum_{KK'} e^{-i(K-K')R_l} C_K^+ C_{K'} ,
\end{aligned}
\tag{2.5}
$$

where

$$
X = B_S(S\alpha) = \frac{2S+1}{2S} \coth\left(\frac{2S+1}{2}\, \alpha\right) - \frac{1}{2S} \coth\left(\frac{\alpha}{2}\right)
$$

is the Brillouin function.

The mean-field equations of motion are classical ones. We hope to take quantal effects adequately into account by quantizing the mean-field equations. Quantal dynamics is then described by the bosonized Hamiltonian given by:

$$
\begin{aligned}
(\mathscr{H})_B = &-NJzS^2X^2 + JzSX \sum_K [\gamma_K B_K C_K + \gamma_K B_K^+ C_K^+ + B_K^+ B_K + C_K^+ C_K]\\
&+ Hg\mu_B\Big[NSX - \sum_K B_K^+ B_K\Big] + Hg\mu_B\Big[-NSX + \sum_K C_K^+ C_K\Big] ,
\end{aligned}
\tag{2.6}
$$

with z the number of nearest neighbours of an atom and $\gamma_K = 1/z \sum_\delta e^{iK\delta}$ (the sum being over the z nearest neighbours).

In order to diagonalize $(\mathscr{H})_B$, we make use of the well-known Bogoliubov transformation for B_K, B_K^+, C_K, C_K^+ to new boson operators α_K, α_K^+, β_K, β_K^+, which is defined by

$$
\begin{aligned}
\alpha_K &= u_K B_K - \nu_K C_K^+ , \quad \alpha_K^+ = u_K B_K^+ - \nu_K C_K ,\\
\beta_K &= u_K C_K - \nu_K B_K^+ , \quad \beta_K^+ = u_K C_K^+ - \nu_K B_K ,
\end{aligned}
\tag{2.7}
$$

where u_K and ν_K are real numbers satisfying $u_K^2 - \nu_K^2 = 1$. This relationship assures that

$$
[\alpha_K, \alpha_K^+] = 1, \quad [\beta_K, \beta_K^+] = 1, \quad [\alpha_K, \beta_K] = 0 . \tag{2.8}
$$

In the new boson variables the Hamiltonian (2.6) reads as

$$
\begin{aligned}
(\mathscr{H})_B = &-NJzS^2X^2 + \sum_K \omega_K(\beta_K^+ \beta_K + \alpha_K^+ \alpha_K)\\
&- JzSX \sum_K (1 - \sqrt{1-\gamma_K^2}) + Hg\mu_B \sum_K (\beta_K^+ \beta_K - \alpha_K^+ \alpha_K) ,
\end{aligned}
\tag{2.9}
$$

with $\omega_K(X) = JzSX\sqrt{1-\gamma_K^2}$. Expression (2.9) may still be written as

$$(\mathcal{H})_B = -NJzS^2X^2 - JzSX\sum_K (1-\sqrt{1-\gamma_K^2}) + \sum_K A_K n_K + \sum_K A_K^+ n_K' , \tag{2.10}$$

with $h = Hg\mu_B/JzSX$, $A_K^\pm = JzSX(\sqrt{1-\gamma_K^2} \pm h)$, $n_K = \alpha_K^+\alpha_K$ and $n_K' = \beta_K^+\beta_K$. The energy consists therefore of a mean-field term, a zero-point energy and two boson terms. The zero point energy reflects the complicated structure of the exact ground state.

The free energy is obtained adding to (2.10) the statistical expectation value of the entropy (which consists also of a mean-field and a boson term)

$$F = -NJzS^2X^2 - JzSX\sum_K (1-\sqrt{1-\gamma_K^2}) + 2N\beta^{-1}\left[\log\left(\sinh\left(\frac{\alpha}{2}\right)\Big/\sinh\left(\frac{2S+1}{2}\alpha\right)\right) + S\alpha X\right]$$

$$+\beta^{-1}\sum_K \log[1-e^{-\beta A_K^-}] + \beta^{-1}\sum_K \log[1-e^{-\beta A_K^+}] , \tag{2.11}$$

with $\beta = (k_B T)^{-1}$, k_B being the Boltzmann constant and T the temperature. The free energy should be minimized with respect to the parameter α. This minimization procedure leads to a transcendental equation which gives X as a function of the temperature. In the case of $H = 0$, we have:

$$\alpha - \beta JzSX + \frac{\beta}{NS}\sum_K \omega_K(1) n_K(X) = 0 \tag{2.12}$$

where $\omega_K(1)$ is the antiferromagnetic spin wave energy at absolute zero temperature and zero external field.

3. Results

We have solved numerically eq. (2.12), considering crystals of NaCl- (sc lattice) and CsCl- (bcc lattice) types. Following ref. [2], we have introduced a cut-off in momentum space $k_m < k_{Bz}$ (with k_{Bz} corresponding to the boundary of the first Brillouin zone). The first Brillouin zone was replaced by a sphere with an adequate radius and the sums over k were replaced by integrals.

There are fundamental reasons for using the cut-off. As discussed in [3], only the collective degrees of freedom should be bosonized (the intrinsic degrees of freedom are supposed to be well described by the mean-field). Although more sophisticated techniques may be utilized [3], it is enough for the present purposes to take the standard form of the thermal boson expansion with a phenomenological value of k_m, as in ref. [2].

The sublattice magnetization as a function of the temperature is given by:

$$M = \frac{g\mu_B}{V}\left[NSX - \sum_K \frac{n_K}{\sqrt{1-\gamma_K^2}} - \frac{1}{2}\sum_K\left(\frac{1}{\sqrt{1-\gamma_K^2}} - 1\right)\right], \tag{3.1}$$

where V the volume of the sample. The first term is a mean-field result, the second is a spin wave effect and the last is the zero-point spin deviation. At low temperatures ($X \simeq 1$), eq. (3.1) yields the asymptotic behaviour characteristic of antiferromagnets (it goes with T^2).

The parallel susceptibility $\chi_\parallel$ is defined by:

$$\chi_\parallel = -\frac{1}{V}\frac{\partial^2 F}{\partial H^2}\bigg|_{H=0} . \tag{3.2}$$

Using eq. (2.11) we obtain

$$\chi_\parallel = 2\beta g^2 \mu_B^2 \sum_K \frac{e^{-\beta\omega_K}}{(1-e^{-\beta\omega_K})^2} . \tag{3.3}$$

On the other hand, it is straightforward to conclude that the perpendicular susceptibility, $\chi_\perp$, in the harmonic approximation, is independent of the temperature ($\chi_\perp = 560 \times 10^{-6}$ per gram).

Expression (3.3) with $X = 1$ agrees with the first term of the expansion in powers of temperature, around $T = 0$, of $\chi_\parallel$ given by Oguchi [4].

We are now going to evaluate for each spin the value of the cut-off required for reproducing the empirical critical temperature, as given by the Rushbrooke–Woods formula [5]:

$$\frac{k_B T_N}{J} = \frac{k_B T_C}{2J}\left(1 + \frac{2}{3zS(S+1)}\right) , \tag{3.4}$$

with

$$\frac{k_B T_C}{J} = \frac{5}{96}(z-1)[11S(S+1)-1] .$$

Our "critical" point corresponds to the maximal temperature at which eq. (2.12) has a real solution. From table I, which shows the results for the maximal temperature without any cut-off ($k_m = k_{Bz}$), we conclude that such a cut-off is indeed required to achieve an agreement with formula (3.4). This is particularly true for spins $S \gg 1$, as in the ferromagnetic case [2], but even for $S = \frac{1}{2}$ the cut-off is needed.

We note that the upper point at which the self-consistent equation (2.12) still has a solution should not be interpreted as a physical (continuous) phase transition, but corresponds to the breakdown of the present temperature dependent spin wave scheme. The maximal temperature attained without any momentum cut-off is systematically lower than the real critical point.

Table I
Critical temperature in units of J/k_B for bcc and sc antiferromagnets (note that the J used in the present work is twice of that normally used). RW denotes Rushbrooke-Woods formula and TBM denotes thermal boson method.

S	bcc		sc	
	RW	TBM ($k_m = k_{Bz}$)	RW	TBM ($k_m = k_{Bz}$)
0.5	1.47	1.24	1.08	0.94
1.0	3.99	3.15	2.89	2.36
1.5	7.50	5.67	5.40	4.25
2.0	12.01	8.81	8.62	6.61
2.5	17.53	12.56	12.56	9.41

Table II
Cut-off momentum in units of the Brillouin momentum for the bcc and the sc antiferromagnets, which gives the Rushbrooke–Woods Néel temperature.

S	bcc k/k_{Bz}	sc k/k_{Bz}
0.5	0.649	0.694
1.0	0.569	0.633
1.5	0.542	0.610
2.0	0.533	0.599
2.5	0.524	0.588

In table II we show the values of the momentum cut-off necessary for obtaining the Néel temperature, as given by Rushbrooke and Woods. We conclude that approximately half of the phase space should be excluded. This procedure for determining k_m is phenomenological and no claim of an "ab initio" evaluation of that parameter is made.

We wish now to compare the magnetization and the parallel susceptibility obtained in the framework of the present approach and the experimental data available for MnF_2, which is one of the most representative Heisenberg antiferromagnets. This crystal is in fact the best experimentally known antiferromagnet insulator. It has a perovskite structure. The magnetic ions (Mn^{++}) form a bc tetrahedric lattice ($S = \frac{5}{2}$). The antiferromagnetic ions are not the nearest neighbours (these are in fact weakly ferromagnetic) but the *next* nearest neighbours.

The magnetic structure of MnF_2 shows a small amount of anisotropy, which is not taken into account in our theoretical description. It is known that $RbMnF_3$ is a better example of an isotropic Heisenberg antiferromagnet but, due to its isotropy, some magnetic measurements turn out to be much more difficult than in the case of MnF_2 (or even impossible).

Figure 1 allows us to compare the reduced magnetization $M(T)/M(0)$, obtained by the present method, with the experimental data of Jaccarino [6], obtained from NMR in MnF_2.

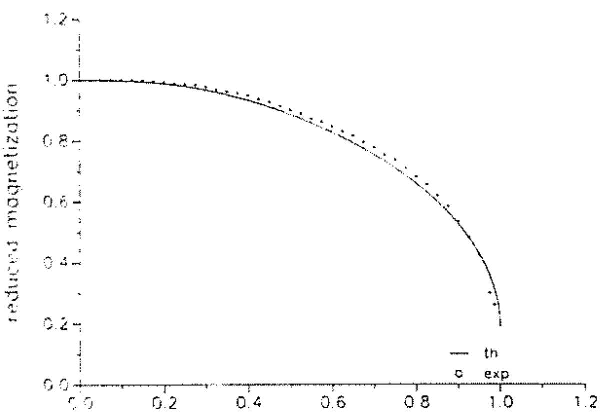

Fig. 1. Comparison of the sublattice magnetization obtained with the thermal boson method in the case of MnF_2 and obtained experimentally from NMR [6].

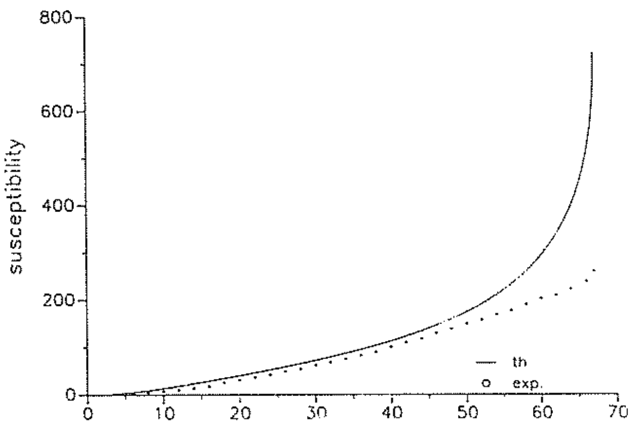

Fig. 2. Comparison of the parallel magnetic susceptibility (in units 10^{-6} per gram) obtained with the thermal boson method in the case of MnF_2 and obtained experimentally [7].

For the sake of consistency, we have taken $k_m = 0.485\, k_{Bz}$ instead of the value $k_m = 0.524\, k_{Bz}$, indicated on table II for $S = \frac{5}{2}$, since the Rushbrooke–Woods formula does not reproduce very well the empirical Néel temperature of MnF_2 ($k_B T_N/J = 18.7$, corresponding to $T_N = 67$ K, if $J = 2 \times 1.79 = 3.58$ K). The results have the same quality as in the ferromagnetic case. At low temperatures, the spin wave results are reproduced, while near the phase transition some deviation of the present theory from experiment is apparent. The thermal boson expansion was in fact expected to fail around the Néel point.

In fig. 2 the calculated parallel susceptibility $\chi_{\|}$ is displayed together with the experimental data for MnF_2 [7]. The calculated susceptibility reproduces the trend of the experimental points up to roughly $0.75\, T_N$. The discrepancy in the vicinity of the Néel temperature is striking. Although part of it should be attributed to the lack of anisotropy in the present description, certainly its major explanation should have to do with the failure of the spin wave picture at the critical point.

4. Conclusions

We conclude that the thermal boson results for Heisenberg antiferromagnets are able to describe well the data for both the magnetization and the magnetic parallel susceptibility, within a wide range of temperatures. In the immediate neighbourhood of the critical point, the thermal boson expansion, truncated at the harmonic term, is no longer expected to provide a good description of the real phenomena. In that region, fluctuations of all orders are relevant and any extrapolations of low-temperature results would fail. These conclusions are therefore in agreement with those obtained in the case of ferromagnetic insulators.

Let us make a few comments on the work of other authors on the problem of spin waves at finite temperature in antiferromagnets. In ref. [8], Bloch has discussed shortly the so-called "dynamical renormalization" (or magnon renormalization) of spin waves in bcc antiferromagnets. Though within that framework no restriction in momentum space seems necessary to make a guess of the Néel temperature within an error of 10%, we point out that in those kind of calculations the kinematical correlations, which are more important in antiferromagnets than in ferromagnets (at $T = 0$ and "a fortiori" in the critical region) are left out. The quality of the fit, if not accidental, remains therefore to be theoretically understood.

Along similar lines, Low [9] has calculated the spin wave spectrum at finite temperatures of MnF_2, concluding that the agreement with the experimental information obtained through neutron scattering [10] is fairly good till $T = 0.9\, T_N$, when the concept of well-defined spin waves ceases to make sense anymore.

Several other authors have addressed afterwards the problem of the dynamical interaction between antiferromagnetic magnons [11–13] trying different approaches.

In contrast to all those works, we have emphasized in the present article kinematical correlations as described by a variational approach to the linear free energy. The interplay of kinematical and dynamical interactions of collective excitations within the ordered phase of antiferromagnets deserves certainly further studies.

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