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Heuristic generalization of the Bogolyubov-Tyablikov magnetization equation to arbitrary isotropic spin Hamiltonians

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The Bogolyubov-Tyablikov (BT) magnetization equation for the Heisenberg Hamiltonian with an arbitrary elementary spin is shown to reduce to the mean-field result in the limit of infinite-range interactions. Comparison with the mean-field magnetization equation for an arbitrary isotropic spin Hamiltonian, ferromagnetic as well as ferrimagnetic, suggests a generalization of the BT magnetization equation and of the effective, temperature-dependent, magnon spectrum corresponding to any such Hamiltonian. The condition for a first-order phase transition in a ferromagnetic system is derived, showing that mean-field theory underestimates the magnitude of the quartic term required. The validity of the results conjectured is verified for a certain class of non-Heisenberg Hamiltonians.

The Heisenberg model, which has had an enormous impact on the development of the theory of magnetism, is insufficient to account for the vast variety of experimentally observed phenomena concerning the properties of magnetic materials.[1,2]

Typical extensions of the Heisenberg Hamiltonian involve the addition of terms which are of higher order than bilinear in the spin operators. A considerable amount of effort has been invested in the study of systems represented by spin Hamiltonians involving biquadratic terms such as $(s_i s_j)^2$ as well as three- and four-center terms such as $(s_i s_j)(s_j s_k)$ and $(s_i s_j)(s_k s_l)$. Such terms arise in accurate treatments of the many-electron problem containing direct exchange,[3] superexchange,[4] and spin-orbit coupling.[5,6] They also appear as a consequence of spin-phonon coupling, giving rise to magnetostriction. The thermodynamic properties of such Hamiltonians were studied in Refs. 7—11.

Complicated effective spin Hamiltonians are also applied in the description of nonmagnetic phenomena such as the orientational ordering in solid hydrogen, whose effective Hamiltonian is antiferromagnetic with a large biquadratic term.[12] Three- and four-particle exchange terms among nuclear spins are of crucial significance in the theory of the phase diagram of ^{3}He around 2 mK.[13,14] Spin Hamiltonians containing anisotropic and higher than bilinear terms were also investigated in the framework of a lattice model of quantum fluids.[15—17]

In spite of the very impressive progress in the understanding and accurate theoretical treatment of Heisenberg-type Hamiltonians, the methods developed for them cannot be transferred straightforwardly to more complex cases.

Several developments in mean-field theory have taken place recently. Some of the relevant contributions are presented in Refs. 18—21. The present authors have studied a formulation of mean-field theory which is applicable to spin Hamiltonians of arbitrary form. This formulation was applied to isotropic[22,23] and anisotropic[24] magnetic systems. It enabled the study of heat magnetization[25] magnetostriction,[26] the phase diagram[27] and dynamics[28] of magnetic systems with axial symmetry, and a new mechanism of ferromagnetism[29] which has also been discovered in a recent experimental study.[30]

In order to account for short-range correlations a treatment which is more accurate than mean-field theory is needed. One such approach, which is based on decoupling the hierarchy of two-time Green's functions, was formulated by Bogolyubov and Tyablikov[31,32] for the Heisenberg Hamiltonian with spin $\frac{1}{2}$. This approach was generalized to a Heisenberg Hamiltonian with arbitrary spin[33,34] as well as to the Heisenberg antiferromagnet.[35] A variational formulation enabled the extension to a system consisting of species with internal structure which interact via an effective anisotropic Heisenberg Hamiltonian, ferromagnetic as well as antiferromagnetic.[36—38] This method was successfully applied to a number of concrete systems.[39,40]

The BT model has several attractive features which suggest that it provides overall a rather satisfactory description of the properties of the spin system. In the limit of infinite-range interactions it reduces to the mean-field approximation.[41] At low temperatures the BT model reduces to the spin-wave treatment, which at that limit is exact.[42] For $T > T_c$ it coincides with the spherical model,[41] which in the limit of an infinite-dimensional spin becomes exact.[43,44] As an indication of the relation between the BT and spherical models, note that those critical exponents which were evaluated for the former agree with the latter, both above and below T_c.[34] A further significant advantage of the BT method is that it provides in a unified self-consistent way both the thermodynamic and the dynamic properties, which in most other approaches are treated separately.

The BT Green's-functions treatment of the Heisenberg Hamiltonian

$$\hat{H} = -\tfrac{1}{2}\sum J_{ij} s_i \cdot s_j \tag{1}$$

results, among other things, in a self-consistency equation for the magnetization

$$S = \sigma B_\sigma(\sigma \ln(1+\Phi^{-1})) \,, \tag{2}$$

where B_σ is Brillouin's function and

$$\Phi=(1/N)\sum_{\mathbf{q}}\{\exp[2\beta\omega(\mathbf{q},T)]-1\}^{-1} \quad (3)$$

with $\beta=1/k_BT$ and

$$\omega(\mathbf{q};T)=[J(\mathbf{0})-J(\mathbf{q})]S(T) . \quad (4)$$

Here S is the magnetization per site and $J(\mathbf{q})$ is the Fourier transform of J_{ij}.

The structure of these results can be interpreted by noting that $\omega(\mathbf{q};T)$ is the effective, temperature-dependent, magnon spectrum which differs from the mechanical ($T\rightarrow 0$) magnon spectrum

$$\omega(\mathbf{q})=[J(\mathbf{0})-J(\mathbf{q})]\sigma \quad (5)$$

by the replacement of the elementary spin σ with $S(T)$. Therefore, Φ is just the distribution function of the effective magnons, evaluated using Bose statistics. In order to proceed we note that in the limit of infinite-range interactions

$$J(\mathbf{q})=J(\mathbf{0})\delta(\mathbf{q}) , \quad (6)$$

so that, for an isotropic Heisenberg Hamiltonian,

$$\Phi=\{\exp[\beta J(\mathbf{0})S]-1\}^{-1} \quad (7)$$

and

$$S=\sigma B_\sigma(\beta\sigma J(\mathbf{0})S) . \quad (8)$$

Equation (8) is the standard mean-field magnetization equation for the Heisenberg Hamiltonian.

The generalization of Eq. (8) to an arbitrary isotropic infinite-range spin Hamiltonian $H(S)$ is[22]

$$S=\sigma B_\sigma(-\beta\sigma\,\partial H/\partial S) . \quad (9)$$

The similarity in form of Eqs. (8) and (9) reveals the fact that the appearance of the Brillouin function is a universal characteristic of the degeneracy of the spin states, independent of the form of the Hamiltonian.[29] The comparison of these two equations suggests that the effective field, which in the Heisenberg case is of the form $J(\mathbf{0})S$, can in general be written as $-\partial H/\partial S$. A second universal feature of the BT formalism as expressed by Eq. (2) and (3) is the form of the distribution function Φ, which reflects the bosonic nature of the magnons.

The above interpretation strongly suggests that a heuristic procedure for the generalization of the BT formalism can be based on a reversal of the logical sequence presented by Eqs. (1)–(8).

Let us start with the generalized magnetization equation [Eq. (9), instead of Eq. (8)], make the appropriate changes in Eqs. (7), (6), (5), (4), and (3), and assume that Eq. (2) remains unchanged.

The generalization of Eq. (7), i.e., the infinite-range limit of the Bose distribution function of the "generalized" effective magnons, should be of the form

$$\Phi=(e^{-\beta\partial H/\partial S}-1)^{-1} , \quad (10)$$

so as to obtain Eq. (9) by substituting this expression in Eq. (2). At this stage it seems almost inevitable that the generalization of Eq. (3) should be

$$\Phi=(1/N)\sum_{\mathbf{q}}\{\exp[\beta\Omega(\mathbf{q};\partial H/\partial S)]-1\}^{-1} . \quad (11)$$

Ω is the generalized effective magnon spectrum, depending on the temperature through its dependence on $\partial H/\partial S$, which is a function of the magnetization. Here, H is the infinite-range limit of the microscopic Hamiltonian $\hat{H}$.

It is clear that in the infinite-range limit Ω should be independent of $\mathbf{q}$, except for a singularity at $\mathbf{q}=0$, which, however, has no effect on Φ. Moreover, to agree with Eq. (10), the infinite-range limit of Ω should be

$$\Omega(\mathbf{q};\partial H/\partial S)\rightarrow -\partial H/\partial S . \quad (12)$$

We do not yet know how to derive Ω from first principles. What we do know is that the low-temperature limit of $\Omega(\mathbf{q};\partial H/\partial S)$ is the effective mechanical magnon spectrum corresponding to the general spin Hamiltonian under investigation, which can presumably be obtained from the equations of motion for the Fourier transform of the spin operators, making the appropriate generalization of the familiar linearization procedure for the Heisenberg-type spin waves.

As an additional heuristic argument supporting Eq. (12) we point out that the infinite-range form of the equations of motion for an arbitrary anisotropic spin Hamiltonian is[28]

$$\dot{s}_\pm=i[H,s_\pm]=iF_\pm(s^2,s_z)s_\pm , \quad (13)$$

where

$$F_\pm=N\{1-\exp[\pm(1/N)\partial/\partial s_z]\}H(s^2,s_z) , \quad (14)$$

N being the number of elementary spins. In the thermodynamic limit ($N\rightarrow\infty$) the equations of motion become

$$\dot{s}_\pm=\pm i(\partial H/\partial s_z)_s\cdot s_\pm . \quad (15)$$

If H depends on s_z only this result implies that the mechanical magnon frequency is $-(\partial H/\partial s_z)_{s_z=\sigma}$ and the effective temperature-dependent magnon frequency is $-\partial H/\partial S$ where S is the magnetization. This last result is actually valid for any $H(s^2,s_z)$, for all $\mathbf{q}\neq 0$. This is in harmony with Eq. (12).

At this stage we would like to rewrite Eq. (4) in the form

$$\omega(\mathbf{q};T)=\omega(\mathbf{q})\cdot[J(\mathbf{0})\cdot S] , \quad (4')$$

where

$$\omega(\mathbf{q})=[1-J(\mathbf{q})/J(\mathbf{0})] , \quad (16)$$

and offer the following interpretation of the two factors in it. $J(\mathbf{0})S$ is just the effective field, which, as we have already seen, should be replaced by $-\partial H/\partial S$ in the general case. On the other hand, $1-J(\mathbf{q})/J(\mathbf{0})$ is a kinematical factor which depends on the lattice structure and the assumed range of interaction. It therefore appears plausible that the generalized effective magnons will be of the form

$$\Omega=-\omega^*(\mathbf{q})\cdot\partial H/\partial S . \quad (17)$$

$\omega^*(\mathbf{q})$ is the reduced magnon frequency, which is independent of the temperature and the scale of energy, and which has to be determined for each case according to the lattice symmetry and the range of interaction assumed. Equation (17) should be viewed as a conjecture concerning the result of the linearization of the equations of motion for the generalized magnons.

Substitution of Eq. (17) in Eq. (11) and substitution of the latter in Eq. (2) provides the generalized BT magnetization equation for an arbitrary ferromagnetic isotropic spin Hamiltonian.

An application of the BT formalism to the nearest-neighbor antiferromagnetic Heisenberg Hamiltonian was proposed by Hewson and ter Haar.[35] Their equations for the two sublattice magnetizations can be written in the form

$$S_\pm=\sigma B_\sigma(\sigma \ln(1+\Phi_\pm^{-1}))\ , \tag{18}$$

where

$$\Phi_\pm=(1/2N)\sum_{\mathbf{q}}\{[(e^{\beta E_1}-1)^{-1}+(e^{\beta E_2}-1)^{-1}]\pm[2J(S_+-S_-)/(E_1-E_2)][(e^{\beta E_1}-1)^{-1}-(e^{\beta E_2}-1)^{-1}]\} \tag{19}$$

and

$$E_{1,2}=J\{\pm[(S_++S_-)^2-4S_+S_-\omega^2(\mathbf{q})]^{1/2}+(S_++S_-)\} \tag{20}$$

with

$$\omega^2(\mathbf{q})=1-\gamma^2(\mathbf{q}),\ \ \gamma(\mathbf{q})=\tfrac{1}{2}\sum_{\mathrm{NN}}\exp(i\mathbf{q}\cdot\boldsymbol{\delta})\ .$$

To generalize these results to an arbitrary two-sublattice isotropic spin Hamiltonian we make the following three observations:

(a) The mechanical magnon spectrum of a two-sublattice Heisenberg Hamiltonian with elementary spins σ_1 and σ_2 (Ref. 45) is essentially identical to Eq. (20), provided that S_+ and S_- are replaced by σ_1 and σ_2.

(b) The infinite-range limit of Eq. (18) is

$$S_\pm=\sigma B_\sigma(-\beta\sigma J S_\mp)\ . \tag{21}$$

(c) The infinite-range sublattice magnetization equations for an arbitrary isotropic two-sublattice spin Hamiltonian are[29]

$$S_i=\sigma_i B_{\sigma_i}(-\beta\sigma_i\,\partial H/\partial S_i)\ ,\ \ i=1,2\ . \tag{22}$$

Following the same heuristic approach which was applied in the ferromagnetic case we conjecture that the BT equations for an arbitrary isotropic two-sublattice spin Hamiltonian, whose infinte-range limit is $H(S_1,S_2)$, are

$$S_i=\sigma_i B_{\sigma_i}(\sigma_i \ln(1+\Phi_i^{-1}))\ ,\ \ i=1,2 \tag{23}$$

where

$$\Phi_i=(1/2N)\sum_{\mathbf{q}}\{[(e^{\beta E_1}-1)^{-1}+(e^{\beta E_2}-1)^{-1}]+(-1)^i 2(h_1-h_2)[(e^{\beta E_1}-1)^{-1}-(e^{\beta E_2}-1)^{-1}]/(E_1-E_2)\} \tag{24}$$

and

$$E_i=(-1)^i\{(h_1+h_2)^2-4h_1h_2[\omega^*(\mathbf{q})]^2\}^{1/2}+(h_1+h_2) \tag{25}$$

with

$$h_i=\partial H/\partial S_i,\ \ i=1,2\ .$$

$\omega^*(\mathbf{q})$ is the temperature-independent structural factor.

A simply manageable example, in which the generalization of the BT formalism can be carried out so as to support the above conjectures, can be introduced in terms of the Hamiltonian

$$\mathscr{H}=\sum_n a_n H^n\ ,$$

where H is the isotropic Heisenberg Hamiltonian and a_n are arbitrary coefficients. Noting that

$$i\dot{s}_i^{(\pm)}=[s_i^{(\pm)},\mathscr{H}]=\sum_n a_n\sum_{m=0}^{n-1}H^m[s_i^{(\pm)},H]H^{n-m-1} \tag{26}$$

and approximating H outside of the commutator brackets by its infinite-range form $\overline{H}$ we obtain

$$i\dot{s}_i^{(\pm)}=[\partial\mathscr{H}(\overline{H})/\partial\overline{H}]\cdot[s_i^{(\pm)},H]\ . \tag{27}$$

The Fourier transform of Eq. (27) is

$$i\dot{s}_q^{(\pm)}=[\partial\mathscr{H}(\overline{H})/\partial\overline{H}]\cdot[s^{(\pm)},H]_q\ . \tag{28}$$

Treating the commutator in the usual way we obtain

$$i\dot{s}_q^{(\pm)}=(\pm\partial\mathscr{H}/\partial\overline{H})\cdot S\cdot\omega(\mathbf{q})\cdot s_q^{(\pm)}\ , \tag{29}$$

where $\omega(\mathbf{q})$ is given by Eq. (16). Noting that $\overline{H}=-J(0)S^2/2$ we finally obtain

$$i\dot{s}_q^{(\pm)}=\pm[\partial\mathscr{H}(S)/\partial S]\cdot\omega(q)\cdot s_q^{(\pm)} \tag{30}$$

in agreement with Eq. (17).

Let us return to the general case of an arbitrary ferromagnetic spin Hamiltonian. If the phase transition is of second order the behavior near the critical temperature is determined by the leading quadratic term in S, so that both the critical temperature and the critical exponents are identical to those obtained for the Heisenberg Hamiltonian.[34] However, the phase transition for a general spin Hamiltonian can also be of first order. To obtain the condition for a first-order phase transition we determine the slope of the magnetization curve evaluated from Eq. (2) with Φ given by Eqs. (11) and (17), at the temperature T^* at which $S\to 0$.

Using the fact that in the vicinity of T^*, S as well as $\partial H/\partial S$ are small, we obtain

$$\Phi^{-1}\simeq -\beta\,\partial H/\partial S\cdot F^{-1}(-1)\{1-[1/2F(-1)]\cdot\beta\,\partial H/\partial S+[3-F(-1)F(1)]/[12F(-1)^2](\beta\,\partial H/\partial S)^2+\cdots\}\ ,$$

where

$$F(n)=(1/N)\sum_{\mathbf{q}}[\omega^*(\mathbf{q})]^n\ . \tag{31}$$

The magnetization equation becomes

$$S=\sigma B_\sigma(\beta\sigma\{[-(\partial H/\partial S)/F(-1)+\beta^2(\partial H/\partial S)^3[F(-1)F(1)-1]/[12F(-1)^3]\})\ . \tag{32}$$

Only the two leading terms in

$$H=-(J/2)S^2-(\alpha/4)S^4+\dots$$

are relevant. By comparison with a similar analysis in Ref. 22 it follows that the condition for a first-order transition is

$$\alpha>\{3J/4[\sigma(\sigma+1)]^2\}\{[(2\sigma+1)^2+1]/5+F(1)-F^{-1}(-1)\}\ . \tag{33}$$

In the infinite-range limit $F(1)=F(-1)=1$ and the result reduces to that of Ref. 22. Note that $F(1)$ is the arithmetic mean of the reduced magnon frequencies and $F^{-1}(-1)$ is their harmonic mean. By a well-known theorem[46] it follows that $F(1)>F^{-1}(-1)$, which means that mean-field theory underestimates the magnitude of the coefficient in the quartic term required for a first-order phase transition. Note that this result is independent of the specific form of the reduced magnon spectrum $\omega^*(\mathbf{q})$, which means that it holds for any lattice type and any range of interaction.

We plan to extend the heuristic approach discussed in the present contribution to anisotropic spin Hamiltonians as well as to systems with internal structure. The appropriate infinite-range results have recently been derived.[24,27,47,48] It is of course desirable to replace the heuristic arguments presented by a more satisfactory derivation, either along the lines of the original Green's-functions approach[34] or generalizing the method used by Suzuki.[37]

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