Antiferromagnet of arbitrary spin in a strong magnetic field

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An approach developed previously for spin 1/2 is generalized to an arbitrary spin. An antiferromagnet is considered in a magnetic field H close to the critical strength H_c at which the sublattice structure disappears at absolute zero. In this region the antiferromagnet may be thought of as a Bose gas, and the antiferromagnetic ordering would correspond to a state with a Bose condensate. The ground-state energy of the system is derived and compared with the energy found in the self-consistent field approximation. The temperature of the antiferromagnetic transition is described as a function of the magnetic field by $T_c \sim (H_c - H)^{2/3}$.

1. INTRODUCTION

In an antiferromagnet in a strong magnetic field, a so-called collapse of sublattices occurs; i.e., the sublattice structure disappears (all the spins become parallel at absolute zero).

A new approach in the theory of antiferromagnets was proposed in Ref. 1 on the basis of an analogy with a Bose gas. This analogy holds near the sublattice collapse point at a sufficiently low temperature. The small parameter of the problem (along with the temperature) is the ratio

$$(H_c - H)/H_c \ll 1, \tag{1}$$

where H is the magnetic field, and H_c is the critical field (the point of this transition at absolute zero). An antiferromagnetic state occurs at $H < h_c$, and a state with a Bose condensate corresponds to antiferromagnetic order in this approach.

This approach is quite different from the approximation of a self-consistent field (the approximation of classical spins), which serves well in the large-spin limit; away from this limit, its validity is dubious. Indeed, a comparison of the results[1] for spin 1/2 shows that the self-consistent field approximation breaks down near H_c.

The spin-1/2 case was studied in Ref. 1. A question which naturally arises is whether this approach can be generalized to the case of an arbitrary spin (the Hamiltonian used in Ref. 1 cannot be generalized directly). We attempt to resolve this question in the present paper.

Our primary task is to construct a Bose Hamiltonian which is equivalent to the original spin Hamiltonian near H_c (Section 2). The problem can then be solved by analogy with the problem of a slightly nonideal Bose gas[2,3] (Section 3). This part of the study has much in common with the spin-1/2 case,[1] so that the corresponding parts of this study will be either summarized or omitted altogether.

2. EQUIVALENT BOSE HAMILTONIAN

We begin with the Heisenberg spin Hamiltonian

$$\mathscr{H} = \frac{1}{2}\sum_{n\neq n'} I_{nn'}\mathbf{S}_n\mathbf{S}_{n'} + H\sum_n S_{nz}, \tag{2}$$

where $\mathbf{S}_n$ is the spin operator of site n, and H is the magnetic field expressed in energy units.

We first discuss the concept of a transition to a Bose Hamiltonian, which is equivalent to (2) near H_c. In a field $H > H_c$ all the spins are parallel (at zero temperature) and have a maximum (in modulus) z projection $-S$. In a field $H < H_c$, if H is quite close to H_c, there are a few spins with other projections: $\sigma - S$, $\sigma = 1, 2, \ldots, 2S$. The point which is important for our purposes is that all these excitations (of the initial state with parallel spins) can be treated in a common way by introducing Bose particles. Specifically, with the projection $1 - S$ we associate a single Bose particle at the given site; with the $2 - S$ projection we associate two Bose particles; etc.—precisely as in Holstein-Primakoff transformations. We then make use of the fact that there are only a few such particles, since H is close to H_c. In this case we are dealing with a gas, so that we can ignore three-body collisions, including the possibility that three or more particles will meet at a given site. It is thus sufficient to consider the $1 - S$ and $2 - S$ (in addition to the $-S$) projections at each site. The corresponding Bose Hamiltonian can then be written on the basis of the condition that all the transitions which are incorporated in the problem (all those which are important) are described by this Hamiltonian in precisely the same ways as they are described by the original Hamiltonian, (2).

We turn now to the construction of the equivalent Bose Hamiltonian. We first rewrite (2) in the more convenient form

$$\mathscr{H} = \frac{1}{2}\sum_{n\neq n'} I_{nn'}\{S_{nz}S_{n'z} + S_n^+S_{n'}^-\} + H\sum_n S_{nz},$$

$$S_n^\pm = S_{nx} \pm iS_{ny}. \tag{3}$$

We recall which matrix elements of the spin operators are nonzero:

$$(S^+)_{M,M-1} = (S^-)_{M-1,M} = \{(S+M)(S-M+1)\}^{1/2},$$
$$(S_z)_{M,M} = M, \tag{4}$$

where M is the z projection of the spin (and the index of the corresponding wave function). For brevity here we are omitting the site index n.

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According to this discussion, the expression for S_z in terms of the Bose operators will be the same as in terms of Holstein-Primakoff transformations. As for S^+ and S^-, we note that the corresponding expressions are written on the basis of the condition that the matrix elements in the class of functions 0, 1, 2 (in the particle occupation numbers), i.e., in the class of functions $-S$, $1-S$, $2-S$ (in spin projections), agree with (4). It is not difficult to see that this condition is satisfied by the following relations:

$$\begin{gathered}S_z=-S+\beta^+\beta,\\ S^+\approx(2S)^{1/2}\beta^+[1+(K-1)\beta^+\beta],\\ S^-\approx(2S)^{1/2}[1+(K-1)\beta^+\beta]\beta,\\ K\equiv(1-1/2S)^{1/2},\end{gathered}\tag{5}$$

where $\beta(\beta^+)$ is the annihilation (creation) operator for a Bose particle (each site has its own operator; the site index is omitted). The symbol $\approx$ means that the correspondence holds only for occupation numbers 0, 1, 2. Here we have introduced a special notation for an expression which we will be using frequently below.

Relations (5) were derived without reference to the Holstein-Primakoff transformations, but they could also be derived directly from these transformations. Briefly, here is the procedure: we consider the Holstein-Primakoff expression for S^+, for example:

$$S^+=(2S)^{1/2}\beta^+(1-\beta^+\beta/2S)^{1/2}.$$

Here the radical is understood as a power series in $\beta^+\beta/2S$. We write this series, and using the commutation relations we put the operators β^+ and β in each of the terms of the series in the normal order (with creation operators to the left of annihilation operators). Collecting the terms of first and third order in the Bose operators in the resulting expression for S^+, we find relation (5).

We note that (5) holds for an arbitrary spin, including spin 1/2. In this case, the infinite repulsion at a site[1] can be taken into account in a different way, by means of the replacement $\beta^+\to\beta^+(1-\beta^+\beta)$ (a similar approach has been used for the Hubbard model with strong correlation[4]); we then have a complete correspondence with (5).

We now substitute (5) into Hamiltonian (3) and find a Hamiltonian which contains terms with even powers of the Bose operators up to the sixth. The latter terms (of sixth degree) describe three-body collisions, which are unimportant in a gas (furthermore, to take them into account would go beyond the accuracy of our treatment), so we must discard them. Finally, we find the following Bose Hamiltonian:

$$\begin{gathered}\mathcal{H}=\mathcal{E}_0(H)+\mathcal{H}_0+\mathcal{H}_1,\\ \mathcal{E}_0(H)=\frac{1}{2}S^2\sum_{n\neq n'}I_{nn'}-SHN,\\ \mathcal{H}_0=S\sum_{n\neq n'}I_{nn'}\beta_n{}^+\beta_{n'}+\sum_n\Big(H-S\sum_{n'}I_{nn'}\Big)\beta_n{}^+\beta_n,\\ \mathcal{H}_1=\frac{1}{2}\sum_{n\neq n'}I_{nn'}\{\beta_n{}^+\beta_{n'}{}^+\beta_{n'}\beta_n\\ +2S(K-1)[\beta_n{}^+\beta_n{}^+\beta_n\beta_{n'}+\text{H.a.}]\},\end{gathered}\tag{6}$$

where N is the number of sites in the lattice.

Transforming to quasimomenta in (6) by means of the standard formulas

$$\beta_n=\frac{1}{N^{1/2}}\sum_{\mathbf{p}}\beta_{\mathbf{p}}\exp(i\mathbf{p}\mathbf{R}_n)$$

($\mathbf{R}_n$ is the radius vector of site n), and omitting the constant $\mathcal{E}_0(H)$, which is not important for the discussion below, we find

$$\mathcal{H}\to\sum_{\mathbf{p}}\omega(\mathbf{p})\beta_{\mathbf{p}}{}^+\beta_{\mathbf{p}}+\frac{1}{2SN}\sum_{\mathbf{p}_i}\beta_{\mathbf{p}_1}{}^+\beta_{\mathbf{p}_2}{}^+\beta_{\mathbf{p}_3}\beta_{\mathbf{p}_4}\times\{\varepsilon(\mathbf{p}_1-\mathbf{p}_4)+2S(K-1)[\varepsilon(\mathbf{p}_1)+\varepsilon(\mathbf{p}_4)]\}.\tag{7}$$

In the second sum here, the ordinary quasimomentum conservation law is understood to hold, and we are using the notation

$$\begin{gathered}\varepsilon(\mathbf{p})=S\sum_{n'}I_{nn'}\exp[i\mathbf{p}(\mathbf{R}_{n'}-\mathbf{R}_n)],\\ \omega(\mathbf{p})=[\varepsilon(\mathbf{p})-\varepsilon_0]+(H-H_c),\\ \varepsilon_0=\min\varepsilon(\mathbf{p}),\qquad H_c=S\sum_{n'}I_{nn'}-\varepsilon_0.\end{gathered}\tag{8}$$

Here $\omega(\mathbf{p})$ is the energy of an isolated magnon in a state with parallel spins. If $H>H_c$, its energy is positive, and the state with parallel spins is stable. If $H<H_c$, the energy is negative near the minimum, the state with parallel spins becomes unstable, and magnons accumulate in one of the states (a Bose condensate forms). In this case the number of particles for a given parameter (1) is determined by the interaction [the second term in (7)].

Our basic goal has thus been reached. We have derived a Bose Hamiltonian, (6), (7), which is equivalent to the original spin Hamiltonian, (2), near H_c. This equivalence follows from the method by which the Bose Hamiltonian is constructed. It can also be checked directly by comparing the matrix elements of operators (6) and (3) between the states mentioned in connection with the derivation of (5).

To conclude this section we note the following. The Bose Hamiltonian written in Ref. 1 is equivalent to the original Heisenberg Hamiltonian for spin 1/2 for an arbitrary field H. However, that Hamiltonian can be used (i.e., it solves the problem) only near H_c, where the gas approximation can be used. In the present paper we have derived a Hamiltonian which is equivalent to the initial spin Hamiltonian for an arbitrary spin. Although this is true only near H_c, we require nothing more, because the problem is solved only near H_c.

RESULTS

In this section we use the equivalent Hamiltonian (7) to calculate the ground-state energy, and we compare it with the energy derived in the self-consistent field approximation. We consider the simplest case of a single magnon minimum [a single minimum of the function $\varepsilon(\mathbf{p})$]. This minimum necessarily lies at the boundary of the Brillouin zone, at equivalent points which differ from each other by a reciprocallattice vector. It is thus sufficient to introduce the coordinate $\mathbf{p}_0$ of one of these points:

$$\varepsilon(\mathbf{p}_0)=\varepsilon_0=\min\varepsilon(\mathbf{p}).$$

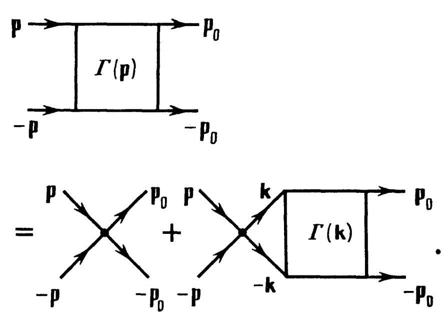
For the specific calculations we use a simple cubic lattice and a body-centered cubic lattice with a nearest-neighbor interaction. For the simple cubic lattice, for example, we have

$$\varepsilon(\mathbf{p})=2SI(\cos p_x+\cos p_y+\cos p_z),\quad \mathbf{p}_0=\pi(1,1,1),\quad m=1/2SI, \tag{9}$$

where $I>0$ is the interaction of nearest neighbors, whose separation is assigned a unit value. We have also given here the effective mass m at the minimum.

The ground state of the system described by Hamiltonian (7) is a state with a Bose-Einstein condensate ($H<H_c$). The interaction between particles is not weak (it is weak only if $S\gg1$), so that we must first find the complete amplitude for the scattering of two free particles (ignoring the effects of other particles) at a minimum of the zone. We can then use the standard expressions for the energy and other characteristics of the system.[2,3]

The vertex part Γ, which describes the scattering of two particles, can be found by a diagram technique. In accordance with the discussion above, we calaculate it in the ladder approximation, using zeroth-order (interaction-ignoring) one-particle Green's functions (in this case there is nothing other than the ladder); i.e., we find the following diagram equation for it:



(10)

The sum frequency in $\Gamma(\mathbf{p})$ is chosen in accordance with the discussion above, i.e., it is set equal to $2\omega(\mathbf{p}_0)$.

After an elementary integration over the intermediate frequency, we find that Eq. (10) becomes

$$2S\Gamma(\mathbf{p})=\varepsilon(\mathbf{p}-\mathbf{p}_0)+2S(K-1)[\varepsilon(\mathbf{p})+\varepsilon_0] -\frac{1}{N}\sum_{\mathbf{k}}\frac{\varepsilon(\mathbf{p}-\mathbf{k})+2S(K-1)[\varepsilon(\mathbf{p})+\varepsilon(\mathbf{k})]}{\varepsilon(\mathbf{k})-\varepsilon_0}\Gamma(\mathbf{k}). \tag{11}$$

This equation can be simplified slightly by making use of the property

$$\langle\varepsilon\rangle\equiv\frac{1}{N}\sum_{\mathbf{p}}\varepsilon(\mathbf{p})=0. \tag{12}$$

Summing (11) over $\mathbf{p}$, we find the relation

$$\frac{K}{K-1}\langle\Gamma\rangle=\varepsilon_0\left\{1-\left\langle\frac{\Gamma}{\varepsilon-\varepsilon_0}\right\rangle\right\}. \tag{13}$$

Using this relation, we can replace (11) by

$$2S\Gamma(\mathbf{p})=\varepsilon(\mathbf{p}-\mathbf{p}_0)+2S\langle\Gamma\rangle\left[1+K\frac{\varepsilon(\mathbf{p})}{\varepsilon_0}\right] -\frac{1}{N}\sum_{\mathbf{k}}\frac{\varepsilon(\mathbf{p}-\mathbf{k})}{\varepsilon(\mathbf{k})-\varepsilon_0}\Gamma(\mathbf{k}). \tag{14}$$

It is thus necessary to solve Eq. (14) under condition (13).

It is not difficult to see that in the spin-1/2 case we have for the vertex part Γ the same equations as in any other method of describing the interaction used in Ref. 1 [see Eqs. (15) and (16) of Ref. 1].

For simple cubic and bcc lattices we can seek a solution in the form

$$\Gamma(p)-\langle\Gamma\rangle\sim\varepsilon(\mathbf{p}).$$

Here is the expression found for the unknown quantity $\Gamma(\mathbf{p}_0)$, which is proportional to the amplitude for the scattering of two particles at a zone minimum:

$$\Gamma(\mathbf{p}_0)\equiv\lambda/2=\nu I/(\nu I\tau+1-1/S). \tag{15}$$

Here we have used $\varepsilon_0=-S\nu I$, where ν is the number of nearest neighbors. The constant τ is defined by

$$\tau=\langle(\varepsilon-\varepsilon_0)^{-1}\rangle \tag{16}$$

(for a simple cubic lattice we would have $\tau\approx1/4SI$). In (15) we have introduced a special notation for the quantity $\Gamma(\mathbf{p}_0)$ in which we are interested.

Once we have found λ, we can write (for example) an expression for the energy of the system, $\mathscr{E}$:

$$\frac{\mathscr{E}}{N}={}^1/_2\lambda\rho^2-\mu\rho,\qquad \mu=H_c-H, \tag{17}$$

where ρ is the number of particles per site ($\rho\ll1$; this is a small parameter of the problem), and $-\mu$ is the energy of an isolated particle at a zone minimum [see (8)]. The condition for a minimum of (17) gives us

$$\rho=\mu/\lambda,\quad \mathscr{E}/N=-\mu^2/2\lambda. \tag{18}$$

Using $\mathscr{E}$, we can find the increment in the saturation magnetic moment and the magnetic susceptibility.

Our next task is to compare (18) with the expression found in the self-consistent field approximation (the classical-spin approximation). In this approximation, for the corresponding quantity $\mathscr{E}'$ [i.e., for the total energy minus $\mathscr{E}_0(H)$; see (6)] we easily find

$$\mathscr{E}'/N=-\mu^2/2\lambda',\quad \lambda'=2\nu I. \tag{19}$$

It can be seen that we have $\lambda<\lambda'$. In the case of a simple cubic lattice, for example, we have

$$\lambda\approx2\nu I/(1+1/2S)\quad(\nu=6)$$

[see the comment after (16)]. This result means that the energy of the system is lower, and the magnetic susceptibility larger, than predicted by the self-consistent field approximation.

It is not difficult to see that these results yield known limiting cases, primarily the spin-1/2 case, mentioned earlier; we derive the expression for the limit $S\to\infty$ below. In the latter case the interaction of the Bose particles is weak, so we can use the Born approximation to calculate Γ; i.e., we can make direct use of expression (7) in the calculation of $\mathscr{E}$, retaining in it only the condensate operators $\beta_{\mathbf{p}_0}\beta^+_{\mathbf{p}_0}$, and replacing them (as usual in the case of a weak interaction) by the term $(N\rho)^{1/2}$. We then find (19).

We can also write expressions for the excitation spectrum at absolute zero, $E(\mathbf{k})$, and for the temperature T_c, of the antiferromagnetic transition which are found in the standard way and are of the same form as the corresponding expressions for the spin-1/2 case.[1] Near the minimum the spectrum is

$$E(\mathbf{k})=\{(k^2/2m+\mu)^2-\mu^2\}^{1/2},$$

and far from the minimum it agrees with $\epsilon-\epsilon_0$. The quantity $\mathbf{k}$ is a quasimomentum measured from the value at the minimum of $\mathbf{p}_0$; $\mathbf{k}$ has the meaning of the quasimomentum of an excitation. The temperature of the antiferromagnetic transition is

$$T_c\approx 2,087\frac{1}{m}\left(\frac{H_c-H}{\lambda}\right)^{2/3}.$$

This dependence of T_c on (H_c-H) continues to hold in the arbitrary case of several equivalent valleys in the "seed" magnon spectrum $\omega(\mathbf{p})$. The dependence on the spin in these expressions enters through the interaction λ and the effective mass m.

It is interesting to compare the expression for T_c with the transition temperature T_c' found in the self-consistent field approximation:

$$T_c'\approx\frac{H_c}{2}\left\{\ln\frac{2\nu I}{H_c-H}\right\}^{-1}.$$

The reason for this dependence is the activation nature of the spin excitations in this approximation; for this reason, there is a marked difference from T_c.

We conclude with a few words about the fluctuation region. This region is clearly small for a slightly nonideal Bose gas, since it is zero for an ideal Bose gas. Accordingly, in the (T, H) plane the corresponding region will be a very narrow strip near the transition curve, for which an expression is given above. Furthermore, there is no such region at all at $T=0$ if we are considering a transition in the field. At $H>H_c$ we know the exact ground state—it is a vacuum in terms of magnons—while at $H<H_c$ there are magnons, but the gas approximation, which we have used here, becomes progressively better as H approaches H_c. We thus expect nothing more than insignificant corrections to the expression given here for T_c.

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