Heisenberg model with higher-order exchange: Ground-state properties and excitations

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Properties of Heisenberg models with higher-order exchange, $H=\frac{1}{2}\sum_{l=1}^{l_0}\sum_{n,m}J_{nm}^{(l)}(\vec{S}_n\cdot\vec{S}_m)^l$, are discussed. It is found that there is a one-to-one relation between the set of all coupling constants and the set of the energies of all ν-magnon states with $\nu\leq 2s$, where s is the spin at each site. Necessary and sufficient conditions for a ferromagnetic ground state are derived for systems with only nearest-neighbor interactions as well as for systems which also include next-nearest-neighbor interactions. These conditions are qualitatively different in the quantum and in the classical case. Discussing the elementary excitations, a classification of the ν-magnon bound states is found: an extreme case is the pure exchange bound state, where the ν spin deviations extend around the nearest ν sites; the opposite extreme case is the pure single-ion bound state where the ν spin deviations are most likely at the same site. Between these two cases there exist all combinations of both, if higher-order exchange with $l_0\geq\nu$ is present. The case $\nu=2$ is discussed in more detail.

I. INTRODUCTION

In this paper we investigate the properties at $T=0$ K of the Hamiltonian for the most general isotropic exchange model restricted to pair interactions,

$$H=\frac{1}{2}\sum_{l=1}^{l_0}\sum_{n,m}J_{nm}^{(l)}(\vec{S}_n\cdot\vec{S}_m)^l,\quad 1\leq l_0\leq 2s\ , \tag{1}$$

where $J_{nm}^{(l)}$ are the lth-order exchange coupling constants between sites n and m and $\vec{S}_n$ is the spin operator at site n with spin quantum number s.

Harris and Owen[1] and Rodbell *et al.*[2] were the first to show that (small) biquadratic exchange interactions must be taken into account. Later it was shown that higher-degree pair interactions between magnetic ions may be comparable or even stronger than the dipolar couplings. (For reference see Refs. 11, 17, and references therein.) This led to a lot of papers investigating the thermodynamic behavior of models (1).

Most of the theoretical work[3–16] is concerned with the influence of biquadratic exchange ($l=2$) on properties of the usual Heisenberg model [bilinear exchange ($l_0=1$)] with nearest-neighbor (NN) interactions. Let us review some of these concerning the ground-state properties and excitations of (1).

Munro[3] has proved that the ground state is a singlet if the couplings $J_{nm}^{(l)}$ fulfill certain conditions, thus extending the Lieb-Mattis theorem[4] up to biquadratic exchange. Conditions (necessary and sufficient) for a ferromagnetic ground state (FGS) were derived in Refs. 5 and 7–9 for NN interactions. An interesting numerical result for a chain with eight lattice sites was found by Parkinson.[10] He has shown that for all $J^{(1)}$ and $J^{(2)}$ with $0<J^{(1)}<1.3J^{(2)}$ the ground state is neither ferro- nor antiferromagnetic.

Concerning the elementary excitations, Nauciel-Bloch *et al.*[5] were the first to derive the spin-wave dispersion (1-magnon energy):

$$E_1(\vec{k})=E_F-s[J^{(1)}+2J^{(2)}s(s-1)]\times\sum_{\vec{\delta}_1}[1-\cos(\vec{k}\cdot\vec{\delta}_1)]\ , \tag{2}$$

where $\vec{\delta}_1$ are the NN vectors and E_F is the ferromagnetic energy. The influence of the biquadratic exchange on the 2-magnon states and especially on the bound states was investigated by several authors[6–8,11–14] for $s=1,2$. The most important result is the appearance of a single-ion bound state where the two spin deviations prefer to be located at the same site.

To our knowledge the Schrödinger model[15]

$$H=\frac{1}{4}\sum_{n,m}J_{nm}P_{nm} \tag{3}$$

with P_{nm} the transposition operator, a special case of model (1), is the only case with $l_0>2$ which was studied theoretically. For $s=1$, Uimin[16] has calculated the ground-state energy of model (3) for the linear chain.

The intention of this paper is to study for $T=0$ K the influence of higher exchange interactions on physical interesting quantities like ground-state properties, excitation energies, bound-state behavior, etc. This is important for the interpretation of experimental information as will be shown. In addition, higher-order exchange gives rise to stronger quantum effects.[17–19] This will be seen for the conditions for FGS.

The paper is organized as follows.

(i) First we derive general properties of the spectrum of model (1) for arbitrary l_0. In this context we discuss how

the original coupling constants $J_{nm}^{(l)}$ can be determined from experiments which only allow one to determine the ν-magnon energies, i.e., some renormalized coupling constants. In particular it is shown that from the ν-magnon energies with $\nu<2s$ it is impossible to decide if higher than νth exchange is present.

(ii) In the second section ground-state properties are studied. Here we improve the necessary and sufficient conditions for FGS obtained by other authors[5,7–9] for model (1) with $l_0=2$. Moreover, the influence of higher-order exchange ($l_0>2$), next-nearest-neighbor (NNN) interactions, and an external magnetic field are discussed.

(iii) The last section contains a study of a part of the excitation spectrum. It is shown that the 1-magnon dispersion for $l_0>1$ is equal to that of the bilinear model with a renormalized coupling constant. With specialization to Schrödinger's exchange model a classification of the ν-magnon bound states, which appear for models (1), is found.

These three sections are almost self-contained and not necessarily dependent on each other as far as the results are concerned.

II. GENERAL (SPECTRAL) PROPERTIES

The following applies to model (1) with arbitrary l_0. If not specified, the lattice and the coupling constants $J_{nm}^{(l)}$ are arbitrary.

Let $|m_1,\ldots,m_N\rangle$, $m_i\in\{-s,-s+1,\ldots,s\}$ be the eigenstates of all S_n^z, $n=1,\ldots,N$, where N is the number of lattice sites. Using the eigenstates of $\vec{S}_i\cdot\vec{S}_j$, we obtain

$$H|m_1,\ldots,m_N\rangle=E_F|m_1,\ldots,m_N\rangle - \tfrac{1}{2}\sum_{i,j}\sum_{m_i',m_j'}\left[\sum_{k=1}^{2s-|m_i+m_j|}\langle s,m_i';s,m_j'|2s-k,m_i+m_j\rangle\langle 2s-k,m_i+m_j|s,m_i;s,m_j\rangle B_{ij}^{(k)}\right] \times|m_1,\ldots,m_i',\ldots,m_j',\ldots,m_N\rangle\,, \tag{4a}$$

where $\langle s,m_i;s,m_j|S,m_i+m_j\rangle$ are Clebsch-Gordan coefficients,

$$E_F=\tfrac{1}{2}\sum_{i,j}\sum_{l=1}^{l_0}J_{ij}^{(l)}s^{2l} \tag{4b}$$

is the ferromagnetic energy, and

$$B_{ij}^{(k)}=\sum_{l=1}^{l_0}J_{ij}^{(l)}\left[s^{2l}-\left[s^2-2sk+\frac{k(k-1)}{2}\right]^l\right] \tag{4c}$$

are the renormalized coupling constants. This notation is chosen because all excitation energies are directly related to $B_{ij}^{(k)}$ and not to $J_{ij}^{(l)}$ as follows from (4a).

The significance of the $B_{ij}^{(k)}$ is revealed by the eigenvalues $E^{(2)}$ of the two-spin system:

$$H_2=\sum_{l=1}^{l_0}J_{ij}^{(l)}(\vec{S}_i\vec{S}_j)^l\quad i\neq j\text{ fixed}\,, \tag{5a}$$

$$E^{(2)}(S)-E_F^{(2)}=\sum_{l=1}^{l_0}J_{ij}^{(l)}\{[\tfrac{1}{2}S(S+1)-s(s+1)]^l-s^{2l}\} =-B_{ij}^{(2s-S)}\,, \tag{5b}$$

with S the total spin.

Equations (4) can be used to prove the following.

Statement. Let H and H' be given by (1) for two different sets $\{J_{ij}^{(l)}\}$ and $\{J_{ij}'^{(l)}\}$. Then all eigenvalues and eigenstates of H and H' in the ν-magnon space are equal if and only if

(i) $E_F(\{J_{ij}^{(l)}\})=E_F(\{J_{ij}'^{(l)}\})$,

(ii) $B_{ij}^{(k)}=B_{ij}'^{(k)}$ for all $i,j,k=1,\ldots,\nu$ $(\nu\leq 2s)$.

Proof. Assuming (i) and (ii) it is obvious using (4a) that the eigenvalues and eigenstates for $Ns-\nu\leq M\leq Ns$ are equal. Here M is the quantum number of S^z. Since the matrix built by the product of the Clebsch-Gordan coefficients is regular, the reverse statement is also true. Under more restrictive conditions:

(i') $E_F=E_F'$,

(ii') $B_{ij}^{(2k)}=B_{ij}'^{(2k)}=0$ for all i,j and $2k\leq\nu$,

$B_{ij}^{(2k+1)}=B_{ij}'^{(2k+1)}=B_{ij}^{(1)}$ for all i,j and $2k+1\leq\nu$,

the statement even stays true for different spins $s\neq s'$. With (i') and (ii') it follows that the Hamiltonian restricted to the ν-magnon subspace $\mathscr{H}_\nu$ has the form

$$H|_{\mathscr{H}_\nu}=H'|_{\mathscr{H}_\nu}=E_F-\tfrac{1}{4}\sum_{i,j}B_{ij}^{(1)}+\tfrac{1}{4}\sum_{i,j}B_{ij}^{(1)}P_{ij}\Big|_{\mathscr{H}_\nu}\,. \tag{6}$$

For the proof see the Appendix.

Given $B_{ij}^{(k)}$ for $k=1,2,\ldots,2s$ it can be shown that Eq. (4c) has a unique solution $J_{ij}^{(l)}$, $l=1,2,\ldots,2s$. Using this and the statement above, we find the following.

Conclusion. In order to determine all the model parameters $J_{ij}^{(l)}$ from experiment it is necessary and sufficient to measure all magnon energies up to $\nu=2s$.

Extension of the Munro theorem. As mentioned in the Introduction, Munro[3] proved that for systems (1) with $l_0=2$ on a lattice Λ consisting of two sublattices Λ_A and Λ_B, the ground state is a singlet ($S=0$) if the couplings fulfill some conditions.[3] This statement can easily be extended to systems which include higher exchange interactions: Performing the unitary transformation $U=\exp(-i\prod\sum_{i\in\Lambda_A}S_i^z)$, the nondiagonal elements of $H'=U^{-1}HU$ are not positive in the basis $\{|m_1,\ldots,m_N\rangle\}$ if

$$\langle m_i',m_j'|H_{ij}'|m_i,m_j\rangle\leq 0\ ,$$

for all i,j and $(m_i',m_j')\neq(m_i,m_j)$.

Here $H_{ij}'=U^{-1}H_{ij}U$ with

$$H_{ij}=\sum_{l=1}^{l_0}J_{ij}^{(l)}(\vec{S}_i\vec{S}_j)^l\ .$$

Under these conditions it follows similarly as in Ref. 3 that the ground state is a singlet ($S=0$). For the results for $l_0=3$ and $s=\frac{3}{2},2$, see the Appendix, Sec. 2.

III. SUFFICIENT AND NECESSARY CONDITIONS FOR FGS

We first derive conditions for FGS for the models (1) with only NN interactions:

$$H=\sum_{l=1}^{l_0}J^{(l)}\sum_{\mathrm{NN}}(\vec{S}_n\cdot\vec{S}_{n+\delta_1})^l\ . \tag{7}$$

Here already the case $l_0=2$ shows the qualitative features of G_F, the region in the space of the coupling constants, where the ground state is ferromagnetic.

A. Sufficient conditions

For deriving sufficient conditions we use the methods developed and applied to systems (1) with only bilinear exchange in previous papers.[20–22] The cell we use is the two-spin system (5a). For this, G_F is given by

$$B^{(k)}<0,\quad k=1,\ldots,2s \tag{8}$$

which are all independent, except for $l_0\leq 2$. Note that (8) is sufficient for systems (7) for arbitrary lattice since cell (5a) applies directly to systems (7).

1. $l_0=2$

Let us now consider the case $l_0=2$ in detail. Here (8) reduces to

$$j^{(1)}<-2\left[1-\frac{1}{s}\right]j^{(2)}\ , \tag{9a}$$

$$j^{(1)}<\frac{1}{s}j^{(2)}\ . \tag{9b}$$

$j^{(l)}$ is defined as $j^{(l)}=s^{2l}J^{(l)}$. (9a) and (9b) are presented in Fig. 1.

Let us comment on (9a) and (9b). This will also be useful for deriving necessary conditions.

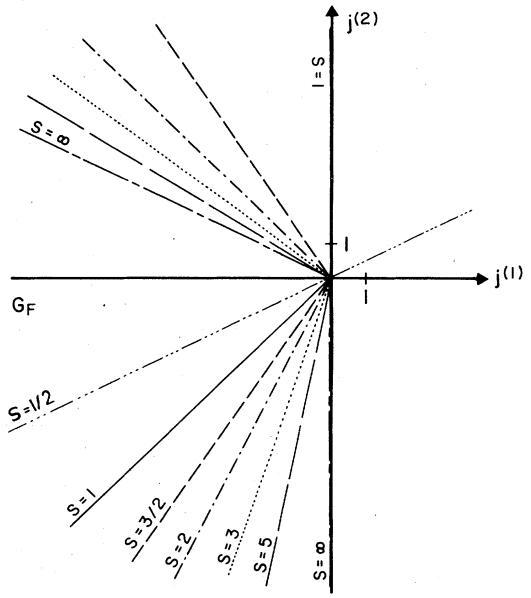

FIG. 1. Spin dependence of G_F for $H_2=J^{(1)}\vec{S}_1\cdot\vec{S}_2+J^{(2)}(\vec{S}_1\cdot\vec{S}_2)^2$. $j^{(k)}=s^{2k}J^{(k)}$ are the reduced coupling constants.

(*i*) *Sector*: $j^{(1)}<0$, $j^{(2)}>0$. The ground state for $j^{(2)}=0$ is ferromagnetic and for $j^{(1)}=0$ nonferromagnetic, except for $s\leq 1$. (For $s=1$ and $j^{(1)}=0$, the FGS is degenerate with nonferromagnetic states.) Thus for $s\leq 1$ there is always FGS whereas for $s>1$ the ground state is expected to be nonferromagnetic, at least for small $j^{(1)}$. This agrees with (9a).

(*ii*) *Sector*: $j^{(1)}<0$, $j^{(2)}<0$. Similar as in (i) for $j^{(1)}=0$, the ground state is antiferromagnetic (with energy $j^{(2)}[1+(1/s)]^2$). Thus it is expected that G_F increases with increasing s, consistent with (9b). This shows that the qualitative behavior at G_F for (9) with $l_0=2$ is as expected.

Improvement. So far we used the smallest cell leading to conditions for all s. The reason was that we were interested more in qualitative results to study the influence of s and higher-order exchange on G_F.

By choosing larger cells we were able to improve the results. We used a cell H_3, which is H_2 extended to three lattice sites, for $s=\frac{3}{2}$, 2, and $\frac{5}{2}$. The results are presented in Table I. (For the calculation see the Appendix.) Compared with the threshold values $\alpha=1/s$ of cell H_2, the results for H_3 in Table I are an essential improvement. The reason is that the antiferromagnetic ground-state energy of H_3 is a much better approach to the N-site system than of H_2. This is similar as for the bilinear case.

2. $l_0>2$

Here we study the influence of higher-order exchange. The sufficient conditions for FGS following from (8) describe a cone of $2s$ planes. This is illustrated for $l_0=3$ in Fig. 2.

TABLE I. Necessary and sufficient conditions for FGS for $H=\sum_{l=1}^{2}J^{(l)}\sum_{\mathrm{NN}}(\vec{\mathbf{S}}_n\vec{\mathbf{S}}_{n+\delta_1})^l$ in the sector $j^{(1)},j^{(2)}<0$. (FGS for $j^{(1)}<\alpha j^{(2)}$.) The sufficient conditions are valid for arbitrary lattice and also represent the threshold conditions for the cells H_2, H_3, and H_3^{cycl}. (See the Appendix for the definition of H_3^{cycl}.) The necessary condition $|\theta,0-0\rangle$ holds also for the arbitrary lattice whereas $|\psi_3(\lambda_1)\rangle$ and the Anderson approximation hold for the linear chain.

	α Sufficient			α Necessary		Anderson
Spin	Cell H_2	Cell H_3	Cell H_3^{cycl}	$\lvert\theta,0-0\rangle$	$\lvert\psi_3(\lambda_1)\rangle,\ldots$	approximation
1	1	1	1	0.5	1	0.363
$\frac{3}{2}$	$\frac{2}{3}$	0.545 454	0.476 19	0.2222	0.465 03	0.242
2	0.5	0.373 724	0.333 33	0.125	0.280 52	0.1815
$\frac{5}{2}$	0.4	0.274 307	0.233 06	0.08		0.1452
$s>\frac{5}{2}$	$\frac{1}{s}$			$\frac{1}{2s^2}$		$\frac{0.363}{s}$

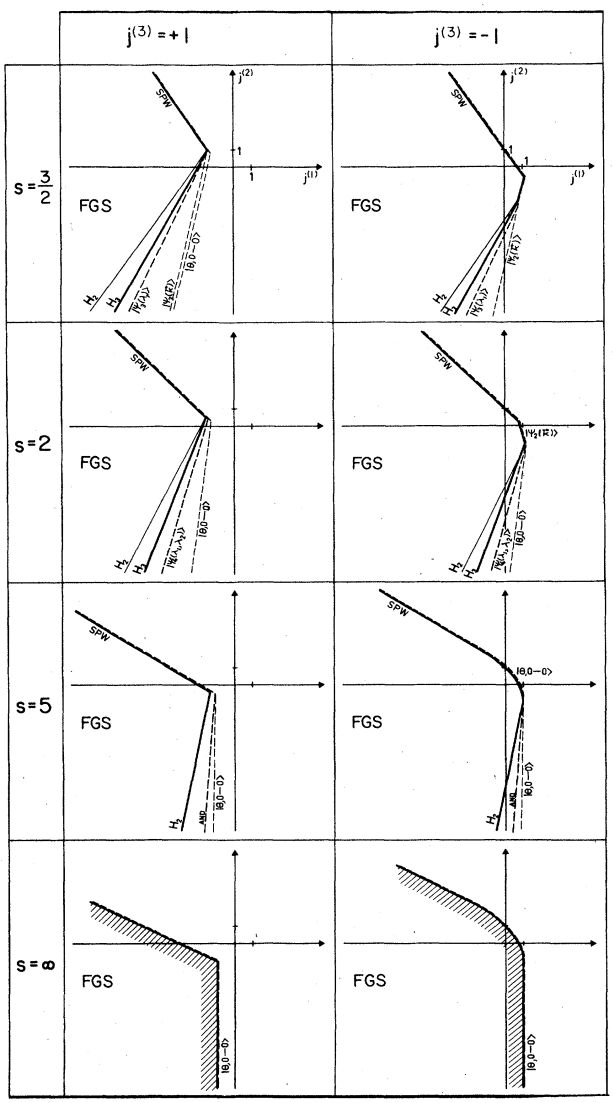

FIG. 2. Necessary (dashed lines) and sufficient conditions (solid lines) for FGS for $H=\sum_{l=1}^{3}J^{(l)}\sum_{\mathrm{NN}}(\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_1})^l$. Conditions obtained by $|\psi_3(\lambda_1)\rangle$, $|\psi_4(\lambda_1,\lambda_2)\rangle$, and AND are valid for the linear chain, $|\psi_3(\vec{k})\rangle$ for the simple-cubic, and all the others for arbitrary lattices. The method of the convex hull was used to obtain conditions denoted by H_3. SPW denotes spin wave (1-magnon) and AND the Anderson approximation.

3. *Magnetic field and NNN interaction*

We also studied the influence of a magnetic field and of NNN interactions on G_F. The Hamiltonian in the latter case is

$$H=\sum_{l=1}^{2}J_1^{(l)}\sum_{\mathrm{NN}}(\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_1})^l+\sum_{l=1}^{2}J_2^{(l)}\sum_{\mathrm{NNN}}(\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_2})^l\,. \tag{10}$$

As cells we again used (5a) with external field and for (10) we used cell H_3 with NNN interactions:

$$\tilde{H}_3=\sum_{l=1}^{2}J_1^{(l)}[(\vec{\mathbf{S}}_1\cdot\vec{\mathbf{S}}_2)^l+(\vec{\mathbf{S}}_2\cdot\vec{\mathbf{S}}_3)^l]+\sum_{l=1}^{2}J_2^{(l)}(\vec{\mathbf{S}}_1\cdot\vec{\mathbf{S}}_3)^l\,. \tag{11}$$

The results for the case of a magnetic field and simple-cubic lattices are presented in Fig. 3 and for system (10) for the linear chain are shown in Fig. 4.

Unfortunately for systems with higher-order exchange the eigenvalue problem is solvable only for simple cells for all s. For example, even for H_3 and $\tilde{H}_3$ the eigenvalues

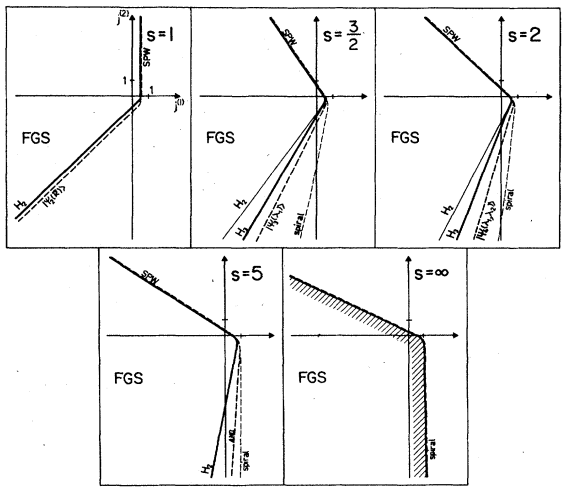

FIG. 3. Necessary (dashed lines) and sufficient conditions (solid lines) for FGS for $H=\sum_{l=1}^{2}J^{(l)}\sum_{\mathrm{NN}}(\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_1})^l-h\sum_n S_n^z$ for simple-cubic lattices. The magnetic field is fixed to $h=z_1/s$. $|\psi_3(\lambda_1)\rangle$, $|\psi_4(\lambda_1,\lambda_2)\rangle$, and AND hold again for the linear chain. [Spiral denotes states (16).]

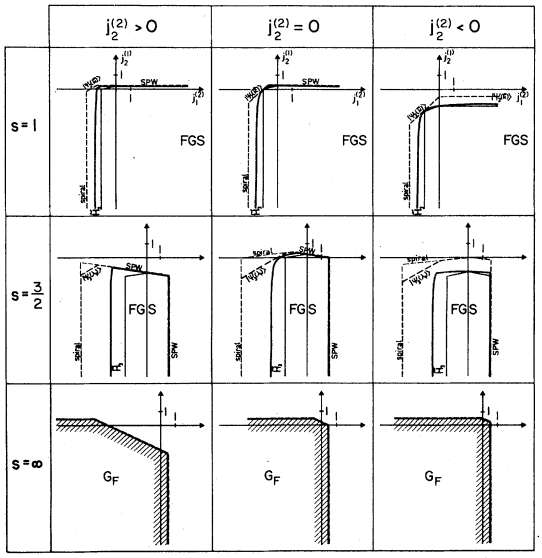

FIG. 4. Necessary (dashed lines) and sufficient conditions (solid lines) for $H=\sum_{l=1}^{2}J_1^{(l)}\sum_{\mathrm{NN}}(\vec{S}_n\cdot\vec{S}_{n+\delta_1})^l+\sum_{l=1}^{2}J_2^{(l)}\times\sum_{\mathrm{NNN}}(\vec{S}_n\cdot\vec{S}_{n+\delta_2})^l$ for the linear chain. The thin solid lines follow from cell $\tilde{H}_3$ with $J_1^{(2)}=0$ and cell H_2 (convex hull).

cannot be given in closed form for arbitrary s. However, using the method of the convex hull[22] we can obtain sufficient conditions for such cases. This method is based on the fact that the convex hull of some regions, each contained in G_F, is itself contained in G_F (Ref. 22). We applied this method to cases (2) and (3), see also Figs. 2—4.

B. Necessary conditions

The general method to obtain necessary conditions is based on the variational principle. The energies of trial states are evaluated and compared with the energy of the ferromagnetic state. Unfortunately there is not a simple successful scheme to obtain such trial states. However, in our cases, the states which were used can be classified into three groups, namely generalized cell states, spin coherent states, and physically adapted states. Since they are defined separately in each case, we shall only present the states and discuss the results.

1. Generalized cell states

We have

$$|\psi_\nu(\vec{k})\rangle=\sum_{\vec{R}_i}e^{i\vec{k}\cdot\vec{R}_i}(S_i^-)^\nu|0\rangle,\quad \nu\le 2s\ . \tag{12}$$

For lattices Λ with sublattices Λ_A and Λ_B, with $B_{ij}^{(k)}=0$ for $i,j\in\Lambda_A$ or $i,j\in\Lambda_B$,

$$\begin{aligned}&|\psi_3(\lambda_1)\rangle=|\tilde{\psi}_{(3)}\rangle+\lambda_1|\tilde{\psi}_{(2,1)}\rangle,\quad s\ge\tfrac{3}{2}\ ,\\ &|\psi_4(\lambda_1,\lambda_2)\rangle=|\tilde{\psi}_{(4)}\rangle+\lambda_1|\tilde{\psi}_{(3,1)}\rangle+\lambda_2|\tilde{\psi}_{(2,2)}\rangle,\ s\ge 2\ ,\end{aligned} \tag{13}$$

and so on, where

$$|\tilde{\psi}_{(k_1,k_2)}\rangle=\sum_{i\in\Lambda_A,\delta_1}|(k_1)_i,(k_2)_{i+\delta_1}\rangle+(-1)^{k_1+k_2}\times\sum_{i\in\Lambda_B,\delta_1}|(k_1)_i,(k_2)_{i+\delta_2}\rangle\ .$$

Here

$$|(k_1)_i,(k_2)_j\rangle=(S_i^-)^{k_1}(S_j^-)^{k_2}|0\rangle$$

with $|0\rangle$ the ferromagnetic state with all spins up.

2. Spin coherent states (Ref. 23)

These states are denoted by $|\theta_1,\phi_1,\ldots,\theta_N,\phi_N\rangle$ with

$$\langle\vec{S}_n\rangle=s\vec{e}_n(\theta_n,\phi_n) \tag{14}$$

and (θ_n,ϕ_n) giving the direction of a classical spin. We use the special cases

$$\text{(a)}\quad |\theta,0\text{—}0\rangle\ ; \tag{15}$$

$$\text{(b)}\quad \theta_n\equiv\theta\ , \tag{16}$$

$$\phi_n=\begin{cases}\vec{k}\cdot\vec{R}_n+\phi, & n\in\Lambda_A\\ \vec{k}\cdot\vec{R}_n, & n\in\Lambda_B\ .\end{cases}$$

3. Physically adapted states

We have

$$|\psi_{\mathrm{AF}}\rangle\text{: ground state of the NN antiferromagnetic bilinear Heisenberg model} \tag{17}$$

(AF is antiferromagnetic). The conditions following from states of types (1) and (2) are calculated in the Appendix. Now we only discuss the results.

We first restrict ourselves to the case (7).

States (1). Extending the 2-site cell states of H_2 [which led to condition (8)] to N-site states we try to approach condition (8). In fact, the 1-magnon states $|\psi_1(\vec{k})\rangle$ and the state $|\psi_2(0)\rangle$ lead to conditions

$$B^{(1)}<0 \tag{18a}$$

and

$$B^{(2)}<0\ , \tag{18b}$$

agreeing with condition (9) for $k=1,2$. Therefore, the hyperplanes belonging to $S=2s-1$ $(k=1)$ and $S=2s-2$ $(k=2)$ of the threshold surface of H_2 also belong to the threshold surface of Hamiltonian (7). The reason is that states $|\psi_1(\vec{k})\rangle$ and $|\psi_2(0)\rangle$ are also eigenstates of H_2 [Eq. (5)] on the hyperplanes $B^{(1)}$ and $B^{(2)}=0$, respectively. Such states do not exist on the hyperplanes $B^{(\nu)}=0$ for $\nu>2$. However, the (corresponding) extended 2-site states (13) are good trial states as shown by the results in Table I for $l_0=2$. Since the evaluation of the $2(s-1)$ states (13)

$$|\psi_3(\lambda_1)\rangle, |\psi_4(\lambda_1,\lambda_2)\rangle, \ldots, |\psi_{2s}(\lambda_1,\ldots)\rangle$$

becomes tedious for large s, we only calculated the cases for s up to 2. But, as will be seen below, states (17) will complete the results of states (13) to larger s.

Remark. For $s=1$ (18a) and (18b) are also sufficient, as follows from (8). Thus for $s=1$ the region G_F is known for an arbitrary lattice. For $J^{(1)}=J^{(2)}$, (7) reduces to the Schrödinger model (3) from which follows also that $J^{(1)}=J^{(2)}<0$ is part of the threshold. Note that for $J^{(1)}=0<J^{(2)}$ the FGS is highly degenerate.

States (3). As mentioned earlier these states extend the results of (13) to larger s. This will be demonstrated now for Hamiltonian (7) with $l_0=2$ and for simple-cubic lattices. Remember that for this case the sufficient conditions are (8). Since (9a) is also necessary [cf. (18a)], we are interested in couplings $j^{(1)},j^{(2)}$ with $j^{(1)},j^{(2)}<0$ and $|j^{(1)}|<|j^{(2)}|/s$. For this case Hamiltonian (7) becomes

$$\tilde{H}=-\left|\frac{j^{(1)}}{j^{(2)}}\right|\sum_{\mathrm{NN}}\frac{\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_1}}{s^2}-\sum_{\mathrm{NN}}\left[\frac{\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_1}}{s^2}\right]^2, \tag{19}$$

where the first term can be considered as a perturbation. The ground state of the second term is some antiferromagnetic state. Using the Néel state as a simple trial state we obtain as a necessary condition

$$j^{(1)}<\frac{1}{2s^2}j^{(2)}. \tag{20}$$

This condition improves, taking quantum corrections to the Néel state into account. Assuming Anderson's spin-wave approximation[24] for the quantum corrections to the energy of a state with Néel-type ordering, we would obtain

$$j^{(1)}<j^{(2)}\frac{1}{s}\frac{2\gamma}{z_1[2+(\gamma/z_1 s)]}. \tag{21}$$

For the values of γ for the various lattices see Ref. 24. z_1 is the number of NN. We should point out that (21) is only an approximation of the threshold and not a necessary condition in a strong mathematical sense, since Anderson's spin-wave energy does not belong to a state in the Hilbert space. More transparent than this approximation is a much simpler procedure, which even leads to a better result. For the ground state $|\psi_{\mathrm{AF}}\rangle$ of the NN antiferromagnetic bilinear Heisenberg model, using

$$\left\langle\psi_{\mathrm{AF}}\left|\sum_{\mathrm{NN}}\vec{\mathbf{S}}_n\cdot\vec{\mathbf{S}}_{n+\delta_1}\right|\psi_{\mathrm{AF}}\right\rangle=-\frac{Nz_1}{2}s^2[1+\gamma(s)],$$

we obtain

$$j^{(1)}<j^{(2)}\gamma(s). \tag{22}$$

Clearly $\gamma(s)$ is not known exactly for $s\geq 1$. But with the Anderson approximation[24] for $\gamma(s)$,

$$\gamma(s)=\frac{\gamma}{z_1 s}+o(1/s^2),$$

we obtain

$$j^{(1)}<j^{(2)}\frac{\gamma}{sz_1}. \tag{23}$$

Again (23) is only an approximation to the threshold and not a necessary condition, in contrast to (22).

The results of states (12)—(17) for Hamiltonian (7) with $l_0=2$ for the linear chain are presented in Table I. The extension to other lattices is straightforward, also see the Appendix.

Finally we mention that the inclusion in (13) of states with spin deviations at three different sites improves the result only a little. For example, for

$$|\tilde{\psi}_3(\lambda_1,\lambda_2)\rangle=|\tilde{\psi}_{(3)}\rangle+\lambda_1|\tilde{\psi}_{(2,1)}\rangle+\lambda_2|\tilde{\psi}_{(1,1,1)}\rangle,$$

where

$$|\tilde{\psi}_{(1,1,1)}\rangle=\sum_{\substack{i\in\Lambda_A,\\ \delta_1,\delta_1\\ \delta_1\neq\delta_1'}}S_i^-S_{i+\delta_1}^-S_{i+\delta_1'}^-|0\rangle-\sum_{\substack{i\in\Lambda_B,\\ \delta_1,\delta_1',\\ \delta_1\neq\delta_1'}}S_i^-S_{i+\delta_1}^-S_{i+\delta_1'}^-|0\rangle,$$

we obtain for the linear chain and $s=\frac{3}{2}$, $\alpha=0.467\,38$ instead of $\alpha=0.465\,03$ with $|\psi_3(\lambda_1)\rangle$. For models including higher-order exchange, a magnetic field, or NNN interactions, trial states (12)—(17) lead to conditions presented in Figs. 2—4.

IV. EXCITATIONS, MAGNONS

In this section we use periodic boundary conditions and NN interactions.

A. 1-magnon states

The 1-magnon states are

$$|\psi_1(\vec{\mathrm{k}})\rangle=\frac{1}{(2Ns)^{1/2}}\sum_i e^{i\vec{\mathrm{k}}\cdot\vec{\mathbf{R}}_i}S_i^-|0\rangle.$$

Using Eq. (4a) one finds the corresponding eigenvalues

$$E_1(\vec{\mathrm{k}})=E_F-\frac{B^{(1)}}{2}\sum_{\vec{\delta}_1}[1-\cos(\vec{\mathrm{k}}\cdot\vec{\delta}_1)], \tag{24}$$

which is a generalization of the result in Ref. 5. Thus measuring the spin-wave dispersion allows one to determine only $B^{(1)}$.

Remark. We should mention that not only the dispersion relation (24) is the same as for the bilinear model, but also all other properties of (1) restricted to the 1-magnon space, e.g., surface states, etc.

B. 2-magnon states

Separating for the 2-magnon states

$$|\psi_2\rangle=\sum_{i,j}\Phi(i,j)S_i^-S_j^-|0\rangle,$$

the center of mass

$$\Phi(i,j)=\exp[\tfrac{1}{2}i\vec{\mathbf{K}}\cdot(\vec{\mathbf{R}}_i+\vec{\mathbf{R}}_j)]\phi(\vec{\mathbf{r}}_{ij}),$$

where $\vec{\mathbf{r}}_{ij}=\vec{\mathbf{R}}_i-\vec{\mathbf{R}}_j$ is the difference between the lattice vectors $\vec{\mathbf{R}}_i$ and $\vec{\mathbf{R}}_j$, the eigenvalue equation

$H|\psi_2\rangle = E_2|\psi_2\rangle$ takes the form

$$B^{(1)}\sum_{\vec{\delta}_1}\left[\cos\left[\frac{\vec{K}\cdot\vec{\delta}_1}{2}\right]\phi(\vec{r}+\vec{\delta}_1)-\phi(\vec{r})\right]+B'\delta(0,\vec{r})\sum_{\vec{\delta}_1}\cos\left[\frac{\vec{K}\cdot\vec{\delta}_1}{2}\right]\left[\phi(\vec{\delta}_1)-\cos\left[\frac{\vec{K}\cdot\vec{\delta}_1}{2}\right]\phi(\vec{0})\right]$$

$$-B\sum_{\vec{\delta}_1}\delta(\vec{r},\vec{\delta}_1)\left[\phi(\vec{\delta}_1)-\cos\left[\frac{\vec{K}\cdot\vec{\delta}_1}{2}\right]\phi(\vec{0})\right]=\epsilon\phi(\vec{r})\,, \quad (25)$$

where

$$B'=\frac{2s}{4s-1}B^{(2)}-B^{(1)}\,,$$

$$B=\frac{2s-1}{4s-1}B^{(2)}-B^{(1)}\,,$$

and

$$\epsilon=E_2-E_F\,.$$

This generalizes the eigenvalue equation obtained by Chiu-Tsao *et al.*[11] to arbitrary spin s and arbitrary high-order exchange. Equation (25) also demonstrates that independent of l_0, the 2-magnon energies are determined only by two parameters, $B^{(1)}$ and $B^{(2)}$.

From (25) follows the 2-magnon continuum

$$\epsilon_{\vec{K}}(\vec{q})=|B^{(1)}|\sum_{\vec{\delta}_1}\left[1-\cos\left[\frac{\vec{K}\cdot\vec{\delta}_1}{2}\right]\cos\left[\frac{\vec{q}\cdot\vec{\delta}_1}{2}\right]\right]\,, \quad (26)$$

where $\vec{q}$ is the relative momentum of both magnons. This holds with the restriction $B^{(1)}=-|B^{(1)}|<0$. Similar to Chiu-Tsao *et al.*[11] for $s=1$ we have discussed the 2-magnon bound states for hypercubic lattices in any dimension limiting ourselves to $\vec{K}=K(1,1,\ldots,1)$. The energies of the bound states in the functions of $\alpha=\cos(K/2)$ and $\beta=B^{(2)}/B^{(1)}$ are qualitatively independent of s. Thus the detailed discussion of Chiu-Tsao *et al.*[11] also applies to arbitrary s. As an illustration we give the "phase" diagrams for the existence of bound states as a function of α and β. For s-type[11] bound states the "phase" diagram is presented in Fig. 5. d-type[11] bound states exist if

$$\beta<\frac{4s-1}{2s-1}\frac{I_D-D\alpha}{I_D}\,, \quad (27)$$

where

$$I_D=\frac{1}{(2\pi)^D}\int d^Dq\frac{\cos^2q_1-\cos q_1\cos q_2}{1-(1/D)\sum_j\cos q_j}$$

and D is the dimension.

Figure 5 shows that for FGS (i.e., $\beta>0$) in the three-dimensional case no bound state exists for $\vec{K}\approx 0$. This is necessary for the application of the spin-wave theory to low-temperature thermodynamics.

Besides the [at most D (Ref. 25)] exchange bound states, a single-ion bound state can exist, depending on the coefficients B', B, and $B^{(1)}$ of the "point interaction," the "NN interaction," and the "kinetic energy" in Eq. (25), respectively. This generalizes the result of other authors[6–8,11] to arbitrary s and higher-order exchange. The existence of a single-ion bound state is illustrated in Fig. 6 which represents, for the linear chain, the ratio

$$\frac{W(0)}{W(1)}=\frac{s\alpha^2}{2s-1}\frac{[t-\alpha^2-\mathrm{sgn}(t)(t^2-\alpha^2)^{1/2}]^2}{(t-\alpha^2)^2[t-\mathrm{sgn}(t)(t^2-\alpha^2)^{1/2}]^2}\,, \quad (28)$$

where $t=1+(\epsilon/2B^{(1)})$ and $W(r)$ is the probability of finding the two spin deviations at rth neighbor sites.

Remark. If the coefficient $B^{(1)}$ of the kinetic energy approaches 0, i.e., if $\beta\to+\infty$, then a single-ion bound state exists. This is based on the uncertainty relation and is similar to the effect of $\hbar\to 0$ on the bound states of the H atom.

C. ν-magnon states

For arbitrary $\{J^{(l)}\}$ the ν-magnon states for $\nu>2$ probably cannot be determined analytically, even for one dimension. Therefore, we will study the special case, where model (1) reduces to the Schrödinger model (3),

$$H=\frac{J}{2}\sum_{\mathrm{NN}}P_{i,i+\delta_1}\,. \quad (29)$$

This simplifies the eigenvalue problem as follows.

(1) The subspaces spanned by the vectors

$$|(\nu_1)_{i_1},\ldots,(\nu_n)_{i_n}\rangle=(S_{i_1}^-)^{\nu_1}\cdots(S_{i_n}^-)^{\nu_n}|0\rangle\,,$$

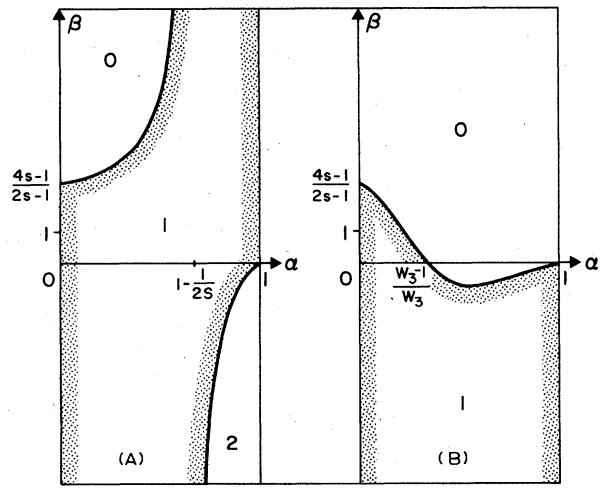

FIG. 5. "Phase diagram" for the existence of 0,1,2 s-type 2-magnon bound states below the continuum. Case (A) (with $D=1,2$) and case (B) (with $D=3$), $\vec{K}=K(1,\ldots,1)$, $\alpha=\cos(K/2)$, and $\beta=B^{(2)}/B^{(1)}$. $W_3=1.516$.

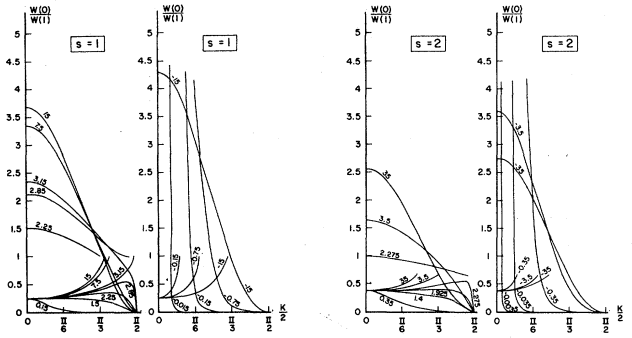

FIG. 6. Probability ratio $W(0)/W(1)$ for the 2-magnon bound states for the linear chain ($D=1$). Single-ion bound states exist below ($\beta<0$) and above ($\beta>0$) the continuum. $\beta=B^{(2)}/B^{(1)}$ is the parameter.

where $\sum_{m=1}^{n}\nu_{i_n}=\nu$ and $i_m\neq i_{m'}$ for all $m\neq m'$ are invariant with respect to Hamiltonian (29).

(2) The eigenvalue problem for (29) in the subspaces of the same type are identical. By subspaces of the same type we mean

$$[\,|\,1_i\rangle]\sim[\,|\,2_i\rangle]\sim[\,|\,3_i\rangle]\sim\cdots\,,$$

$$[\,|\,1_i,1_j\rangle]\sim[\,|\,2_i,2_j\rangle]\sim\cdots\,,$$

$$[\,|\,2_i,1_j\rangle]\sim[\,|\,3_i,1_j\rangle]\sim[\,|\,3_i,2_j\rangle]\sim\cdots\,,$$

and so on.

(3) The eigenstates of (29) in the subspaces $[\,|\,1_{i_1},\ldots,1_{i_\nu}\rangle]$ are equivalent to the ν-magnon states of the Heisenberg model with $s=\frac{1}{2}$.[26]

Let us now consider the 1-, 2-, and 3-magnon states in more detail.

$\nu=1$: There is of course no new insight.

$\nu=2$: For $s\geq 1$ there are two subspaces $[\,|\,2_i\rangle]$ and $[\,|\,1_i,1_j\rangle]$. In the first subspace the eigenvalues are equal to the 1-magnon energies

$$E_2(\vec{K})=E_1(\vec{K})=E_F-J\sum_{\vec{\delta}_1}[1-\cos(\vec{K}\cdot\vec{\delta}_1)]\,, \qquad (30)$$

as follows from above.

In the subspace $[\,|\,1_i,1_j\rangle]$ the bound states are exchange bound states. For the linear chain the eigenvalues of these states are

$$E_2(K)=E_F-J\sin^2\left[\frac{K}{2}\right].$$

$\nu=3$: The two subspaces $[\,|\,3_i\rangle]$ and $[\,|\,1_i,1_j,1_k\rangle]$ are already discussed, using remarks (2) and (3). The third subspace $[\,|\,2_i,1_j\rangle]$ can be decomposed into a symmetric and an antisymmetric part,

$$[\,|\,2_i,1_j\rangle\pm|\,2_j,1_i\rangle]\,.$$

The eigenstates for the symmetric part are again identical to the 2-magnon states for the Heisenberg model with $s=\frac{1}{2}$. Thus there exists a 3-magnon bound state (dependent on the total momentum and dimension) which is composed of a single-ion 2-magnon bound state and a 1-magnon bound state. The antisymmetric eigenstates have eigenvalues, which are in the continuum.

For arbitrary ν the following classification of the bound states is now obvious: Besides the pure single-ion and pure exchange bound states there also exists all combinations of both. The reason is that for model (29) in the ν-magnon equation, similar to (25), the coefficients of all the interaction terms and the kinetic energy are equal. We believe that this classification for model (1) stays valid for more general coupling constants $\{J_{ij}^{(l)}\}$. If not all higher-order exchange interactions up to $2s$ are present, then it is expected that only part of these types of bound states appear. This was already true for $\nu=2$. (See Fig. 6.)

V. DISCUSSION

For the Heisenberg model with higher-order exchange we showed that for determining the coupling constants, all ν-magnon energies up to $\nu=2s$ have to be measured. In particular in the $\nu=1$ [$(\nu=2)$] magnon space it is impossible to decide if higher than bilinear (biquadratic) exchange is present if $s>\frac{1}{2}$ ($s>1$).

Extending the Munro theorem[3] to bicubic interactions we obtained conditions for the couplings such that the ground state is a singlet. The results indicate that this method is not so successful as if only bilinear interactions are present. For example, for NN interactions, $l_0=3$ and $s\to\infty$, it is obvious that the ground state is antiferromagnetic for couplings $j^{(1)}$, $j^{(3)}>0$, and $j^{(2)}<0$ whereas the conditions of the Munro theorem only describe a part of this domain.

Generalizing the methods presented earlier we derived conditions for FGS for such models. Let us comment on the results.

(i) The spin-wave stability[20] is stringent in a subspace of the coupling space as the figures show. However, in contrast to the bilinear case, it represents only a part of the threshold in the classical limit $s\to\infty$. This could indicate that even in the classical case for higher exchange interactions the local stability of FGS is not sufficient.

(ii) Besides the spin-wave states, we used trial states which were "extended-cell states." This procedure improved the results.

(iii) The typical situation of the spin dependence of G_F in the coupling space is given in Fig. 1, which was discussed in Sec. III. Here we only want to point out the qualitative difference between the classical and the quantum case: For couplings $J^{(1)},J^{(2)}<0$ there is always FGS in the classical case, in contrast to the quantum case. The same holds for each pair of an odd and even power exchange. For example, the spin dependence of the region for FGS in Fig. 4 can be discussed as those in Fig. 1.

(iv) The coupling constants belonging to even powers of the exchange influence the domain G_F in a similar way, which differs only by numeric values from one power to another. The same holds for the odd powers. For bilinear and bicubic interactions this follows from Fig. 2: There is only a quantitative difference between the diagrams of Fig. 2 and the corresponding ones where $j^{(1)}=\pm 1$ fixed, instead of $j^{(3)}$.

(v) The inequalities[22] for the thresholds of the Ising, classical, and quantum models are no longer true as can be seen in Fig. 1. For the corresponding Ising model the threshold is

$$j^{(1)}<0\ ,$$

$$j^{(1)}<-2j^{(2)}\left[1-\frac{1}{2s}\right]\ .$$

Note that including higher-order exchange, the ground state of the Ising model with spin s may be different from that of the $s=\frac{1}{2}$ Ising model. Clearly $G_F^{qm}\to G_F^{cl}$ as $s\to\infty$, consistent with the classical spin limit.

(vi) The threshold surfaces are composed of curved and flat pieces. On the curved pieces the ferromagnetic states are degenerate with a state which changes continuously along the surface, whereas on a flat piece the degenerate states are constant. On the intersection, the ground state changes its symmetry discontinuously (Fig. 4).

(vii) For models (1) with FGS, there exist no 2-magnon bound states for $\vec{K}\approx\vec{0}$ in the case of a single-cubic lattice. This is important for the theory of low-temperature thermodynamics.

Finally, studying the elementary excitations of the Schrödinger model, which contains all powers of the exchange, the ν-magnon bound states were classified into pure single-ion, pure exchange bound states and all combinations of both. Similar as in the case of the 2-magnon state, it is expected that this classification stays valid for more general couplings $J_{nm}^{(l)}$, if still $l_0=\nu$. If $l_0<\nu$ then only part of these types of bound states appear. Thus higher-order exchange interaction manifests in the types of bound states which appear.

(viii) Finally, let us say a few words about the molecular-field approximation (MFA). For only bilinear interactions, to obtain G_F, the MFA is a first simple approximation. Using the spin-wave stability condition, the results are improved. In a further step, s-dependent results were obtained using the cell method and better adapted trial states.[20–22]

Of course MFA can be extended to higher exchange interactions. It then includes at least l_0 ordering parameters (magnetization, quadrupole momentum, ...) and must be performed for each spin s separately.[5,27] Therefore, for determining G_F in functions of s, it is rather tedious and does not necessarily give a good result. This is explicitly shown for Hamiltonian (1) with $s=1$, $s=\frac{3}{2}$, and $l_0=2$.

MFA: $s=1$,

$$\tilde{j}^{(1)}<0,\ \ \tilde{j}^{(1)}<\tilde{j}^{(2)}\ ,$$

$s=\frac{3}{2}$,

$$\tilde{j}^{(1)}+\tfrac{2}{3}\tilde{j}^{(2)}<0,\ \ \tilde{j}^{(1)}-\tfrac{2}{5}\tilde{j}^{(2)}<0\ ,$$

where $\tilde{j}^{(k)}=\sum_m s^{2k}J_{nm}^{(k)}$.

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APPENDIX

1. Proof of the statement for $s\neq s'$

Let $|(\nu_1)_{i_1},\ldots,(\nu_n)_{i_n}\rangle$ be the normalized state with ν_k spin deviations at site i_k, $k=1,\ldots,n$. Clearly these states with $\sum_{i=1}^n\nu_i=\nu$ and $n\leq\nu$ form a basis of the ν-magnon space $\mathscr{H}_\nu$. The analogous states exist also in $\mathscr{H}'_\nu$, the ν-magnon space of the system with spin s'. Thus for $\nu\leq\min\{2s,2s'\}$

$$H\,|_{\mathscr{H}_\nu}\equiv H'\,|_{\mathscr{H}'_\nu}$$

means that

$$(H-H')\,|(\nu_1)_{i_1},\ldots,(\nu_n)_{i_n}\rangle=0$$

for all these states with $\sum_{i=1}^n\nu_i=\nu$.

Proof of the statement by induction. Suppose that from $H\,|_{\mathscr{H}_\nu}\equiv H'\,|_{\mathscr{H}'_\nu}$ follows (i') and (ii') for $\nu=2k_0+1$. Using this we obtain from

$$H\,|(\nu+1)_i\rangle=H'\,|(\nu+1)_i\rangle$$

that

$$B_{in}^{(\nu+1)}=B_{in}'^{(\nu+1)}=0\ ,$$

and from

$$H\,|(\nu+2)_i\rangle=H'\,|(\nu+2)_i\rangle$$

that

$$B_{in}^{(\nu+2)}-B_{in}^{(1)}=B_{in}'^{(\nu+2)}-B_{in}^{(1)}=0\ .$$

Thus (i') and (ii') are true for $\nu=2k_0+2$, $2k_0+3$. The reverse statement follows directly from (4a).

Consider now H_2 of Eq. (5a) with

$$\begin{aligned}&B_{ij}^{(2k)}=0\ \text{ for all } 2k\leq\nu\ ,\\ &B_{ij}^{(2k+1)}=B_{ij}^{(1)}\ \text{ for all } 2k+1\leq\nu\ .\end{aligned}\tag{A1}$$

Then it can be seen, using (5b), that

$$H_2\,|_{\mathscr{H}_\nu}=E_F^{(2)}-\tfrac{1}{4}B_{ij}^{(1)}+\tfrac{1}{4}B_{ij}^{(1)}P_{ij}\,|_{\mathscr{H}_\nu}\ .$$

Since (A1) holds for all i,j, it follows that $H\,|_{\mathscr{H}_\nu}$ has the form (6).

2. Conditions following from the extension of Munro's theorem

We have for $s=\frac{3}{2}$,

$$J_{ij}^{(2)} \le \tfrac{11}{4} J_{ij}^{(3)} \le 0 ,$$
$$J_{ij}^{(1)} - \tfrac{1}{2} J_{ij}^{(2)} + \tfrac{103}{16} J_{ij}^{(3)} \le 0, \qquad i,j \in \Lambda_A \text{ or } \Lambda_B$$
$$J_{ij}^{(1)} - \tfrac{5}{2} J_{ij}^{(2)} + \tfrac{191}{16} J_{ij}^{(3)} \le 0 ,$$

$$J_{ij}^{(2)} \le \tfrac{5}{4} J_{ij}^{(3)} , J_{ij}^{(3)} \ge 0 ,$$
$$J_{ij}^{(1)} + \tfrac{3}{2} J_{ij}^{(2)} + \tfrac{63}{16} J_{ij}^{(3)} \ge 0 , \qquad -i \in \Lambda_A,\ j \in \Lambda_B \text{ or vice versa} .$$

For $s=2$,

$$J_{ij}^{(2)} \le 5 J_{ij}^{(3)} \le 0 ,$$
$$J_{ij}^{(1)} - 2 J_{ij}^{(2)} + 19 J_{ij}^{(3)} \le 0 , \qquad i,j \in \Lambda_A \text{ or } \Lambda_B$$

$$J_{ij}^{(2)} \le -J_{ij}^{(3)} \le 0 ,$$
$$J_{ij}^{(1)} + 4 J_{ij}^{(2)} + 16 J_{ij}^{(3)} \ge 0 , \qquad i \in \Lambda_A,\ j \in \Lambda_B \text{ or vice versa} .$$

3. Threshold condition for cell H_3 and $\tilde{H}_3$

We have

$$H_3 = J(\vec{S}_1 \cdot \vec{S}_2 + \vec{S}_2 \cdot \vec{S}_3) + J'[(\vec{S}_1 \cdot \vec{S}_2)^2 + (\vec{S}_2 \cdot \vec{S}_3)^2] ,$$
$$\tilde{H}_3 = H_3 + K \vec{S}_1 \cdot \vec{S}_3 + K'(\vec{S}_1 \cdot \vec{S}_3)^2 . \tag{A2}$$

The matrix elements for $\tilde{H}_3$ were calculated by Griffith[28] for $s=1$ and $\frac{3}{2}$ using the technique of irreducible tensor operators. Clearly from these the threshold conditions follow. Here we only give the conditions for $K=K'=0$. (Conditions for $K,K' \neq 0$, see Fig. 4.)

For $s=1$

$$J<0, \quad J<J' , \tag{A3}$$

for $s=\frac{3}{2}$

$$J < \tfrac{189}{154} J', \quad J < -\tfrac{3}{2} J' , \tag{A4}$$

for $s=2$

$$J < 1.495 J', \quad J < -4J' , \tag{A5}$$

and for $s=\frac{5}{2}$

$$J < 1.714 J', \quad J < -\tfrac{15}{2} J' . \tag{A6}$$

Cyclic cell (can be applied to hexagonal lattice),

$$H_3^{\text{cycl}} = J(\vec{S}_1 \cdot \vec{S}_2 + \vec{S}_2 \cdot \vec{S}_3 + \vec{S}_3 \cdot \vec{S}_1) + J'[(\vec{S}_1 \cdot \vec{S}_2)^2 + (\vec{S}_2 \cdot \vec{S}_3)^2 + (\vec{S}_3 \cdot \vec{S}_1)^2] . \tag{A7}$$

From the eigenvalues of H_3^{cycl} in Ref. 26 it follows for the threshold condition

$$J < -2_s(s-1)J' \text{ (spin waves)} ,$$
$$J < \alpha_s J' , \tag{A8}$$

with $\alpha_s = 1, \frac{15}{14}, \frac{4}{3}$, and $\frac{1}{55}[24(\sqrt{19}-1) - \frac{1}{2}]$ for $s = 1, \frac{3}{2}$, 2, and $\frac{5}{2}$, respectively. From (A2) to (A8) sufficient conditions follow for FGS which are listed in Table I and given in Figs. 2–4, respectively.

Illustration of the method of the convex hull.

(1) *Model (7) with* $l_0=3$. The sufficient condition following from H_2 is a cone of $2s$ planes (see Fig. 2). In the plane $J^{(3)}=0$ this condition improves using cell H_3. Now, from the convexity property of G_F it follows that the convex hull of the conditions obtained by using H_2 and H_3 is also sufficient (see Fig. 2).

(2) *Model (11).* Cell H_2 leads to a condition in the subspace $J_2^{(1)}=J_2^{(2)}=0$. The same is true for cell $\tilde{H}_3$ with $J_1^{(2)}=0$ for the subspace $J_1^{(2)}=0$. Again the convex hull of both is sufficient for FGS (see Fig. 4). Note that this is easily feasible for arbitrary s, since $\tilde{H}_3$ with $J_1^{(2)}=0$ can easily be diagonalized.

TABLE II. Values α, β, and γ for the simple-cubic lattices.

	Linear chain	Quadratic	Simple-cubic
α	1.046 312	0.7946	0.703 13
β	1.122 06	0.905 75	0.699 32
γ	0.726	0.632	0.58

4. Necessary conditions

States $|\psi_\nu(\vec{k})\rangle$: (12). For the Hamiltonian (1) with external magnetic field we obtain as necessary conditions, using (4a),

$$\sum_{l=1}^{\nu}[\tilde{B}^{(l)}(0)+(-1)^l\tilde{B}^{(l)}(\vec{k})]\frac{(2s)!\nu!(4s-2l+1)(4s-l-\nu)!}{(2s-\nu)!l!(\nu-l)!(4s-l+1)!}<h\nu\ , \tag{A9}$$

where

$$\tilde{B}^{(l)}(\vec{k})=\sum_n B_{in}^{(l)}e^{i\vec{k}(\vec{R}_i-\vec{R}_n)}\ .$$

States $|\psi_3(\lambda_1)\rangle$: Using the matrix elements of Hamiltonian (7), including a magnetic field, for simple-cubic lattices in the subspace $|\psi_3(\lambda_1)\rangle$ are

$$\begin{aligned}
A_{11}&=E_F-z_1\left[\frac{3(s-1)}{4s-3}B^{(1)}+\frac{s}{4s-3}B^{(3)}\right]+3h\ ,\\
A_{12}&=\frac{[3s(s-1)z_1]^{1/2}}{4s-3}(B^{(1)}-B^{(3)})=A_{21}\ ,\\
A_{22}&=E_F-\frac{s}{4s-3}B^{(1)}-\frac{3(s-1)}{4s-3}B^{(3)}-(z_1-1)\left[B^{(1)}+\frac{s}{4s-1}B^{(2)}\right]+3h\ .
\end{aligned} \tag{A10}$$

Similarly one obtains the matrix elements in the subspace $|\psi_4(\lambda_1,\lambda_2)\rangle$.

Spin-coherent states. For the Hamiltonian (1) with $l_0=3$ and a magnetic field we obtain

$$\begin{aligned}
&\langle\theta_1,\phi_1,\ldots,\theta_N,\phi_N|H|\theta_1,\phi_1,\ldots,\theta_N,\phi_N\rangle\\
&=\frac{s^2}{2}\sum_{i,j}J_{ij}^{(1)}(\vec{e}_i,\vec{e}_j)+\tfrac{1}{2}\sum_{i,j}J_{ij}^{(2)}[s^2(s-\tfrac{1}{2})^2(\vec{e}_i,\vec{e}_j)^2-\frac{s^2}{2}(\vec{e}_i,\vec{e}_j)+s^2(s+\tfrac{1}{4})]\\
&\quad+\tfrac{1}{2}\sum_{i,j}J_{ij}^{(3)}\Bigg[s^2(s-1)^2(s-\tfrac{1}{2})^2(\vec{e}_i,\vec{e}_j)^3-2s^2(s-\tfrac{1}{2})^2(\vec{e}_i,\vec{e}_j)^2\\
&\qquad+s^2\left[3s^2-\tfrac{7}{4}s^2+\frac{s}{2}+\tfrac{1}{4}\right](\vec{e}_i,\vec{e}_j)+s^3\left[\frac{s}{2}-1\right]\Bigg]-hs\sum_i\cos\theta_i\ .
\end{aligned} \tag{A11}$$

We specialize now to the cases Hamiltonian (1) considered in Sec. III.

a. Hamiltonian (7) with $l_0=2$: Arbitrary lattice and spin

We have

$$|\psi_1(\vec{k})\rangle,|\theta,0\text{—}0\rangle:\ J^{(1)}<-2s(s-1)J^{(2)}\ ,$$

$$|\psi_{2s}(\vec{k})\rangle,|\theta,0\text{—}0\rangle:\ J^{(1)}<\tfrac{1}{2}J^{(2)}\ .$$

Simple-cubic lattices. For $s=\frac{3}{2}$

$$|\psi_3(\lambda_1)\rangle:\ J^{(1)}<\alpha J^{(2)}\ ,$$

for $s=2$

$$|\psi_4(\lambda_1,\lambda_2)\rangle:\ J^{(1)}<\beta J^{(2)}\ ,$$

and for s large

$$|\psi_{\mathrm{AF}}\rangle:\ J^{(1)}<s\frac{\gamma}{z_1}J^{(2)}\ ,$$

Anderson approximation: $J^{(1)}<s\dfrac{2\gamma}{z_1[2+(\gamma/z_1 s)]}J^{(2)}$,

where α, β, and γ are listed in Table II.

b. Hamiltonian (7) with $l_0=3$: Arbitrary lattice and spin

We have

$$|\psi_1(\vec{k})\rangle:\ J^{(1)}+2s(s-1)J^{(2)}+(3s^4-6s^3+4s^2)]^{(3)}<0\ ,$$

$$|\psi_2(\vec{k})\rangle:\ J^{(1)}+(2s^2-4s+1)J^{(2)}+(3s^4-12s^3+19s^2-8s+1)J^{(3)}<0\ ,$$

$$|\psi_3(\vec{k})\rangle:\ J^{(1)}+(2s^2-4s+1)J^{(2)}+(3s^4-12s^3+21s^2-11s+2)J^{(3)}<(s-1)(2s-1)J^{(3)}\frac{1}{z_1}\sum_{\delta_1}\cos(\vec{k}\,\vec{\delta}_1)\ ,$$

$|\theta,0\text{—}0\rangle$: follows directly from (A11) .

Simple-cubic lattices. For $s=\frac{3}{2}$

$$|\psi_3(\lambda_1)\rangle:\ \tfrac{1}{2}(9z_1-1)(J^{(1)})^2-\tfrac{1}{8}(15z_1+57)(J^{(2)})^2$$
$$+\left[\frac{92\,745z_1-6561}{512}\right](J^{(3)})^2+\tfrac{3}{2}(z_1-1)J^{(1)}J^{(2)}+\left[\frac{951z_1-15}{16}\right]J^{(1)}J^{(3)}+\left[\frac{669z_1+627}{32}\right]J^{(2)}J^{(3)}>0\ .$$

$s=2$: $|\psi_4(\lambda_1,\lambda_2)\rangle$ is a complicated condition, we only give the result in Fig. 2. For s larger

$$|\psi_{\mathrm{AND}}\rangle:\ J^{(1)}-s\frac{2\gamma}{z_1[2+(\gamma/sz_1)]}J^{(2)}+s^4\left[\frac{2+(3\gamma/sz_1)}{2+(\gamma/sz_1)}\right]J^{(3)}<0\ .$$

c. Hamiltonian (7) with $l_0=2$ and a magnetic field h: Simple-cubic lattices

We have

$$|\psi_1(\vec{k})\rangle:\ J^{(1)}+2s(s-1)J^{(2)}<\frac{h}{2sz_1}\ ,$$

$$|\psi_2(\vec{k})\rangle:\ J^{(1)}+(2s^2-4s+1)J^{(2)}<\frac{h}{sz_1}\ .$$

States (15) with $\phi=0$. We have

$$(1+\cos\theta)[J^{(1)}+2s(s-1)J^{(2)}]-2(s-\tfrac{1}{2})^2(1+\cos\theta)^2(1-\cos\theta)J^{(2)}<\frac{h}{sz_1}\ ,$$

for $s=\frac{3}{2}$

$$|\psi_3(\lambda_1)\rangle:\ \frac{z_1(9z_1-1)}{4}(J^{(1)})^2-\frac{3z_1(5z_1+19)}{16}(J^{(2)})^2+\frac{3z_1(z_1-1)}{4}J^{(1)}J^{(2)}-h\left[3z_1J^{(1)}+\frac{(z_1-4)}{2}J^{(2)}\right]+h^2>0\ .$$

$s=2$: $|\psi_4(\lambda_1,\lambda_2)\rangle$ is a complicated condition, the result is given in Fig. 3. For s larger

Anderson approximation: $J^{(1)}-s\dfrac{2\gamma}{z_1[2+(\gamma/sz_1)]}J^{(2)}$
$$<\frac{2h}{sz_1}\frac{1}{[2+(\gamma/sz_1)]}\ ,$$

$$|\psi_{\mathrm{AF}}\rangle:\ J^{(1)}-s^2\gamma(s)J^{(2)}<\frac{2h}{sz_1[2+\gamma(s)]}\ .$$

We therefore have the Anderson approximation

$$J^{(1)}-s\frac{\gamma}{z_1}J^{(2)}<\frac{2h}{sz_1[2+(\gamma/z_1s)]}\ .$$

d. Hamiltonian (10)

Similarly as above; necessary conditions can be derived for this case.

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