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Objekttyp: **Article**

Zeitschrift: **Helvetica Physica Acta**

Band (Jahr): **41 (1968)**

Heft 6-7

PDF erstellt am: **22.07.2024**

Persistenter Link: https://doi.org/10.5169/seals-113929

Magnetically Coupled Impurities in a Linear Chain Heisenberg Ferromagnet

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Zusammenfassung. Ein Fremdspin verändert das Spinwellenspektrum einer ferromagnetischen Kette, und im Grundzustand treten um den Fremdspin herum Spinabweichungen auf, die eine örtliche magnetische Polarisation bewirken. Diese werden, besonders im Falle des antiferromagnetisch gekoppelten Fremdspins, untersucht. Der Effekt eines beliebigen äusseren Magnetfeldes und verschiedener g-Faktoren wird mitberücksichtigt.

I. Introduction

Materials such as EuS [1], $CrBr_3$ [2], CrO_2 [3] and EuO [4] are ferromagnetic insulators. The properties of these substances are considerably changed by the introduction of other magnetic ions in the metal positions. A particular difficulty in the understanding of the impurity-induced properties stems from the fact that perturbation theories break down when applied. The reason for this breakdown is that the magnetic coupling constant of the impurity ion to its surrounding is often largely different from that of the host metal ions to each other.

Therefore, more sophisticated theories have to be used, such as the technique of Green functions [5]. The important features of the solution are: (1) The appearance of localized states outside the spin wave band, (2) Resonant states in the band. For a ferromagnetically coupled impurity, the problem has been extensively treated [6]. The antiferromagnetically coupled impurity presents more difficulties, because the total spin of the ground state may deviate by more than one unit from the totally aligned state. An approximate analysis [7] leads to ground state energies which are 50% higher than those obtained in an exact solution of single spin deviation states for the case of impurity spin 1/2 [8].

The fact that the excitation energies of spin deviations from the fully aligned state are negative for states below the spin wave band for an extended range of parameters leads us to a variational calculation of the ground state in which the mixing of higher (> 1) spin deviation states is considered. This will be described in a subsequent publication.

It is the object of this paper to treat the linear chain with arbitrary host and impurity spins S and S_0, exchange constants J and J_0 and Landé-factors g and g_0,

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respectively. An external magnetic field H of arbitrary magnitude is applied to the system.

It should be pointed out that we use the linear spin-wave approximation, but in contrast to previous work, obtain the correct ground state energy (as known for $S_0 = 1/2$) [8]. This is achieved by using the fully aligned state as the state with no excitations both for ferro- and antiferromagnetic spin waves.

II. The Green Function Formalism

Figure 1 shows the linear chain Heisenberg ferromagnetic with an impurity at the origin. Its Hamiltonian is

$$\hat{\mathcal{H}} = -J \sum_{i=1}^{N-2} \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_{i+1} - J_0 \, \hat{\boldsymbol{S}}_0 \cdot (\hat{\boldsymbol{S}}_1 + \hat{\boldsymbol{S}}_{N-1}) - \mu \, g \, H \sum_{i=1}^{N-1} \hat{S}_i^z - \mu \, g_0 \, H \, \hat{S}_0^z , \tag{1}$$

where J is the host exchange integral and is positive, J_0 is the impurity exchange integral, $\hat{\boldsymbol{S}}_i$ is the spin operator for site i, N is the number of atoms (even), H is an external magnetic field in the z direction, g and g_0 are the host and impurity Landé-factors, and μ is the Bohr magneton. We impose periodic boundary conditions by requiring $\hat{\boldsymbol{S}}_N = \hat{\boldsymbol{S}}_0$.

In terms of the HOLSTEIN-PRIMAKOFF [9] boson operators Equation [1] may be written as

$$\begin{aligned} \hat{\mathcal{H}} = J \, S \sum_{i=1}^{N-2} (\hat{a}_i^+ \, \hat{a}_i + \hat{a}_{i+1}^+ \, \hat{a}_{i-1} - \hat{a}_i \, \hat{a}_{i+1}^+ - \hat{a}_i^+ \, \hat{a}_{i+1}) + J_0 \{ S_0 \, (\hat{a}_1^+ \, \hat{a}_1 + \hat{a}_{N-1}^+ \, \hat{a}_{N-1}) \\ + 2 \, S \, \hat{a}_0^+ \, \hat{a}_0 - \sqrt{S_0 \, S} \, [\hat{a}_0 \, (a_1^+ + \hat{a}_{N-1}^+) + \hat{a}_0^+ \, (\hat{a}_1 + \hat{a}_{N-1})] \} \\ + \mu \, g \, H \sum_{i=1}^{N-1} a_i^+ \, \hat{a}_i + \mu \, g_0 \, H \, \hat{a}_0^+ \, \hat{a}_0 + \sum_{i=1}^{\infty} \hat{\mathcal{H}}_i . \end{aligned} \tag{2}$$

Here, $\hat{\mathcal{H}}_i$ is of order $2\,i + 2$ in boson operators and has matrix elements only between states with at least $i + 1$ boson excitations. The state with no excitations $|0\rangle$ is the one where all spins are fully aligned along the positive z-direction. Its energy is taken as 0. The states $a_i^+ \, |0\rangle$ represent states with a single spin deviation.

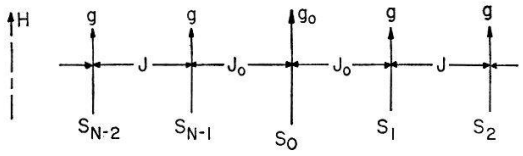

Figure 1

The linear chain Heisenberg ferromagnet with an impurity at the origin. The symbols are defined in the text.

We neglect all $\hat{\mathcal{H}}_i$ ($i = 1, \ldots, \infty$) in Equation (2). If the number of spin deviations excited is small, these terms will make only a small contribution. Our results will be exact for the single spin deviation states, since the $\hat{\mathcal{H}}_i$ have no matrix elements between such states.

Proceeding to diagonalize $\mathcal{H}_0$, the remaining part of $\hat{\mathcal{H}}_i$ we introduce new boson operators $\hat{b}_j$ by the definition

$$\hat{b}_j = \sum_{i=0}^{N-1} c_{j\,i}\, \hat{a}_i\,, \quad j = 0, \ldots, \quad N-1\,, \tag{3}$$

where the matrix of the $c_{j\,i}$ coefficients must be unitary in order that $\hat{b}_j$ satisfy the boson commutation rules. We wish to determine the $c_{j\,i}$ such that $\hat{\mathcal{H}}_0$ takes on the form $\hat{\mathcal{H}}_0 = \Sigma_j E_j\, \hat{b}_j^+\, \hat{b}_j$. Utilizing the equations of motion for the operators $\hat{b}_j$, we require that

$$[\hat{b}_j, \hat{\mathcal{H}}_0] = E\, \hat{b}_j\,, \quad j = 0, \ldots, \quad N-1\,. \tag{4}$$

In contrast to the conventional notation, we define matrix elements through the commutator relationships as

$$[\hat{a}_i, \hat{\mathcal{H}}_0] = \sum_{j=0}^{N-1} \mathcal{H}_{0\,ij}\, \hat{a}_j\,, \quad i = 0, \ldots, N-1\,. \tag{5}$$

Equations (2), (3), (4) and (5) then lead to the following set of N homogeneous equations in the N unknowns $c_{j\,i}$ for each value of the free index j,

$$\sum_{i=0}^{N-1} (\mathcal{H}_{0\,i\,l} - E\, \delta_{i\,l})\, c_{j\,i} = 0\,, \quad j = 0, \ldots, N-1\,. \tag{6}$$

Since the impurity only interacts with its nearest neighbors, the following Green function technique will reduce the N Equations (6) to three. First we rewrite $\mathcal{H}_{0\,i\,l}$ in the form

$$\mathcal{H}_{0\,i\,l} = \mathcal{H}^0_{i\,l} + \mathcal{V}_{i\,l}\,, \tag{7}$$

where $\mathcal{H}^0_{i\,l}$ are matrix elements of the Hamiltonian of the perfect chain (impurity replaced by host). Explicitly, we have

$$\mathcal{H}^0_{i\,l} = (2\,J\,S + \mu\,g\,H)\,\delta_{i\,l} - J\,S\,(\delta_{i,\,l+1} + \delta_{i,\,l-1})\,, \tag{8}$$

and

$$\mathcal{V}_{i\,l} = [2\,S\,(J_0 - J) + \mu\,(g_0 - g)\,H]\,\delta_{i0}\,\delta_{l0} + (J_0\,S_0 - J\,S)\,(\delta_{i1}\,\delta_{l1} + \delta_{i,n-1}\,\delta_{l,n-1})$$
$$+ (J\,S - J_0\sqrt{S_0\,S})\,[\delta_{i0}\,(\delta_{l1} + \delta_{l\,n-1}) + \delta_{l0}\,(\delta_{i1} + \delta_{i,n-1})]\,. \tag{9}$$

The resolvent $\hat{\mathcal{G}}^0 = (E - \hat{\mathcal{H}}^0)^{-1}$ of the perfect chain has the matrix elements (as defined by Equation (5))

$$\mathcal{G}^0_{m\,n}(E) = \frac{1}{N} \sum_{k}^{Bz\,1} e^{ik(m-n)}\, [E - 2\,J\,S\,(1 - \cos k) - \mu\,g\,H]^{-1}\,, \tag{10}$$

between one spin deviation states, where k ranges over the first Brillouin zone ($-\pi \leqslant k < \pi$) in the reciprocal lattice. By definition of the Green function above,

$$\sum_{l=0}^{N-1} \mathcal{G}^0_{m\,l}(E)\, (E\,\delta_{l\,i} - \mathcal{H}^0_{l\,i}) = \delta_{m\,i}\,. \tag{11}$$

Using $\mathcal{H}^0_{i\,l} = \mathcal{H}^0_{l\,i}$ and $\mathcal{V}_{i\,l} = \mathcal{V}_{l\,i}$, we multiply Equation (6) by $\mathcal{G}^0_{m\,l}(E)$ and sum over l, to obtain

$$c_{j\,m} = \sum_{l,\,i}{}' \mathcal{G}^0_{m\,l}(E)\, \mathcal{V}_{l\,i}\, c_{j\,i}\,, \quad j, m = 0, \ldots, N-1\,, \tag{12}$$

where the prime indicates that l and i need range only over $N-1$, 0 and 1, since $\mathcal{V}_{l\,i}$ is zero otherwise. Equation (12) gives all $c_{j\,m}$ in terms of those for the impurity site and its neighbors. Thus, we have only to solve the 3×3 determinant,

$$\left|\delta_{m\,i} - \sum_{l}{}' \mathcal{G}^0_{m\,l}(E)\, \mathcal{V}_{l\,i}\right| = 0\,, \tag{13}$$

which will supposedly have N solutions E_j, counting degeneracies.

III. The Single Spin Deviation Solutions

It is convenient to express the equations in terms of the dimensionless parameters

$$\left.\begin{aligned} &\xi = -J/J_0\,, \quad \gamma = S_0/S\,, \quad h = \mu\, H/2JS \\ \text{and} \qquad &\\ &\eta = (g_0 - g)\, h\,, \quad \varepsilon = E/2JS\,, \quad \varepsilon' = \varepsilon - g\, h \end{aligned}\right\} \tag{14}$$

Also, the dimensionless Green-function $G_{m-n}(\varepsilon') = 2\,J\,S\,\mathcal{G}_{m\,n}(E)$ will be used. The determinant, Equation (13) splits into two factors, giving rise to symmetric (S) and antisymmetric (A) solutions with respect to the plane of symmetry of the chain, located at the site 0.

a) *Symmetric Solutions*

The energies are roots of the equation

$$[\xi + (1 + \gamma\,\xi)\,(\varepsilon' - \eta)]\, f^{-1}(\varepsilon') = G_0(\varepsilon')\,, \tag{15}$$

with

$$f(\varepsilon') = \eta\,\gamma\,\xi + [(1-\gamma)\,\xi - (1+\gamma\,\xi)\,\eta]\,\varepsilon' + (1+\gamma\,\xi)\,\varepsilon'^2\,. \tag{16}$$

The coefficients (for $m \neq 0$) are given by

$$c^S_{j\,m} = f(\varepsilon'^S_j)\; G_m(\varepsilon'^S_j)\; c^S_{j\,0}/\xi\sqrt{\gamma}\,, \tag{17}$$

where $c^S_{j\,0}$ is determined by unitarity. We recall that the coefficients $c_{j\,m}$ represent the probability amplitude for a unit spin deviation to be found at site m when the chain is in its eigenstate of energy ε_j.

We note that the wave function is symmetrical, so that

$$c^S_{j\,m} = c^S_{j,\,N-m}\,. \tag{18}$$

b) *Antisymmetric Solutions*

The energies are obtained from

$$\frac{(1+\gamma\,\xi)\,\varepsilon' - \gamma\,\xi}{(1+\gamma\,\xi)\,\varepsilon'\,(\varepsilon'-2)} = G_0(\varepsilon')\,. \tag{19}$$

Since for the antisymmetric solutions

$$c^A_{l\,0} = c^A_{l,\,N/2} = 0\,, \tag{20}$$

on account of the relation

$$c^A_{l\,m} = -\,c^A_{l,\,N-m}\,, \tag{21}$$

the coefficients are expressed in terms of $c^A_{l,1}$, for $m \neq 0$ or 1, as

$$c^A_{lm} = \frac{1}{2}(1+\gamma\xi)\,[G_{m+1}(\varepsilon'^A_l) - G_{m-1}(\varepsilon'^A_l)]\,c^A_{l,1}\,. \tag{22}$$

The eigenvalues of $\hat{\mathcal{H}}^0$ are doubly degenerate at every value of k, except at 0 and π, since the excitation energy for $-k$ is the same as for k.

As may be seen from the root diagram, Figure 2a, this degeneracy is totally lifted by the impurity: There are $(1/2)\,N$ symmetric and $(1/2)\,N-1$ antisymmetric solutions. The missing solution is the state in which each spin deviates by $1/N$ from the z-axis, i.e. the state of zero wavenumber k.

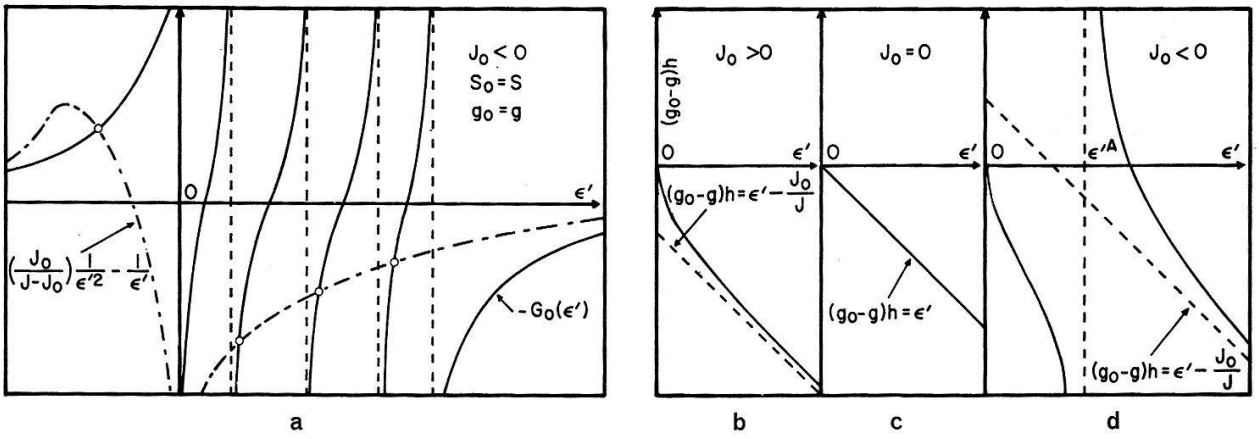

Figure 2

Schematic diagrams of the excitation energies for the symmetric states of the linear chain Heisenberg ferromagnet with one impurity spin and an infinite number of host spins. In Figure 2a, only 5 of the infinite set of poles of the Green function $G_0(\varepsilon')$ in the spin wave band are shown. The excitation energies (open circles) are solutions of the equation $J_0/(J-J_0)\,\varepsilon'^2 - 1/\varepsilon' = -G_0(\varepsilon')$. In Figures 2b, 2c and 2d, $(g_0-g)\,h$ is shown as a function of ε' for representative values of S_0/S and J_0/J. In Figure 2d, ε'^A is the excitation energy for the antisymmetric states and has the value $-J_0^2\,S_0^2/2JS\,(JS-J_0\,S_0)$. The energy ε' is negative (decreases to the right) and $\varepsilon' = \varepsilon - g\,h$. Thus ε' is shifted from the true excitation energy ε if a field is present. The symbols are defined in Figure 3.

Within the spin wave band, the wave function coefficients are all of the order $N^{-1/2}$, but oscillate from site to site, very much like the standing spin waves of the unperturbed spin system. However, although of the order $N^{-1/2}$, c_0 or c_1 may become large relative to the other coefficients for certain ranges of the parameters, giving rise to virtual states which manifest themselves as peaks in the density of the spin wave states in the band [6].

IV. Impurity States Below the Spin Wave Band

For certain values of the parameters, solutions exist with energy below the spin wave band. The form of the Green function in this region is found by direct integration [10] for the limit $N \to \infty$ to be

$$G_m(\varepsilon') = -\alpha^\beta/\sqrt{\varepsilon'\,(\varepsilon'-2)}\,, \tag{23}$$

where

$$\alpha = 1 - \varepsilon' - \sqrt{\varepsilon'(\varepsilon' - 2)} \quad \text{and} \quad \beta = \begin{cases} m & \text{if } m \leqslant N/2, \\ N - m & \text{if } m > N/2. \end{cases} \tag{24}$$

Note that $0 < \alpha < 1$, and as ε' varies from 0 to $-\infty$, α varies from 1 to 0. Equations (23) and (17) show that the spin deviations decrease exponentially with distance from the impurity. The range of localization decreases with increasing $|\varepsilon'|$.

a) *Symmetric Solutions*

Inserting Equation (23) into Equation (15), the equation for ε' becomes cubic but reduces to quadratic if $\eta = 0$ or $\gamma\,\xi = -1$. If $\eta = 0$, we have one solution $\varepsilon' < 0$. If $\gamma = 1$ and $\xi = -1$, the root is given by

$$\varepsilon'^{S} = 1 - \sqrt{1 + \eta^2}, \quad \eta < 0. \tag{25}$$

The schematic root diagram in Figure 2a for $\eta = 0$ and $\gamma = 1$ shows how the states in the upper portion of the band 'press' downward as the impurity exchange J_0 decreases from J, 'pushing' a localized mode out of the bottom of the band as the interaction becomes antiferromagnetic.

For general values of η we do not present the equations, but plot η against ε'^{S} for fixed values of γ and ξ in Figures 2b, c and d. Also plotted is the line $\eta = \varepsilon'^{S} + \xi$, to which Equation (15) is asymptotic as $\varepsilon'^{S} \to -\infty$. If $\xi < 0$, we find a solution below the band, if $\eta < 0$. If $\xi = 0$, no solutions of this form exist, since then there is no interaction coupling the impurity spin to the rest of the lattice. If $\xi > 0$, we find 2 solutions below the band, if $\eta < 0$, one of which is always lower than the other, separated by a pole of Equation (15) at ε'^{A}, the antisymmetric solution.

The symmetric wave function coefficients for $\varepsilon' < 0$ become

$$c_m^S = -f\,\alpha^\beta\, c_0^S/\xi \sqrt{\gamma\,\varepsilon'(\varepsilon' - 2)}\,\Big|_{\varepsilon' = \varepsilon'^S}, \quad m \neq 0, \tag{26}$$

These coefficients are represented in Figures 3a and b for particular values of the parameters. For $\xi > 0$ we notice two striking features. First, c_0 has the opposite sign to all other coefficients, and second, for $\eta \leqslant 0$, c_0 varies from 1 to some finite limit as ξ varies from 0 to ∞. This decrease in c_0 as ξ increases is in contrast to the results of WANG and CALLEN [8] for the three dimensional simple cubic lattice, for which c_0 increases from 0 to a finite limit. In both cases, the limit as $\xi \to \infty$ can be understood in terms of the states of an isolated system consisting only of the impurity and its nearest neighbors. In our case, for instance, the actual limits are $|c_0|^{-2} \to 1 + \gamma/2$, $c_1/c_0 \to -\sqrt{\gamma}/2$ and $c_i \to 0$, $i > 1$, which, for a single spin deviation, agree with a calculation of the ground state of a system of 3 spins of magnitude 1/2 each for $\gamma = 1$. But the fact that $c_0 \to 1$ at the bottom edge of the spin wave band for $\eta \leqslant 0$ and $\xi > 0$ appears to be peculiar to the one dimensional case. This particular solution also is unstable for $\eta \approx 0$ near the band edge, since, as Figure 2a shows, if g_0 is increased infinitesimally greater than g, the entire character of the solution changes.

The higher symmetric energy root varies from 0 to ε'^{A} as η varies from 0 to $-\infty$, while c_0 varies from order $N^{-1/2}$ to a maximum to 0, c_1 varies from order $N^{-1/2}$ to a maximum, and the other coefficients vary from order $N^{-1/2}$ to finite values, decreasing faster with distance from the impurity as $\eta \to -\infty$.

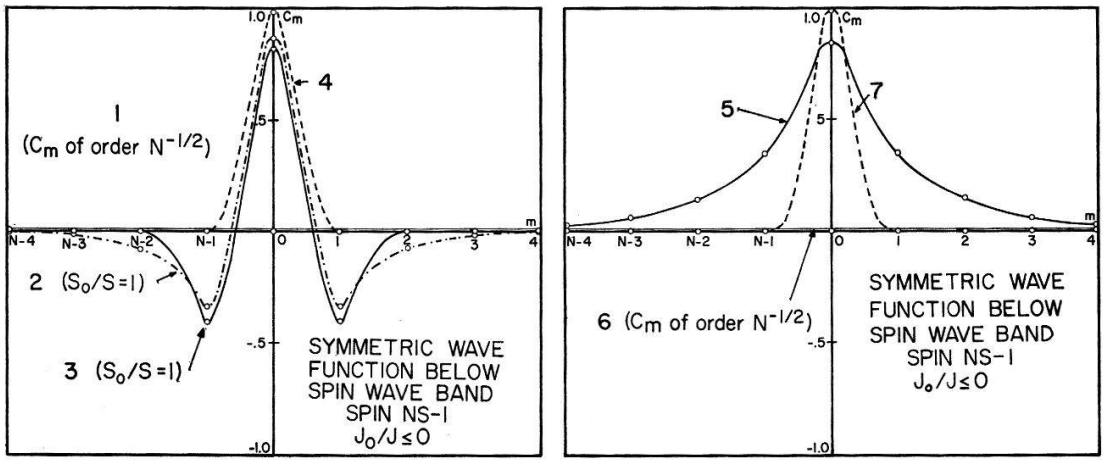

Figure 3

Symmetric localized wave functions of the linear chain Heisenberg ferromagnet with a magnetically coupled impurity. The host and impurity have exchange constants J and J_0, spins S and S_0, and Landé-factors g and g_0, respectively. The energy ε is in units of $2JS$ measured from the energy of the fully aligned state and $h = \mu H/2JS$, where μ = Bohr magneton and H = magnetic field. The different curves are obtained with the parameter values listed in the Table.

Table

Curve No.	J_0/J	$(g_0-g)\,h$	ε
1	$\to 0_-$	>0	$\to 0_- + g\,h$
2	-1	0	$-1.3904 + g\,h$
3	$-\infty$	$>-\infty$ and $<\infty$	$\to -\infty + g\,h$
4	$\to 0_-$	$>-\infty$ and <0	$\to 0_- + g\,h$
	$>-\infty$ and <0	$\to -\infty$	$\to -\infty + g\,h$
5	1	-1	$-0.4142 + g\,h$
6	>0	$\to 0_-$	$\to 0_- + g\,h$
	$\to\infty$	$>-\infty$ and <0	$\to 0_- + g\,h$
7	>0 and $<\infty$	$\to -\infty$	$\to\infty + g\,h$
	$\to 0_+$	<0	$\to\infty + g\,h$

b) *Antisymmetric Solutions*

There exists only one antisymmetric solution below the spin wave band, with energy

$$\varepsilon'^{A} = -\gamma^2\,\xi^2/2\,(1+\gamma\,\xi)\,, \quad \xi > 0\,.$$

No such solutions exist for $\xi < 0$. The corresponding spin deviation at site m is found from the coefficients

$$c_m^A = \alpha^{\beta-1}\sqrt{(1-\alpha^2)/2} \qquad m \geqslant 1\,,$$

where

$$\alpha = 1 + \gamma\,\xi\,.$$

These coefficients become equal and of order $N^{-1/2}$ as the energy approaches the bottom of the spin wave band ($\xi \to 0$), thus becoming delocalized. Also, the coefficients

tend to zero as $\varepsilon' \to -\infty$ ($\xi \to \infty$), except for c_1, which becomes 1. Of course, the antisymmetric solution is independent of η.

V. The Phase Transition and the New Ground State

If the parameters describing the system are such that the excitation energy ε is greater than 0, the fully aligned state is the ground state. At such parameter values that $\varepsilon = 0$, there is a sharp transition to another ground state, which has one or more spin deviations as ε becomes negative. The condition for this transition is gained by the substitution $\varepsilon' = -g\,h$ in Equation (15). If we fix γ and ξ, then Figures 2b, c and d become phase diagrams describing this transition as a function of η and $g\,h$.

Above the solid line, the ground state is the fully aligned state, but the solid line below the pole in Figure 2d should in this context be disregarded, since it corresponds to a state which is always above the lowest energy state below the band. Thus the ground state always has at least one spin deviation below the pole. The line $\eta = \xi - g\,h$ in these figures describes the transition of the corresponding classical spin system from the fully aligned ground state to the ground state with the impurity spin down and all others up.

Since the excitation energy is negative, linear spin wave theory yields the result that everywhere below the solid lines of Figures 2a, b and c, the ground state will have the maximum number of deviations consistent with the restriction that no single spin is deviated more than twice its value. Therefore, we obtain classical behavior for large η and $g\,h$.

When interactions between the states of spin deviations larger than one are taken into account by the variational calculation referred to in the introduction, we expect that the solid lines of Figures 2b, c and d will describe the transition to the ground state having one spin deviation. Also, we expect that new lines below these lines will appear, which will describe the transitions to a ground state of higher spin deviation, as the parameters of the chain are varied.

VI. Acknowledgements

Our sincere thanks are due to Drs. J. JANAK and T. D. SCHULTZ for illuminating discussions and helpful comments.

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