Zeitschrift: Helvetica Physica Acta
Band: 49 (1976)
Heft: 3

Artikel: Multiple excitations in an impure infinite-chain Heisenberg ferromagnet
Autor: Rhodes, Edgar A. / Erdös, Paul
DOI: https://doi.org/10.5169/seals-114771

Helvetica Physica Acta, Vol. 49 (1976), Birkhauser Verlag, Basel

Multiple Excitations in an Impure Infinite-Chain Heisenberg Ferromagnet

by **Edgar A. Rhodes**

School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332[1])

and **Paul Erdös**

Department of Physics, The Florida State University, Tallahassee, Florida 32306[2])

(20. X. 1975)

Abstract. The exact single spin deviation eigenstates (measured from full alignment along an applied field) of the infinite chain Heisenberg ferromagnet containing a substituted magnetically coupled impurity are found and studied in detail, allowing arbitrary host and impurity spins, Landé factors, exchange constants and uniaxial anistropy constants. Depending on the sign and magnitude of impurity–host exchange and the impurity parameters, a number of localized spin deviations are found above and below the spin wave band, and resonant states are found within the band. Using an expansion in terms of these eigenstates, an approximation scheme is formulated for localized double spin deviations. For certain ranges of parameter values, the ground state is found to contain one spin deviation. For antiferromagnetic impurity–host exchange, the ground state can contain two spin deviations, and the conditions for a metamagnetic transition to the two spin deviation ground state are found.

I. Introduction

The magnon excitations of a magnetic insulator, and even the ground state, can be considerably modified by the introduction of different magnetic or non-magnetic atoms into the lattice. Both the experimental and the theoretical aspects of this problem have been discussed in review articles [1–4].

If the impurity–host exchange is quite different from the host–host exchange, as is usually the case, perturbation theory gives no convergent results. However, if the impurity–host interaction is of short range, as is usually also the case, the single spin deviation eigenstates may be found, for a low concentration of impurities, by a Green function technique of Lifshitz [5] in which the non-translationally invariant part of the Hamiltonian is partitioned off to yield an eigenvalue problem in terms of the pure host Green function for single spin deviations.

Wolfram and Callaway [6] applied the technique to the single spin deviation spectrum of a Heisenberg ferromagnet containing a single substituted impurity with ferromagnetic impurity–host exchange, and found resonances near the bottom of the

---

[1]) Present address: Argonne National Laboratory, Argonne, Il 60439, USA.

[2]) Present address: Institut de Physique Théorique, Université de Lausanne, 1015 Lausanne, Switzerland.

spin wave band for impurity–host exchange small compared to the host–host exchange. They also found spin deviations localized about the impurity above the band for impurity–host exchange large in comparison with the host–host exchange. Ishii, Kanamori and Nakamura [7], and Izyumov and Medvedev [8] used this technique for an antiferromagnetically coupled impurity and found localized states below the spin wave band. However, their results are only approximate, since they used linear spin wave theory and chose, as the state of no excitations, the '*Néel state*' (impurity spin down, host spins up), which is not an eigenstate of the Hamiltonian. Wang and Callen [9] pointed this out and obtained the lowest lying exact single spin deviation eigenstate for a spin $\frac{1}{2}$ antiferromagnetic impurity by choosing the fully aligned state as the state of no excitations, while Parkinson [10] noted that the results of Wolfram and Callaway gave the exact single spin deviation eigenstates for an antiferromagnetic impurity as well.

In a previous paper [11], the authors used the Lifshitz technique to find a general solution for the single spin deviation eigenstates of the infinite chain Heisenberg ferromagnet containing a substituted impurity with arbitrary impurity–host exchange and arbitrary host and impurity spins and Landé factors; in an external magnetic field. By choosing the fully aligned state as the state of no excitations, the exact solution was obtained. However, only the eigenstates below the spin wave band, for antiferromagnetic impurity, were investigated. These eigenstates were also studied by Oguchi and Ono [12–13], for spins $\frac{1}{2}$ and zero field. In the present paper, uniaxial anisotropy is added to the Hamiltonian and all single spin deviation eigenstates are investigated in detail, including those above, below, and within the spin wave band. The low-lying double spin deviation states are also studied. The motivation for this extension of the previous work is the following.

The infinite chain impurity problem is interesting from several standpoints. The effects of impurities are most striking in a one dimensional array, because the impurities cannot be avoided by propagating excitations. The solution can be found in simple analytic form in some cases and is quite amenable to numerical computation. Finally, it has been discovered that some materials behave like infinite chain ferromagnets to a good approximation [14–16].

In an impure ferromagnet, when an impurity of spin greater than $\frac{1}{2}$ is coupled strongly and antiferromagnetically to the host, the ground state may contain multiple spin deviations. The question of what is the ground state energy of such a system, or even the simpler question, of what is the total spin deviation of the ground state, has so far not been answered. In the present paper we obtain answers for this problem, valid under certain conditions to be formulated in the text. In particular, we shall study the transition from the one- to the two-spin deviation ground state which occurs in an external magnetic field.

II. Single Spin Deviation Solution in Terms of the Pure Chain Green Function

The Hamiltonian of the linear chain Heisenberg ferromagnet of N spins with an impurity spin at lattice site 0 and periodic boundary conditions ($\hat{\mathbf{S}}_N = \hat{\mathbf{S}}_0$) is given by

$$\hat{H} = -J\sum_{j=1}^{N-2}\hat{\mathbf{S}}_j\cdot\hat{\mathbf{S}}_{j+1} - J_0\hat{\mathbf{S}}_0\cdot(\hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_{N-1}) - \mu gH\sum_{j=1}^{N-1}\hat{S}_j^z$$
$$-\mu g_0 H\hat{S}_0^z - K\sum_{j=1}^{N-1}(\hat{S}_j^z)^2 - K_0(\hat{S}_0^z)^2 \tag{2.1}$$

where the host ions have spin S, the impurity ion has spin S_0, the host–host and impurity–host exchange constants are J and J_0, the host and impurity Landé factors are g and g_0, the external magnetic field is H, and host and impurity anisotropy constants are K and K_0. All parameters are arbitrary, except that J, g, and g_0 must be positive.

Since $\hat{H}$ commutes with the operator of the z-component of the total spin $\hat{S}^z = \sum_{j=0}^{N-1} \hat{S}_j^z$, the eigenstates of $\hat{H}$ are eigenstates of $\hat{S}^z$, one of which is the fully aligned state, with spin eigenvalue $S^z = (N-1)S + S_0$. Our task is to find the eigenstates of $\hat{H}$ for which $S^z = (N-1)S + S_0 - 1$. States with this value of S^z are called *single spin deviation states*. The states $|j\rangle$ having a unit spin deviation from the fully aligned state at lattice site j form an orthonormal basis of the subspace of spin states with a single spin deviation. The proper linear combinations of these states,

$$|n) = \sum_{j=0}^{N-1} c_{nj}|j\rangle, \tag{2.2}$$

must be found which are the eigenstates $|n)$ of $\hat{H}$. Thus, we must solve the N homogenous equations

$$\sum_{j=0}^{N-1} (H_{lj} - E\delta_{lj})c_{nj} = 0, \qquad l \text{ and } n = 0, \ldots, N-1, \tag{2.3}$$

for c_{nj} and eigenenergy E_n for each value of n, where H_{lj} is the matrix element of H between the states $|l\rangle$ and $|j\rangle$ and δ_{lj} is the Kronecker symbol.

Because the impurity interacts only with its neighbors, the N equations may be reduced to three [5, 6]. First, we partition H_{lj} into

$$H_{lj} = H_{lj}^0 + V_{lj}, \tag{2.4}$$

where H_{lj}^0 are the matrix elements of the translationally invariant Hamiltonian of the pure chain (impurity replaced by host ion). Explicitly,

$$H_{lj}^0 = [E_0 + 2JS + \mu gH + (2S-1)K]\delta_{jl} - JS(\delta_{j,l+1} + \delta_{j,l-1}) \tag{2.5}$$

and

$$\begin{aligned} V_{lj} = {} & [2S(J_0 - J) + \mu(g_0 - g)H + (2S_0 - 1)K_0 - (2S-1)K]\delta_{j0}\delta_{l0} \\ & + (J_0 S_0 - JS)(\delta_{j1}\delta_{l1} + \delta_{j,N-1}\delta_{l,N-1}) + (JS - J_0\sqrt{S_0 S})[\delta_{j0}(\delta_{l1} + \delta_{l,N-1}) \\ & + \delta_{l0}(\delta_{j1} + \delta_{j,N-1})], \end{aligned} \tag{2.6}$$

where all contribution to the energy E_0 of the fully aligned state of $\hat{H}$ has been placed in H_{lj}^0, so that V_{lj} is non-zero only for indices $N-1$, 0, 1.

The resolvent $\hat{G}^0 = (E - \hat{H}^0)^{-1}$ of the pure chain is now introduced. Since the single spin deviation eigenstates of the pure chain are the spin waves $N^{-1/2}\sum_{j=0}^{N-1} e^{ikj}|j\rangle$, the matrix elements of $\hat{G}_0$ are easily found to be

$$\begin{aligned} G_{ml}^0(E) = {} & N^{-1} \\ & \times \sum_k e^{ik(m-l)}[E - E_0 - 2JS(1 - \cos k) - \mu gH - (2S-1)K]^{-1}. \end{aligned} \tag{2.7}$$

Here, the wave number k is given by $k = 2\pi m/N$, where the integer m varies in steps

of one between $-(N-1)/2$ and $(N-1)/2$ if N is odd, and between $-N/2$ and $N/2-1$ if N is even. By definition of the resolvent, we have

$$\sum_{l=0}^{N-1} G^0_{ml}(E)(E\delta_{lj} - H^0_{lj}) = \delta_{mj}. \tag{2.8}$$

We multiply equation (2.3) by the Green function G^0_{ml}, sum over l, and use equations (2.4) and (2.8) to obtain

$$c_{nm} = \sum_{lj} G^0_{ml}(E)V_{lj}c_{nj}, \qquad n \text{ and } m = 0, \ldots, N-1. \tag{2.9}$$

Since equation (2.9) expresses all wave function coefficients in terms of those for the impurity and its neighbors, the eigenenergies E_n are found by solving the 3×3 determinantal equation

$$\det(\delta_{jm} - \sum_{l} G^0_{ml}(E)V_{lj}) = 0, \qquad j = N-1, 0, 1; m = N-1, 0, 1. \tag{2.10}$$

A schematic root diagram illustrating the solution of equation (2.10) may be found in Refs. [11] and [17].

Other quantities of physical interest include the average value of $\hat{S}^z_j$ when the chain is in the eigenstate $|n)$,

$$(n|\hat{S}^z_j|n) = S_j - |c_{nj}|^2, \tag{2.11}$$

and the transverse correlation function for sites l and j,

$$(n|\hat{S}^\alpha_j\hat{S}^\alpha_l|n) = \sqrt{S_lS_j}\,\mathrm{Re}(c^*_{nl}c_{nj}), \qquad \alpha = x \text{ or } y, j \neq l. \tag{2.12}$$

From equation (2.11), we see that $|c_{nj}|^2$ is the spin deviation at site j in state $|n)$.

It is convenient to introduce the dimensionless parameters

$$\begin{aligned} &\xi = -J_0/J, \quad \gamma = S_0/S, \quad h = \mu H/2JS, \quad \kappa = K/2JS, \\ &\phi = K_0/2JS, \quad \eta = (g_0 - g)h + (2\gamma S - 1)\phi - (2S-1)\kappa, \quad \beta = 1 + \gamma\xi, \\ &\epsilon = (E - E_0)/2JS, \quad \epsilon' = \epsilon - gh - (2S-1)\kappa, \quad G_{m-l}(\epsilon') = 2JSG^0_{ml}(E). \end{aligned} \tag{2.13}$$

The periodic boundary condition dictates that $G_{N-m}(\epsilon') = G_m(\epsilon')$ and every Green function may be expressed in terms of $G_0(\epsilon')$ by using equations (2.5) and (2.8). Equation (2.10) factors into

$$f_S(\epsilon')f_A(\epsilon') = 0, \tag{2.14}$$

where

$$f_S(\epsilon') = \sigma(\epsilon') - [\lambda(\epsilon') - (1-\epsilon')\sigma(\epsilon')]G_0(\epsilon') \tag{2.15}$$

and

$$f_A(\epsilon') = 1 - \beta(1-\epsilon') - \beta\epsilon'(\epsilon' - 2)G_0(\epsilon'). \tag{2.16}$$

In equation (2.15),

$$\lambda(\epsilon') = [1 + (\epsilon' - \eta)/\xi]/\sqrt{\gamma}, \qquad \sigma(\epsilon') = [1 + \beta(\epsilon' - \eta)/\xi]/\sqrt{\gamma}. \tag{2.17}$$

First we consider the energy eigenvalues given by

$$f_S(\epsilon') = 0, \tag{2.18}$$

which will be denoted ϵ'_{Sj}. From equations (2.9) and (2.18), we find that $c_{Sj,N-1} = c_{Sj,1}$, that

$$c_{Sj,1} = \lambda(\epsilon'_{Sj})c_{Sj,0}, \tag{2.19}$$

and finally that

$$c_{Sj,m} = [\lambda(\epsilon'_{Sj}) - (1 - \epsilon'_{Sj})\sigma(\epsilon'_{Sj})]G_m(\epsilon'_{Sj})c_{Sj,0}, \qquad m \neq 0 \text{ or } N, \tag{2.20}$$

where $c_{Sj,0}$ is determined by the unitarity condition

$$\sum_{m=0}^{N-1} c_{n,m}c_{n,m} = 1. \tag{2.21}$$

with $n = Sj$.

From equation (2.20), it is seen that $c_{Sj,N-m} = c_{Sj,m}$, so that the wave function is symmetric about the impurity site.

Now consider the eigenvalues given by

$$f_A(\epsilon') = 0, \tag{2.22}$$

denoted ϵ'_{Aj}. From equations (2.9) and (2.22), we find that $c_{Al,0} = 0$, $c_{Al,N-1} = -c_{Al,1}$, and that

$$c_{Al,m} = \tfrac{1}{2}\beta[G_{m+1}(\epsilon'_{Al}) - G_{m-1}(\epsilon'_{Al})]c_{Al,1}, \qquad 1 < m < N - 1, \tag{2.23}$$

where $c_{Al,1}$ is determined by equation (2.21). It is seen that $c_{Al,N-m} = -c_{Al,m}$, so that the wave function is antisymmetric about the impurity site. The antisymmetric solutions are independent of the impurity single ion parameter η since $c_{Al,0} = 0$, as can be seen from equations (2.16) and (2.23). This means that neither the Landé factor nor the anisotropy constant of the impurity influence the behavior of the antisymmetric solutions.

III. States Below the Spin Wave Band

For certain ranges of parameter values, eigenstates will exist with energies below the spin wave band ($\epsilon' < 0$). In this region, the Green function has no poles and is found by integration of equation (2.7) to have the form

$$G_m(\epsilon') = -\alpha(\epsilon')^{\bar{m}}/\sqrt{\epsilon'(\epsilon' - 2)}, \tag{3.1}$$

in the limit $N \to \infty$, where

$$\alpha(\epsilon') = 1 - \epsilon' - \sqrt{\epsilon'(\epsilon' - 2)}, \qquad \bar{m} = \begin{cases} m & \text{for } m \leqslant N/2 \\ N - m & \text{for } m > N/2. \end{cases} \tag{3.2}$$

Since $\epsilon' < 0$, we have $0 < \alpha < 1$, $\lim(\epsilon' \to 0_-)\alpha = 1_-$ and $\lim(\epsilon' \to -\infty)\alpha = 0_+$. Thus, equations (2.20), (2.23), and (3.1) show that the spin deviations decrease exponentially with distance from the impurity and that the decay is greater when ϵ' is farther below the spin wave band.

First, we examine the antisymmetric solutions. Substituting equation (3.1) into equation (2.22), we find the eigenenergy,

$$\epsilon'_{A1} = -(\beta - 1)^2/2\beta, \qquad \beta > 1. \tag{3.3}$$

There is no antisymmetric state below the band for a ferromagnetic impurity ($\beta < 1$). From equations (2.23), (3.1), and (3.3), the antisymmetric wave function coefficient $c_{A1,m}$ at site m is found. These coefficients become equal in magnitude and of order $N^{-1/2}$ as the energy approaches the bottom of the spin wave band ($\beta \to 1_+$). As $\epsilon'_{A1} \to -\infty$, i.e. $\beta \to \infty$, the coefficients approach zero, except that $c_{A1,1} \to 1$.

Now we examine the symmetric solutions. Substituting equation (3.1) into equation (2.18), an equation is obtained for the eigenenergies ϵ'_{Sj}. Using equations (2.20), (3.1), and (2.18), we find the corresponding wave function coefficient $c_{Sj,m}$ at site m. The behavior of the energy ϵ'_{Sj} and of the corresponding wave functions is discussed in detail in Refs. [11] and [17], and we give only a summary here. The definitions of η and ϵ in Ref. [11] are generalized in equation (2.13) to include uniaxial anisotropy. A symmetric state $S1$ appears below the band if $\xi > 0$ or if $\eta < 0$. If both $\xi > 0$ and $\eta < 0$, another symmetric state $S2$ appears below the band in addition to $S1$. State $S1$ always has the lowest energy and the energies are ordered as follows: $\epsilon_{S1} < \epsilon_{A1} < \epsilon_{S2}$. For $\eta = 0$ a simple analytic solution is obtained,

$$\epsilon'_{S1} = -\xi\{4 + \gamma\xi(2 + \gamma) + \gamma\sqrt{\xi[8 + \xi(2 + \gamma)^2]}\}/4(1 + \gamma\xi). \qquad \xi > 0, \tag{3.4}$$

For $\gamma\xi = -1$, the solution is

$$\epsilon'_{S1} = \{\eta + \xi[\eta - \xi - \sqrt{(\eta - \xi)^2 - 2\eta}]\}/(1 + 2\xi), \qquad \eta < 0. \tag{3.5}$$

A schematic graph of ϵ'_{S1} and ϵ'_{A1} as functions of ξ is shown in Figure 1 for $\eta = 0$.

For $\xi > 0$, $\eta = 0$, $h = 0$, and $S_0 = \frac{1}{2}$, the ground state of the Hamiltonian is

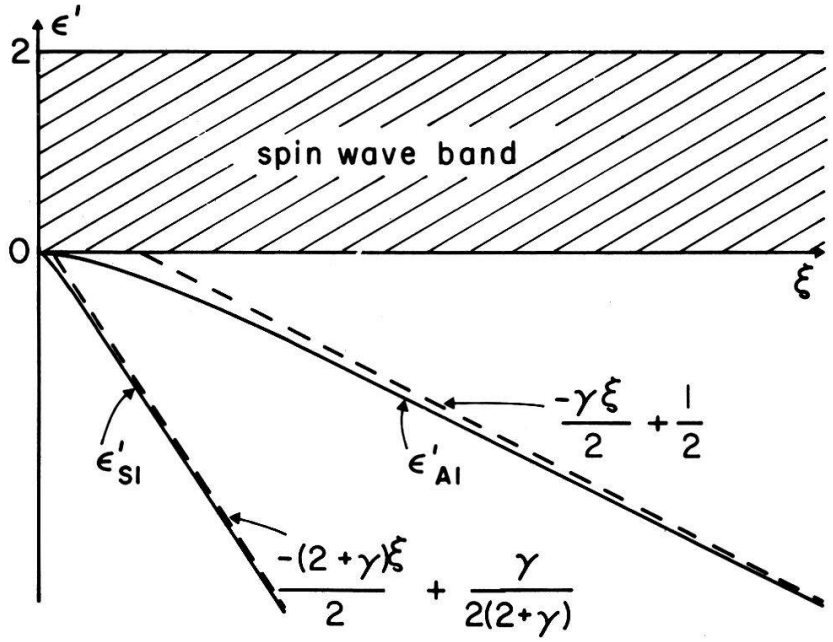

Figure 1
Schematic diagram of the symmetric and antisymmetric single spin deviation eigenenergies ϵ'_{S1} and ϵ'_{A1} below the spin wave band as a function of ξ, for the case $\eta = 0$. Dashed lines are asymptotes. Slopes of the $S1$ and $A1$ curves at $\xi = 0$ are -1 and 0, respectively. The parameters ξ, γ and ϵ' are defined in equation (2.13) in terms of the parameters of the Hamiltonian, equation (2.1).

expected to contain one spin deviation. This state should approach the Néel state in character as $\xi \to 0_+$, because the exchange of the impurity with its neighbors is antiferromagnetic and there is neither external field nor anisotropy present. Indeed, the preceding equations yield for $\xi \to 0_+$: $\epsilon'_{S1} \to 0_-$, $c_{S1,0} \to 1$, and $c_{S1,m} \to 0$ $(m \neq 0)$.

Also, for $\xi \to \infty$, $\eta = 0$, $h = 0$, and $S_0 = \frac{1}{2}$ the ground state should approach the lowest state of the system consisting of an isolated cluster of three spins (the impurity and its two neighbors) with the remaining spins fully aligned. The equations yield for $\xi \to \infty$: $\epsilon'_{S1} \to -\infty$, $c_{S1,0} \to (1 + \frac{1}{2}\gamma)^{-1/2}$, $c_{S1,1} \to -\frac{1}{2}\sqrt{\gamma}\, c_{S1,0}$, and $c_{S1,m} \to 0$ $(m > 1)$, which is the required result.

For $\xi > 0$ and $\eta = 0$, $c_{S1,0}$ is opposite in sign to $c_{S1,m}$, $m \neq 0$, so we see from equation (2.12) that the impurity is negatively correlated to all host spins in state $S1$. As pointed out by Wang and Callen [5], Ishii, Kanamori, and Nakamura [7] do not obtain this characteristic because they choose the 'Néel state' as the state of no excitations, giving an incorrect positive correlation which leads to an energy that is too high.

If the single spin deviation eigenstate $S1$ is the ground state for $\xi > 0$, $\eta = 0$, $h = 0$, and $S_0 = \frac{1}{2}$, then if an external magnetic field is applied, a *metamagnetic transition* to the fully aligned state will occur when $\epsilon = 0$ for some critical field h_c. The fully aligned state will be the ground state for $h > h_c$. Noting the definition of ϵ' in equation (2.13), we see that the critical field may be found by substituting $\epsilon' = -gh_c - (2S - 1)\kappa$ into equation (3.5). If $\kappa = \phi = 0$, a simple analytic solution is obtained for the critical field given by

$$h_c = \xi\{4 + \gamma\xi(2 + \gamma g_0/g) + \gamma\sqrt{\xi[8g_0/g + \xi(2 + \gamma g_0/g)^2]}\}/4g_0(1 + \gamma\xi), \qquad \xi > 0. \tag{3.6}$$

A detailed discussion of this metamagnetic transition, extended to $S_0 > \frac{1}{2}$ and multiple spin deviations in the non-interacting spin wave approximation, appeared in Ref. [11]. The effect of the inclusion of uniaxial anistropy (positive κ and ϕ) and spin wave interactions on the metamagnetic transition is discussed in Ref. [17]. It is found that the ground state is always the fully aligned state for $\xi < 0$ and positive κ and ϕ, and that the ground state contains at least one spin deviation if $gh + (2S - 1)\kappa < -\epsilon'_{A1}$ regardless of the value of η.

Another case in which the ground state should contain one spin deviation is for $S_0 = 1$, $gh + (2S - 1)\kappa > 0$, and infinite impurity easy plane anisotropy $(\phi \to -\infty)$, h remaining finite. In this limit $\eta \to -\infty$, so that the spin deviation in state $S1$ becomes concentrated on the impurity, while $\epsilon'_{S1} \to -\infty$ [11]. Since the host spins are then fully aligned, state $S1$ minimizes the host energy when $gh + (2S - 1)\kappa > 0$, and since the impurity spin lies in the $x - y$ plane for $S_0 = 1$, the impurity anisotropy energy is also minimized. The interaction energy between the impurity and host in this limit is given simply by $(S1|\hat{H}'|S1)$, where $\hat{H}' = -J_0\hat{\mathbf{S}}_0 \cdot (\hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_{N-1})$. All higher order perturbation terms are zero because neither $\hat{H}$ nor $\hat{H}'$ mixes states containing different numbers of spin deviations, so that only single spin deviation states contribute and the energy denominators in the perturbation expansion become infinite. And since $(S1|\hat{H}'|S1) = 0$ in this limit, the impurity becomes uncoupled from the host. One would expect the state $S1$ to be the ground state also for very large but finite impurity easy plane anisotropy and small external field when $S_0 = 1$, as long as the host anisotropy is of the easy axis type and large enough so that the host spin deviation is small.

IV. States Above the Spin Wave Band

For certain ranges of parameter values, eigenstates will exist with energies above the single spin deviation spin wave band ($\epsilon' > 2$). In this region, the Green function is found by integration of equation (2.7) to have the same form as for $\epsilon' < 0$, except for a change in sign of the square root in equations (3.1) and (3.2). However, since $\epsilon' > 2$, we have $-1 < \alpha < 0$, $\lim(\epsilon' \to 2_+)\alpha = -1_+$ and $\lim(\epsilon' \to \infty)\alpha = 0_-$. Thus the spin deviations decrease exponentially with distance from the impurity, just as for $\epsilon' < 0$, and the decay is greater when ϵ' is farther above the band. But, since $\alpha < 0$, spin deviations on neighboring sites are negatively correlated throughout the chain, raising the energy of these states above the band. Equation (3.3) remains correct for the antisymmetric state above the band, except one must have $\beta < -1$. We denote the antisymmetric state above the band as $A2$.

One can easily find η as a function of ϵ' to study the symmetric eigenenergies for $\epsilon' > 2$. Schematic graphs of η vs. ϵ' are shown in Figure 2 for fixed values of γ and ξ

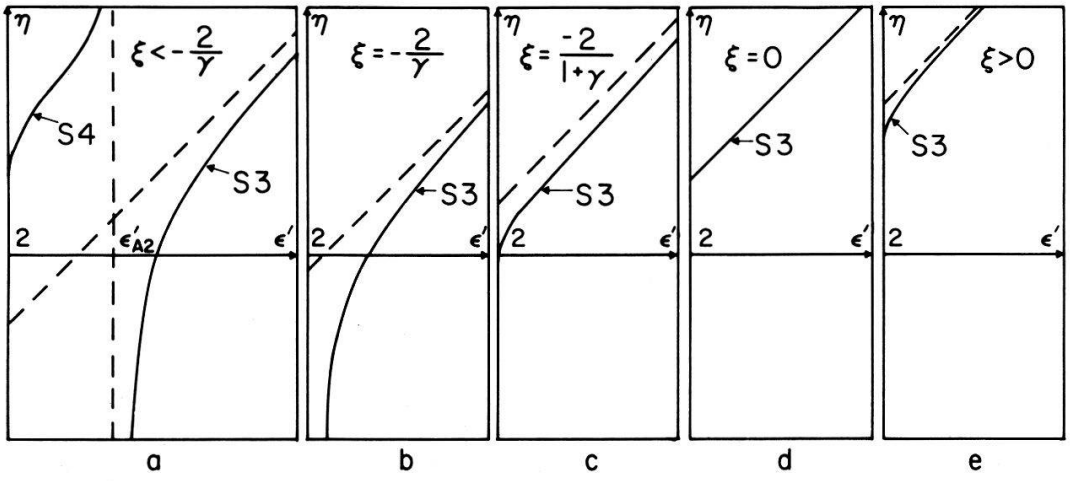

Figure 2
Schematic diagrams of η as a function of ϵ' for the two symmetric single spin deviation eigenenergies $S3$ and $S4$ above the spin wave band. The ϵ' axes begin at 2. The dashed line asymptote in the figures is given by $\eta = \epsilon' + \xi$. At the point $\epsilon' = 2$, $\eta = 2[1 + \xi/(2 + \gamma\xi)]$. The figures a, b, c, d, and e are drawn for different values of ξ. The parameters ξ, γ, η, ϵ' are defined in equation (2.13). ϵ'_{A2} is the energy of the antisymmetric state.

and for pertinent parameter ranges. It is seen that for $\xi > -2/\gamma$, one symmetric state, $S3$, exists above the band, but only if $\eta > 2[1 + \xi(2 + \gamma\xi)^{-1}]$. For $\xi < -2/\gamma$, i.e. $\beta < -1$, state $S3$ exists for all η values, along with state $A2$, and another state, $S4$, appears for $\eta > 2[1 + \xi(2 + \gamma\xi)^{-1}]$. We see that η has a simple pole at the energy ϵ'_{A2} of the antisymmetric state which separates ϵ'_{S3} from ϵ'_{S4}, so that $\epsilon'_{S3} > \epsilon'_{A2} > \epsilon'_{S4}$. A simple analytic solution exists for ϵ'_{S3} for $\eta = 0$, $\xi < -2/(1 + \gamma)$, given by equation (3.4) with a change in the sign of the square root. A schematic diagram of ϵ'_{S3} and ϵ'_{A2} as function of ξ is shown in Figure 3 for $\eta = 0$.

V. States Within the Spin Wave Band

With help of equation (2.7) it may be shown [18] that for energies within the spin wave band ($0 \leqslant \epsilon' \leqslant 2$), G_0 has the form

$$G_0(\theta) = \cot(N\theta/2)/\sin\theta, \qquad N \gg 1, \tag{5.1}$$

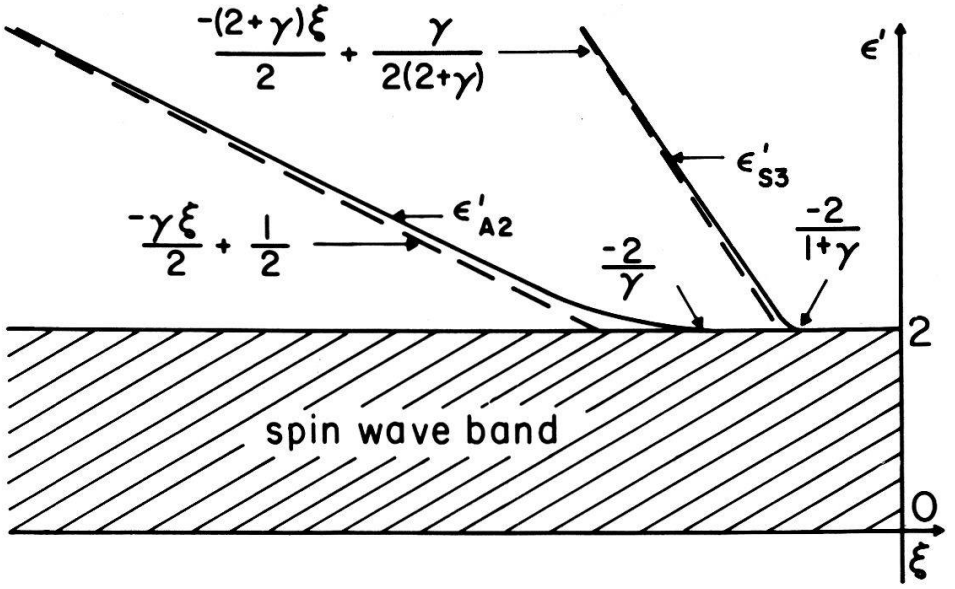

Figure 3
Schematic diagram of the symmetric and antisymmetric single spin deviation eigenenergies ϵ'_{S3} and ϵ'_{A2} above the spin wave band as a function of ξ, for the case $\eta = 0$. Dashed lines are asymptotes. Slopes are zero of the $S3$ and $A2$ curves at $\xi = -2/(1 + \gamma)$ and $\xi = -2/\gamma$, respectively. The parameters ξ, γ and ϵ' are defined in equation (2.13).

where

$$\theta = \text{Arccos}(1 - \epsilon'), \qquad 0 \leqslant \theta \leqslant \pi. \tag{5.2}$$

G_0 has $N/2$ poles. By use of equations (2.5), (2.7), and (2.8), G_m can be shown to have the form

$$G_m(\theta) = G_0(\theta) \cos \bar{m}\theta + \sin \bar{m}\theta/\sin \theta, \qquad m > 0, \tag{5.3}$$

indicating the oscillatory character of the band eigenstates throughout the chain. These Green functions lead to standing spin waves in the lattice, rather than traveling waves, and yield eigenstates having explicit site dependence, rather than wave number dependence. One may also consider the scattering problem [18] by adding a small imaginary part to the energy in equations (2.7), (2.15), and (2.16) and obtain traveling wave solutions.

The form of G_0 given in equation (5.1) is not suitable for calculation. In determining the eigenstates, we eliminate G_0 by use of equations (2.18) and (2.22). Since the eigenenergies form a quasi-continuum and ϵ' becomes an independent variable, the subscripts numbering the band eigenenergies will be suppressed. From equations (2.20) and (5.3), we find the coefficient of the symmetric wave function at site m to be

$$c_{Sm}(\theta) = c_{S0}(\theta)[\sigma \cos \bar{m}\theta + (\lambda - \sigma \cos \theta) \sin \bar{m}\theta/\sin \theta], \qquad m \neq 0, \tag{5.4}$$

where $\sigma = \sigma(\theta)$ and $\lambda = \lambda(\theta)$ as defined by equation (2.17) and c_{S0} is determined by equation (2.21).

All the coefficients are of order $N^{-1/2}$ and the wave functions are basically nonlocal, just as for the spin waves in a pure chain. In the same manner, equation (2.23) yields the coefficients of the antisymmetric wave function $c_{Am}(0)$.

For the low-lying states, the wave function varies like a sine wave of long wavelength, whereas for the high-lying states, the wave function varies like a wave of short

wavelength with an amplitude envelope in the form of a sine wave of long wavelength. The shape and wavelength of the wave functions are independent of the parameters except at the impurity site, although the phase shifts are not. Behavior of the antisymmetric wave functions is similar, except of course $c_{A0} = 0$ and $c_{A,N-m} = -c_{A,m}$.

In Figure 4 the spin deviation at the impurity site for the symmetric states is shown as a function of energy for representative values of ξ, for $\gamma = 1$ and $\eta = 0$. The impurity site spin deviation is of course zero for the antisymmetric states and the impurity neighbor spin deviation for the antisymmetric state does not exhibit the narrow peaks as a function of energy seen in Figure 4 for the symmetric states. We mention here a general feature of the band eigenfunctions: as a parameter approaches a value for which a state leaves the band, the corresponding wave function coefficients at the corresponding band edge approach the values assumed by the state just outside the band, so there does not appear to be any discontinuous change in the wave function.

Since the spin wave band is a quasi-continuum of states, an important quantity of physical interest is the density of states, which can be written [19]

$$R(E) = -\frac{1}{\pi}\lim_{t\to 0}\mathrm{Im}[\mathrm{Tr}(E + it - \hat{H})^{-1}] \tag{5.5}$$

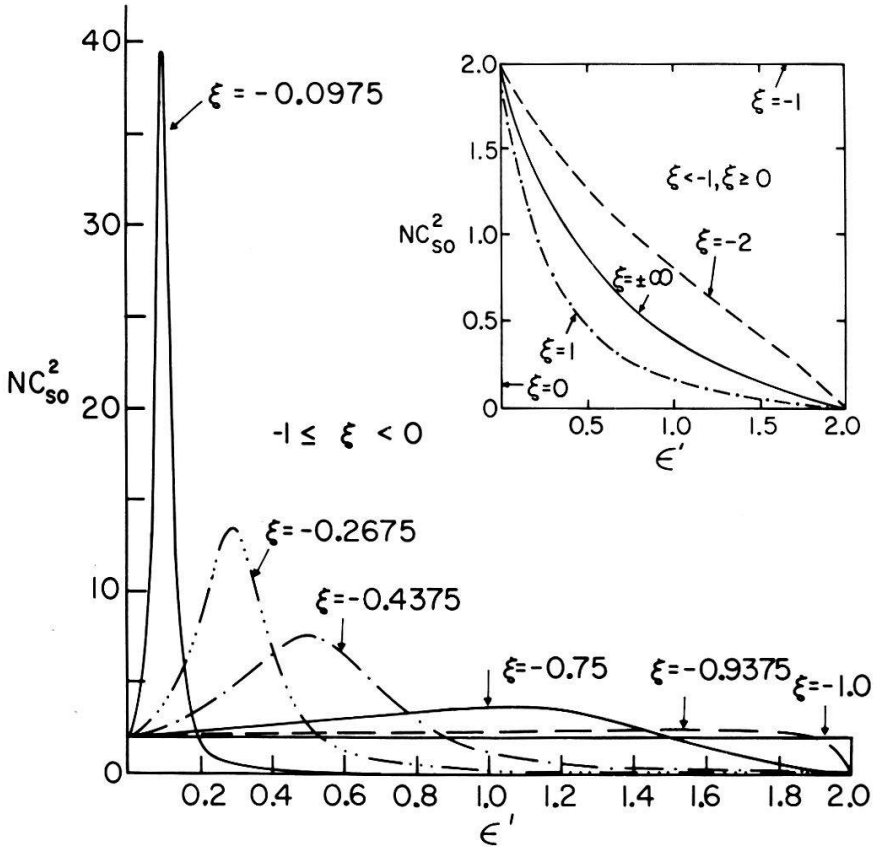

Figure 4

Spin deviation c^2_{s0} (multiplied by the number N of spins in the chain) of the impurity as a function of energy in the spin wave band for symmetric single spin deviation eigenstates, for $\eta = 0$, $\gamma = 1$, and various values of ξ. For $\xi = 0$, spin deviation is 1 at $\epsilon' = 0$, 0 for $\epsilon' > 0$. A symmetric eigenstate exists below the band if $\xi > 0$ and above the band if $\xi < -1$. Nc^2_{s0} for these cases is shown in the insert. The parameters are defined in equation (2.13).

for energy E in the quasi-continuum where Im denotes 'imaginary part' and Tr denotes 'trace'. It is convenient to define the dimensionless density of states

$$\rho(\epsilon') = 2JSR(E), \qquad 0 \leqslant \epsilon' \leqslant 2. \tag{5.6}$$

By use of the same partitioning of the Hamiltonian as indicated in equation (2.4), ρ can be written as [6]

$$\rho(\epsilon') = \rho_0(\epsilon') + \Delta\rho_S(\epsilon') + \Delta\rho_A(\epsilon'), \tag{5.7}$$

where ρ_0 is the density of states for the pure chain and $\Delta\rho_S$ and $\Delta\rho_A$ are the changes due to the symmetric and antisymmetric impurity states. The latter may be found from [6] to be

$$\Delta\rho_l(\epsilon') = -\frac{1}{\pi}\lim_{t\to 0}\operatorname{Im}[f_l^{-1}(\epsilon' + it)\, df_l(\epsilon' + it)/d\epsilon'], \qquad l = S \text{ or } A, \tag{5.8}$$

where f_l is given by equations (2.15) and (2.16). We note that while the factorization in equation (2.14) remains valid for complex ϵ', neither f_S nor f_A is zero.

If $\hat{H}$ is replaced by the pure chain Hamiltonian $\hat{H}^0$, we obtain [17]

$$\rho_0(\epsilon') = N/\pi\sqrt{\epsilon'(2 - \epsilon')}. \tag{5.9}$$

We see that ρ_0 is symmetric about the band center $\epsilon' = 1$ and approaches infinity at the band edges $\epsilon' = 0$ and $\epsilon' = 2$. There are no single spin deviation states outside this band for the pure chain, and the integral of ρ_0 over the band is N, the total number of single spin deviation states.

From equations (2.15), (2.16), and G_0 we may determine $\Delta\rho_S$ and $\Delta\rho_A$. In terms of the variable θ introduced in equation (5.2), we have

$$\begin{aligned}\Delta\rho_S = \rho_0\{&\beta\lambda \sin^2\theta + [(1 + \xi + \beta - \eta)(\cos\theta - 1) \\ &+ \eta(\beta - 1)]\sigma\}/N\sqrt{\gamma\xi}(\lambda^2 - 2\lambda\sigma\cos\theta + \sigma^2)\end{aligned} \tag{5.10}$$

and

$$\Delta\rho_A = \rho_0\beta(\cos\theta - \beta)/N(\beta^2 - 2\beta\cos\theta + 1), \tag{5.11}$$

where $\lambda = \lambda(\theta)$ and $\sigma = \sigma(\theta)$ are defined by equation (2.17) and $\rho_0 = \rho_0(\theta) = N/\pi \sin\theta$. We see that $\Delta\rho_S$ and $\Delta\rho_A$ are of order N^{-1} compared to ρ_0, as might be expected, since the band eigenenergies of the chain containing a single impurity differ from those of the pure chain only by a quantity of order N^{-1} [11, 17], and since only a few eigenstates can exist outside the band. $\Delta\rho_S$ and $\Delta\rho_A$ have the characteristic energy dependence of ρ_0, multiplied by a factor dependent on the impurity parameters, and are zero for the pure chain.

Figure 5 displays $\Delta\rho_S$ as a function of energy for the same parameter ranges as used in Figure 4. The ordinates have been divided by ρ_0 in order to exhibit the structure due to the impurity. A general feature of the figures is an increase in the density of states in the neighborhood of the corresponding band edge when a state is about to leave the band, followed by a depletion in the density of states when the state is outside the band. As the impurity–host interactions become appreciably different from the host–host interactions, these increases become resonances (relatively large, narrow peaks) which move through the band as the parameters change. The actual positions of these resonances are shifted, from those shown in the figures, toward the nearest

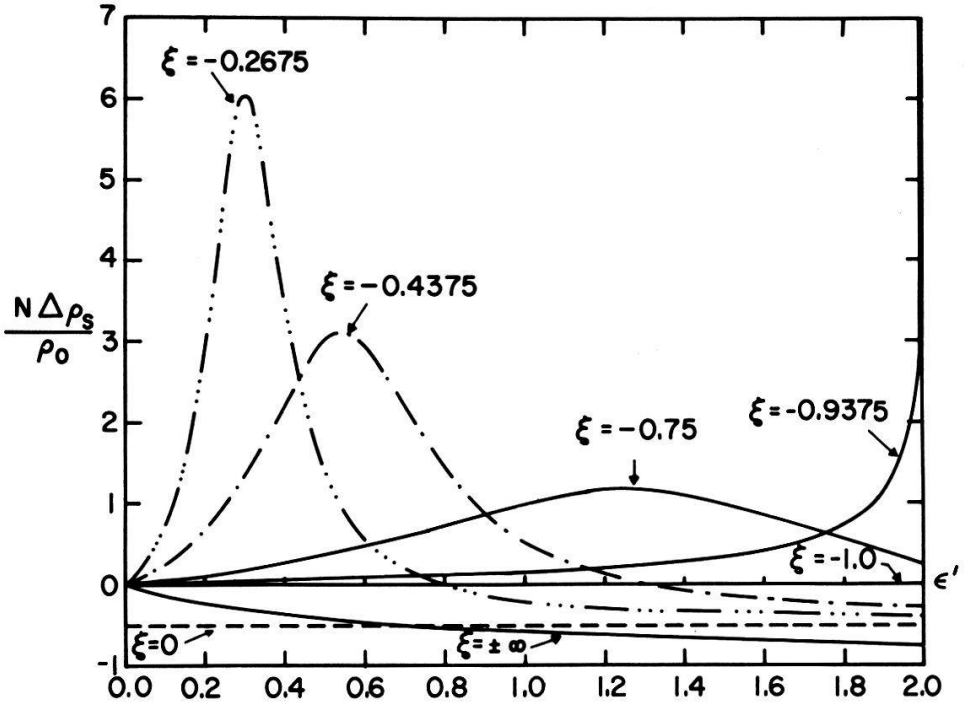

Figure 5
Plot of $N\Delta\rho_s/\rho_0$ as a function of ϵ', for $\gamma = 1$, $\eta = 0$ and various values of ξ. $\Delta\rho_s$ is the change in density of states of the spin wave band due to the impurity for symmetric single spin deviation eigenstates. The density of states of the pure chain is $\rho_0 = N/\pi\sqrt{\epsilon'(2 - \epsilon')}$. N is the number of spins in the chain, and the parameters are defined in equation (2.13).

band edge by the strong increase in ρ_0 away from the center, and their shapes are somewhat distorted. The peak position and width of these resonances, if calculated by the usual resonance formulas [6], would be inaccurate for energies not near the band center, due to this strong variation of ρ_0 with energy. Comparison of Figure 4 depicting $\Delta\rho_S$ with Figure 5 showing c_{S0}^2 reveals a strong correlation between these two quantities. In particular, resonances in the density of states are associated with peaks in the spin deviation at the impurity site as a function of energy. Unlike $\Delta\rho_S$, $\Delta\rho_A$ exhibits no resonances, as might be expected, since $c_{A0} = 0$ and c_{A1}^2 has no narrow peaks as a function of energy.

VI. Conclusions Regarding the One Spin Deviation Eigenstates

In the preceding sections, we have found and studied in detail the exact single spin deviation eigenstates of the infinite chain Heisenberg ferromagnet containing a substituted magnetically coupled impurity. Depending on the values of the impurity parameters, a number of localized states were found above and below the spin wave band, and resonant states were found within the band. For easy axis host anisotropy and small external field, the ground state was found to contain one spin deviation in two cases: (1) impurity spin $\frac{1}{2}$, antiferromagnetic impurity–host exchange, small impurity and host anistropy; (2) impurity spin 1, very large easy plane impurity anisotropy.

In a real physical system, a finite number N_0 of impurities will be found. If the impurities are randomly substituted along our chain and the concentration is small ($c = N_0/N \ll 1$), the probability that two impurities will be near each other is small (order c^2), and the interaction between impurities is small. In this case the

eigenfunctions described here remain approximately valid for each impurity and host ions in the vicinity of the impurity. Eigenenergies will be shifted only by a small amount and will assume a small width, and the change in the density of states due to the impurities will be of order c compared to the density of states of the pure chain [20–22]. For a sufficiently large concentration of impurities, then the eigenstates outside the spin wave band and the resonances inside the band can have a measurable effect on properties of systems which behave approximately like linear chain Heisenberg ferromagnets [14–16].

VII. The Double Spin Deviation Problem

Returning to the Hamiltonian Equation (2.1) and imposing the additional condition that K and K_0 be positive (easy axis anisotropy), we now search for eigenstates which have total spin eigenvalue $S^z = (N-1)S + S_0 - 2$. *These are called double spin deviation states.*

It is not *a priori* clear how many spin deviations (magnons) the ground state and the most important excitations of the system contain. Therefore it is essential to include among the set of basic functions, in terms of which the unknown ground state is to be expanded, states with more than one spin deviation.

In the single spin deviation problem for an impurity with nearest neighbor exchange in a Heisenberg ferromagnet, $\langle m|\hat{V}|n\rangle$ is zero unless m and n are the impurity site or its nearest neighbors, where $|m\rangle$ is a spin deviation at lattice site m from the fully aligned state. Therefore the single spin-deviation eigenvalue problem is only of dimension $(z+1) \times (z+1)$, where z is the number of nearest neighbors.

The double spin deviation impurity problem has received little attention in the literature and is a much more difficult problem to solve exactly than the single spin deviation impurity problem. If one proceeds as for the single spin deviation problem, using as a basis the states $|m, n\rangle$ containing spin deviations at lattice sites m and n, one finds that $\langle j, l|\hat{V}|m, n\rangle$ is non-zero not only when j, l, m and n are the impurity sites or its neighbors, but also when j (or l) and m (or n) are arbitrary sites anywhere in the lattice. This means that the Lifshitz [5] partitioning technique does not solve the problem. In addition, the two spin deviation eigenstates of the pure host, although known [23–27], are more complicated than the single spin deviation eigenstates of the pure host, and consist of a two magnon band in which there is magnon-magnon scattering and one or more two magnon bound states lying below the band. Oguchi and Ono [12] formulated the Dyson equation for two spin deviations in the infinite chain Heisenberg ferromagnet containing a substituted impurity for spins $\frac{1}{2}$ and no anisotropy or external field, and obtained an approximate solution by ignoring all matrix elements of $\hat{V}$ except those between the impurity and its neighbors and by ignoring magnon–magnon interactions in the pure chain.

We are particularly interested in the cases for which the ground state contains two spin deviations. In the solution of the single spin deviation problem for the infinite chain Heisenberg ferromagnet containing an impurity, it has been found that localized states below the spin wave band can lie below the normal ground state (the fully aligned state) if the impurity host exchange is antiferromagnetic [7–12]. In the Appendix to the present paper, it is found that the ground state can be expected to contain $2S_0$ spin deviations, where S_0 is the impurity spin, for a significant range of parameter values, if and only if the impurity is antiferromagnetic [28]. Therefore we are particularly interested in the case of an antiferromagnetic impurity of spin $S_0 = 1$, with

the other parameters of the problem arbitrary, because this gives rise to a ground state containing two spin deviations. Oguchi and Ono [12] approximated the ground state in this case by requiring one spin deviation to remain on the impurity, while allowing the other spin deviation to be shared among all lattice sites, which is just a modification of the single spin deviation problem. Ono and Endo [29] recently improved upon the approximation by allowing the spin deviation on the impurity to be shared also by its nearest neighbors.

Instead of Oguchi and Ono's approximate ground state, one might choose two spin deviations of type $S1$, the single spin deviation eigenstates of lowest energy [11, 17]. This choice places both spin deviations on an equal footing and is consistent with linear spin wave theory, which regards state $S1$ as a magnon of negative excitation energy localized about the impurity, for antiferromagnetic impurity–host exchange [11], and which ignores magnon–magnon interactions. Thus linear spin wave theory dictates that the ground state has the maximum number of $S1$ magnons consistent with the restriction that no spin S_j is deviated by more than $2S_j$, and since most of the $S1$ spin deviation is concentrated on the impurity, this restriction is reached for two magnons, if $S_0 = 1$. Linear spin wave theory can be regarded as giving a first approximation to the other double spin deviation eigenstates also. The approximation can then be improved by accounting for magnon–magnon interactions.

Our approach to finding the non-band double spin deviation eigenstates is to take the double spin deviation states as given by linear spin wave theory in terms of the single spin deviation eigenstates as basis, to diagonalize the Hamiltonian with respect to these states, and then to treat the effect of the various bands on these states as a perturbation on the wave functions. Our results will be accurate for the region of parameter values where the perturbation is small. This approach can be generalized to more than two spin deviations and to lattices of higher dimension, at a cost in labor and numerical difficulty.

VIII. Basis States

The N single spin deviation eigenstates $|m)$ of $\hat{\mathscr{H}}$ are given as a sum over single spin deviation states $|j\rangle$ at lattice sites j, as $|m) = \sum_{j=0}^{N-1} c_{mj}|j\rangle$, where the wave function coefficients c_{mj} were obtained in the previous sections. They may be written in terms of the magnon creation operator $\hat{b}_m^+ = \sum_{j=0}^{N-1} c_{mj}\hat{a}_j^+$, where $\hat{a}_j^+|0\rangle = |j\rangle$ and the c_{mj} are real, $|0\rangle$ being the fully aligned state with $S^z = (N-1)S + S_0$, as $|m) = \hat{b}_m^+|0\rangle$. The $\hat{a}_j^+$ operators are given in terms of the spin operators by the Holstein–Primakoff transformation [30]. The double spin deviation eigenstates are N^2 in number and are given by linear spin wave theory, which neglects interactions between the single spin deviation magnons, as

$$
\begin{aligned}
|m, n) &\equiv \hat{b}_m^+\hat{b}_n^+|0\rangle = \sum_{jl}^{j\neq l} c_{mj}c_{nl}|j, l\rangle + \sqrt{2}\sum_j c_{mj}c_{nj}|j, j\rangle, \qquad m \neq n \\
|m, m) &\equiv \frac{1}{\sqrt{2}}(\hat{b}_m^+)^2|0\rangle = \frac{1}{\sqrt{2}}\sum_{jl}^{j\neq l} c_{mj}c_{ml}|j, l\rangle + \sum_j c_{mj}^2|j, j\rangle,
\end{aligned}
\tag{8.1}
$$

where $|j, l\rangle \equiv \hat{a}_j^+\hat{a}_l^+|0\rangle$, $j \neq l$, $|j, j\rangle \equiv (1/\sqrt{2})(\hat{a}_j^+)^2|0\rangle$, and j and l range over all lattice sites. The states given by equation (8.1) are orthonormal, and form a complete set if $S_j \geqslant 1$ for all spins. However, if some $S_j = \frac{1}{2}$, these states are 'over-complete',

since they then contain the unphysical state $|j, j\rangle$. From equation (8.1) and orthonormality of the single spin deviation eigenstates, we have

$$(m, n|\hat{S}_l^z|m, n) = S - c_{ml}^2 - c_{nl}^2, \qquad m \neq n, \tag{8.2}$$

where all $|j, j\rangle$ states (whether $S_j = \frac{1}{2}$ or not) are responsible for $4c_{ml}^2 c_{nl}^2$ of the spin deviation $c_{ml}^2 + c_{nl}^2$, and

$$(m, m|\hat{S}_l^z|m, m) = S - 2c_{ml}^2, \tag{8.3}$$

where all $|j, j\rangle$ states are responsible for $2c_{ml}^4$ of the spin deviation $2c_{ml}^2$. We see that when the spin deviation at site l is small, the contribution to the spin deviation from the $|j, j\rangle$ states is very small. It will turn out that for the only $S_j = \frac{1}{2}$ case we will consider, $S_0 = 1$ and $S = \frac{1}{2}$, the contribution from unphysical states is small for the range of parameter values of interest to us. Therefore, rather than encumber our formalism with a prescription for projecting out the unphysical states $|j, j\rangle$ for $S_j = \frac{1}{2}$ [24], which would also cause our basis states to be non-orthogonal, we shall confine the validity of our results to the range of parameter values for which the spin deviation at site j is less than one if $S_j = \frac{1}{2}$.

Just as in the case of one spin deviation, the two spin deviation eigenstates are divided into those symmetric about the impurity site and those antisymmetric about the impurity site. From equation (8.1) and the symmetry $c_{mN-j} = c_{mj}$ derived in Section II for periodic boundary conditions, one sees that $|m, n)$ is symmetric if $|m)$ and $|n)$ are both symmetric or antisymmetric and that $|m, n)$ is antisymmetric if one is symmetric and the other is antisymmetric. $\hat{\mathscr{H}}$ has no matrix elements connecting symmetric states to antisymmetric states. Thus the true double spin deviation eigenstates (which may be expanded in the $|m, n)$ basis) are either symmetric or antisymmetric.

In the solution of the single spin deviation problem, there are $N/2$ symmetric and $N/2$ antisymmetric eigenstates [31]. Although N^2 double spin deviation states may be formed from them, only $N(N+1)/2$ of these are distinguishable, since the boson state $|m, n)$ is indistinguishable from $|n, m)$. Thus $(N/2)(N/2+1)$ symmetric and $N^2/4$ antisymmetric distinguishable double spin deviation states are found.

Two approaches were considered for the evaluation of the matrix elements of $\hat{\mathscr{H}}$ in terms of the basis states $|m, n)$. One was an expansion of $\hat{\mathscr{H}}$ in terms of $\hat{b}$-type bosons, the other an expansion of $|m, n)$ in terms of the states $|j, l\rangle$. $\hat{\mathscr{H}}$ may be written in terms of $\hat{a}$-type boson operators using the Holstein–Primakoff transformation, and the $\hat{a}$-type boson operators may then be expressed in terms of $\hat{b}$-type boson operators. In this approach, terms such as $\sqrt{2S_j - \hat{a}_j^+ \hat{a}_j}$ arise which must be expanded in powers of S_j^{-1} to be evaluated. The leading terms in S_j^{-1} are readily found for an arbitrary number of spin deviations of the $\hat{b}$-type. However, we are interested in the cases for which the ground state contains $2S_0$ spin deviations, and for most of the ranges of parameters, it is expected from analogy with the single spin deviation solution that most of the spin deviation will be concentrated on the impurity in these cases. Thus the successive matrix element terms derived from the S_0^{-1} expansion of $\sqrt{2S_0 + a_0^+ a_0}$ will be of the same order of magnitude, so that each term must be found and the terms must be summed to infinite order in S_0^{-1}. The attempt to do this sum becomes quite laborious, and this approach was abandoned in favor of leaving $\hat{\mathscr{H}}$ intact and expanding $|m, n)$ in terms of $|j, l\rangle$. By use of equation (8.1), the orthonormality of the wave function coefficients c_{mj}, and the relation

$$(m|\hat{\mathscr{H}}|n) = (E_0 + 2JS\epsilon_m)\delta_{mn}, \tag{8.4}$$

where E_0 is the energy of the fully aligned state, the matrix elements in terms of the basis states $|m, n)$ may be written in the forms

$$(m, n|\hat{\mathscr{H}}|p, q)/2JS = (\delta_{mp}\delta_{nq} + \delta_{mq}\delta_{np})(E_0/2JS + \epsilon_m + \epsilon_n) + 2(D_{mnpq} + D_{nmpq} + D_{pqmn} + D_{qpnm}) - (F_{mnpq} + F_{nmpq} + F_{mnqp} + F_{nmqp}) - 2L_{mnpq}, \quad m \neq n, \quad p \neq q, \tag{8.5}$$

$$(m, m|\hat{\mathscr{H}}|n, p)/2\sqrt{2}\,JS = D_{nmmp} + D_{pmmn} + 2D_{mmnp} - F_{mmnp} - F_{mmpn} - L_{mmnp}, \quad n \neq p, \tag{8.6}$$

and

$$(m, m|\hat{\mathscr{H}}|n, n)/2JS = \delta_{mn}(E_0/2JS + 2\epsilon_m) + 2(D_{mmnn} + D_{nnmm} - F_{mnmn}) - L_{mmnn}, \tag{8.7}$$

where

$$D_{mnpq} = s\left(\sum_{j=1}^{N/2-1} c_{m,j+1}c_{nj}c_{pj}c_{qj} + \sum_{j=2}^{N/2} c_{m,j-1}c_{nj}c_{pj}c_{qj} - \sqrt{\gamma}\,\xi c_{m0}c_{n1}c_{p1}c_{q1}\right) - s_0\sqrt{\gamma}\,\xi c_{m1}c_{n0}c_{p0}c_{q0}, \tag{8.8}$$

$$F_{mnpq} = S^{-1}\left(\sum_{j=1}^{N/2-1} c_{mj}c_{n,j+1}c_{pj}c_{q,j+1} - \xi c_{m0}c_{n1}c_{p0}c_{q1}\right), \tag{8.9}$$

and

$$L_{mnpq} = 2\kappa \sum_{j=1}^{N/2} (2 - \delta_{jN/2})c_{mj}c_{nj}c_{pj}c_{qj} + 2\varphi c_{m0}c_{n0}c_{p0}c_{q0} \tag{8.10}$$

In equations (8.5–8.11), $s = \sqrt{1 - 1/2S}$ and $s_0 = \sqrt{1 - 1/2S_0}$. In deriving equations (8.5–8.10), use has been made of the symmetry $c_{mN-j} = \pm c_{mj}$ [32]. The D terms arise from the $\hat{S}_j^{\pm}\hat{S}_{j\pm1}^{\mp}$ exchange terms, the F terms, from the $\hat{S}_j^z\hat{S}_{j+1}^z$ exchange terms, and the L terms, from the $(\hat{S}_j^z)^2$ anisotropy. In this approach, the matrix elements may be evaluated exactly, but the calculation quickly becomes prohibitively laborious for states containing greater numbers of spin deviations.

IX. Diagonalization with Respect to Discrete States

According to linear spin wave theory, the double spin deviation eigenstates are of the form $|m, n)$ as given by equation (8.1) with excitation energy $E_m + E_n$, where E_m is the excitation energy of the single spin deviation eigenstate $|m)$. The single spin deviation eigenstates consist of $\sim N$ symmetric and antisymmetric states of non-local character [33] which form a quasi-continuum of energies throughout the spin wave band (as given by the pure host), together with as many as three states of discrete energy (two symmetric, one antisymmetric), with spin deviation localized about the impurity which lie outside the band for certain ranges of parameters. Thus there are double spin deviation states $|m, n)$ of three different kinds:

(1) Non-local band states arising when $|m)$ and $|n)$ are single spin deviation band eigenstates. Their number is $\approx N^2$.

(2) Partially local band states arising when $|m)$ is within the single spin deviation band and $|n)$ is outside or *vice versa*. Their number is $\approx N$.

(3) Localized states, which arise when $|m)$ and $|n)$ are both outside the single spin deviation band. Their number is $\leqslant 6$, of which 4 are symmetric, and 2 are antisymmetric.

The Hamiltonian is easily diagonalized with respect to the six localized non-band double spin deviation states.
Suppose

$$\hat{\mathscr{H}}|l\} = E_l|l\}, \tag{9.1}$$

where

$$|l\} = \sum_{m=1}^{6} x_{lm}|m)). \tag{9.2}$$

$|l\}$ denotes any one of six eigenstates of equation (9.1), ordered in increasing eigenenergy E_l from $l = 1$ to 6, and $|m))$ denotes any one of the six non-band double spin deviation states, ordered in some fashion. From equations (9.1) and (9.2) and the orthonormality of the $|m))$ states, we obtain the six homogenous equations

$$\sum_{m=1}^{6} [((m'|\hat{\mathscr{H}}|m)) - E_l\delta_{m'm}]x_{lm} = 0, \qquad m' \text{ and } l = 1, \ldots, 6, \tag{9.3}$$

in the six unknowns x_{lm} for each l. The absolute magnitude of x_{lm} is determined by the orthonormality of $|l\}$. Since $\hat{\mathscr{H}}$ does not connect symmetric states to antisymmetric states, equation (9.3) reduces to four equations for the symmetric states and two equations for the antisymmetric states. Of course, depending on the parameter values, fewer than three single spin deviation states may lie outside the one magnon band, further reducing the number of equations.

Besides E_l and x_{lm}, another physical quantity which we calculate is the average z-component of the spin at site n, in state $|l\}$, $\{l|\hat{S}_n^z|l\}$. Equations (8.2), (8.3) and (9.2) yield the result

$$\begin{aligned}\{l|\hat{S}_n^z|l\} = S_n &- 2(x_{l,S1S1}^2 c_{S1,n}^2 + x_{l,S2S2}^2 c_{S2,n}^2 + x_{l,A1A1}^2 c_{A1,n}^2) \\ &- x_{l,S1S2}^2(c_{S1,n}^2 + c_{S2,n}^2) - 2\sqrt{2}\, x_{l,S1S2}(x_{l,S1S1} + x_{l,S2S2})c_{S1,n}c_{S2,n}\end{aligned} \tag{9.4}$$

for $|l\}$ symmetric, where S denotes a symmetric and A an antisymmetric state, and $S1$, $S2$, and $A1$ refer to single spin deviation eigenstates below the one magnon band. A similar result for antisymmetric $|l\}$ is obtained. If parameter values are such that there are single spin deviation eigenstates above the one magnon band, only a change in notation is required.

The matrix elements required in equation (9.3) are calculated exactly in Ref. [17]. For a wide range of parameters the most important matrix element for the calculation of the lowest lying double spin deviation state is

$$[(S1, S1|\hat{\mathscr{H}}|S1, S1) - E_0]/2JS = 2\epsilon_{S1} + v \qquad \text{for } N \to \infty, \tag{9.5}$$

where

$$\begin{aligned}v = 2c_{S1,0}^4\{&2s[\lambda_{S1}^4\alpha_{S1}(1 - \alpha_{S1}^2)^{-1} - \lambda_{S1}^3\sqrt{\gamma}\,\xi] - 2s_0\lambda_{S1}\sqrt{\gamma}\,\xi \\ &- S^{-1}[\lambda_{S1}^4\alpha_{S1}^2(1 - \alpha_{S1}^4)^{-1} - \lambda_{S1}^2\xi] - 2\lambda_{S1}^4\kappa(1 - \alpha_{S1}^4)^{-1} - \varphi\},\end{aligned} \tag{9.6}$$

and $\lambda_{S1} = \lambda(\epsilon_{S1})$ and $\alpha_{S1} = \alpha(\epsilon_{S1})$ are defined by equations (2.17) and (3.2). The state

$S1$ is the lowest lying single spin deviation state below the one magnon band, and equation (9.5) gives the expectation value of $\hat{\mathscr{H}}$ in the double spin deviation state $|S1, S1)$ consisting of two $S1$ magnons to within an error of order N^{-1}, where $2\epsilon_{S1}$ is the excitation energy as given by linear spin wave theory and v is the interaction energy between two $S1$ magnons.

The states $|l\}$ together with the band states excluded from equations (9.1–9.6) form an orthonormal complete basis for the double spin deviation problem. If the effect of the band states on $|l\}$ is small, their effect may be treated as a perturbation, which will produce corrections to $|l\}$ and E_l. This is done in the next section. If E_l lies within a double spin deviation band of states having the same symmetry as $|l\}$, $|l\}$ would be expected to decay into the band states and to have a finite line width. In this situation, our calculation, which does not yield any line width for $|l\}$, cannot be expected to describe $|l\}$ at all, although the appearance of such a state $|l\}$ within a band may possibly indicate a resonance at E_l. Presuming this situation does not occur for the state $|l\}$ of interest and the parameter values considered, we shall refer to the states $|l\}$ as 'discrete' states, since the remaining double spin deviation states of our basis form energy bands. It should be borne in mind, however, that an exact calculation, if it were done, might not only yield energy shifts for the $|l\}$ states, but small line widths as well, even when E_l is not within a band.

X. Perturbational Effect of Band States on Discrete States

Let the true discrete eigenstate $|l]$, which is represented by $|l\}$ in the absence of the perturbation due to the band states, be expanded in terms of our basis as

$$|l] = u_l|l\} + \sum_{l' \neq l} u_{l'}|l'\} + \sum_{b \neq l}^{b \neq l'} u_b|b)), \tag{10.1}$$

where $|b))$ represents any one of the double spin deviation band states and u_i are the expansion coefficients. In this section, the following conventions on arbitrary state indices will be used: i and j will designate states which may be either discrete or band states; l and l' will designate discrete states only; b and b' will designate band states only. Thus $\mathscr{H}_{ll'} = \{l|\hat{\mathscr{H}}|l'\} = 0, l \neq l'$, since the discrete states have been diagonalized among themselves. If the true eigenenergy for $|l]$ is E, the following set of homogeneous equations for the wave function coefficients is found,

$$(E - E_l)u_l = \sum_{i \neq l} \mathscr{H}_{li}u_i, \tag{10.2}$$

and

$$u_i = \left(\mathscr{H}_{il}u_l + \sum_{j \neq i}^{j \neq l} \mathscr{H}_{ij}u_j\right)(E - E_i)^{-1}, \qquad i \neq l, \tag{10.3}$$

where $E_j = \mathscr{H}_{jj}$. Equation (10.3) has been written in a manner suggesting iteration, since it is expected that $|u_i|$, $i \neq l$ will be small compared to $|u_l|$. Substituting equation (10.3) for u_i in equation (10.2), we have the exact relation

$$(E - E_l)u_l = u_l \sum_{i \neq l} \mathscr{H}_{li}\mathscr{H}_{il}(E - E_i)^{-1} + \sum_{i \neq l} \sum_{j \neq l}^{j \neq i} \mathscr{H}_{li}\mathscr{H}_{ij}u_j(E - E_i)^{-1}. \tag{10.4}$$

The iteration can be continued by substituting equation (10.3) for u_j in equation (10.4). If

$$\left|\sum_{j\neq i}^{j\neq l} \mathscr{H}_{ij}u_j\right| \ll |\mathscr{H}_{il}u_l|, \tag{10.5}$$

then it is seen from equations (10.3) and (10.4) that each successive new term in the interaction scheme is much smaller than the preceding term, so that

$$u_i \approx \mathscr{H}_{il}u_l(E - E_i)^{-1}, \qquad i \neq l, \tag{10.6}$$

and

$$E - E_l \approx \sum_{i\neq l} \mathscr{H}_{li}\mathscr{H}_{il}(E - E_i)^{-1}, \tag{10.7}$$

in the first approximation.

Equation (10.7) is an implicit equation for E. To remove E from the denominator, we assume [34]

$$|\Delta E_l| \equiv |E - E_l| \ll |E_l - E_B|, \tag{10.8}$$

where E_B is the energy of the band state lying closest to $|l\}$ in energy. Then equation (10.7) becomes

$$\Delta E_l \approx \sum_b |\{l|\hat{\mathscr{H}}|b))|^2(E_l - E_b)^{-1}. \tag{10.9}$$

To the same approximation, equation (10.6) becomes

$$u_b \approx \mathscr{H}_{bl}u_l(E_l - E_b)^{-1}. \tag{10.10}$$

For discrete states $|l'\}$, $l' \neq l$, equation (10.6) yields $u_{l'} \approx 0$. Applying the condition $\sum_i |u_i|^2 = 1$, we find from equation (10.10) that

$$|u_l|^{-2} \approx 1 + \sum_b |\mathscr{H}_{lb}|^2(E_l - E_b)^{-2}. \tag{10.11}$$

From equation (10.9) it is seen that equation (10.8) implies

$$\sum_b |\mathscr{H}_{lb}|^2(E_l - E_b)^{-2} \ll 1, \tag{10.12}$$

so we may replace u_l by 1 and obtain $u_b \approx \mathscr{H}_{bl}(E_l - E_b)^{-1}$ from equation (10.11) in the first approximation, or

$$|l] \approx |l\} + \sum_b \frac{((b|\hat{\mathscr{H}}|l\}}{E_l - E_b}\,|b)). \tag{10.13}$$

Using equation (10.10), Condition (10.5) can be written self-consistently as

$$\left|\sum_{b'\neq b} \mathscr{H}_{bb'}\mathscr{H}_{b'l}(E_l - E_b)^{-1}\right| \ll |\mathscr{H}_{bl}|. \tag{10.14}$$

At first glance, one might suppose that the left and right hand sides of (10.14) are of the same order of magnitude except for $E_l - E_b$, since both sides are of the same order in N. For instance, if b and b' each represent a state composed of two single spin deviation band eigenstates, then $\mathscr{H}_{b'l}$ and $\mathscr{H}_{bl}$ are of the same order of magnitude, $\mathscr{H}_{bb'}$ is of order N^{-2} (as seen from equations (8.5–8.10), noting from Ref. [17] that

the wave function coefficients appearing are all of order $N^{-1/2}$), and b' ranges over N^2 states. If this were true, Condition (10.14) could be satisfied only if E_l were far away from E_B. However, it is possible that the band states are 'well-diagonalized' such that Condition (10.14) may be satisfied for E_l rather close to E_B, and we shall see in Section XI that this can be true for the lowest lying discrete state. The quantity on the left hand side of Condition (10.14) is rather difficult to calculate. We expect that Condition (10.14) will be satisfied when Condition (10.8) is satisfied, so in practice we shall consider satisfaction of Condition (10.8) as sufficient for validity of equations (10.9) and (10.13).

We now proceed to develop formulas viable to numerical computation. As seen from equations (8.5–8.10), E_b in equation (10.9) may be written as the sum of the corresponding single spin deviation eigenenergies within an error of order N^{-1}. We then have for the symmetric states $|l]$,

$$\begin{aligned}\Delta E_l \approx & \sum_b |\{l|\hat{\mathscr{H}}|S1, Sb)|^2(E_l - E_{S1} - E_b)^{-1} \\ & + \sum_b |\{l|\hat{\mathscr{H}}|S2, Sb)|^2(E_l - E_{S2} - E_b)^{-1} \\ & + \sum_b |\{l|\hat{\mathscr{H}}|A1, Ab)|^2(E_l - E_{A1} - E_b)^{-1} \\ & + \sum_{bb'}^{E_{b'} \geq E_b} |\{l|\hat{\mathscr{H}}|Sb,\ Sb')|^2(E_l - E_b - E_{b'})^{-1} \\ & + \sum_{bb'}^{E_{b'} \geq E_b} |\{l|\hat{\mathscr{H}}|Ab, Ab')|^2(E_l - E_b - E_{b'})^{-1}, \qquad |l\} \text{ symmetric,} \end{aligned} \tag{10.15}$$

where S denotes a symmetric and A denotes an antisymmetric $\hat{b}$-type magnon and $S1$, $S2$, and $A1$ refer to single spin deviation eigenstates below the one magnon band. If parameter values are such that there are single spin deviation eigenstates above the one magnon band, only a change in notation is required. Since the band states form a quasi-continuum for $N \gg 1$, it is necessary to convert the sums in equation (10.15) to integrals, as $\sum_{bS,A} \to \int \rho_{S,A}\, d\epsilon'$, where $\rho_{S,A}$ is the density of symmetric or antisymmetric single spin deviation eigenstates and ϵ' is the corresponding normalized eigenenergy. $\rho_{S,A}$ may be replaced by $\rho_0/2$ to order unity. In the same manner, the energy correction to the antisymmetric discrete states due to the band states can also be found.

From equation (10.13) the correction $\Delta\{l|\hat{S}^z_m|l\}$ to the average of the spin z-component at site n in state $|l\}$ can be written in the first approximation as

$$\Delta\{l|\hat{S}^z_n|l\} \approx 2\sum_b \{l|\hat{\mathscr{H}}|b))((b|S_n - \sum_{ij} c_{in}c_{jn}\hat{b}_i^+\hat{b}_j|l\}(E_l - E_b)^{-1}. \tag{10.16}$$

Since the wave function perturbation is essentially of first order, equation (10.16) is a first order result. Since the energy perturbation is essentially second order, our formulas compute ΔE_l to greater accuracy than $\Delta\{l|\hat{S}^z_n|l\}$.

The matrix elements required in equations (10.15–10.16) are calculated exactly by use of equations (8.5–8.10), the single spin deviation wave function coefficients, trigonometric identities, and several summation formulas for geometric progressions. Their explicit forms are not given here, but may be found in Ref. [17]. No analytic closed form was found for the integrals over these matrix elements; therefore the integrations had to be performed numerically.

XI. Discussion of the Results

For the numerical computations, only the case of $S_0 = 1$, $\xi > 0$ and $\kappa = \varphi = 0$ (no anisotropy) is considered. Instead of the single spin deviation eigenenergy ϵ_j, the parameter $\epsilon'_j = \epsilon_j - gh$ is used because ϵ'_j is independent of gh, as are also the wave function coefficients. Similarly $\mathscr{E}'_l = \mathscr{E}_l - 2gh$ and $\Delta\mathscr{E}_l = \Delta\mathscr{E}'_l$ (where $\mathscr{E}_l = (E_l - E_0)/2JS$) for the double spin deviation problem are independent of gh, as are also the perturbed eigenstates. $\Delta\mathscr{E}_l$ is the change in ϵ_l due to the perturbational effect of the band states. Throughout this section, it is to be understood that $\Delta\epsilon = \Delta\mathscr{E}'$, $\mathscr{E} = \mathscr{E}_l + \Delta\mathscr{E}$, and $\mathscr{E}' = \mathscr{E}'_l + \Delta\mathscr{E}$ refer to the discrete double spin deviation of lowest energy.

The numerical computations were performed on the Florida State University CDC 6400 computer. The numerical errors in all quantities obtained from our theory which are presented in this section are estimated to be $\leqslant 0.1\%$. Details of the numerical computations are given in Ref. [17], where results of our calculation for additional parameter values other than those presented here may also be found.

1. Zero applied field

Before proceeding with the calculation of the metamagnetic transition from a ground state with one spin deviation to one with two spin deviations, we study $\mathscr{E}'$ and $[l|\hat{S}^z_n|l]$ for the case $\eta = (g_0 - g)h = 0$ (such as occurs for zero applied field h) and compare our results with the Oguchi and Ono approximation for the two spin deviation ground state in which one spin deviation is isolated on the impurity while the second deviation is free to move about in the lattice [35]. The results are shown

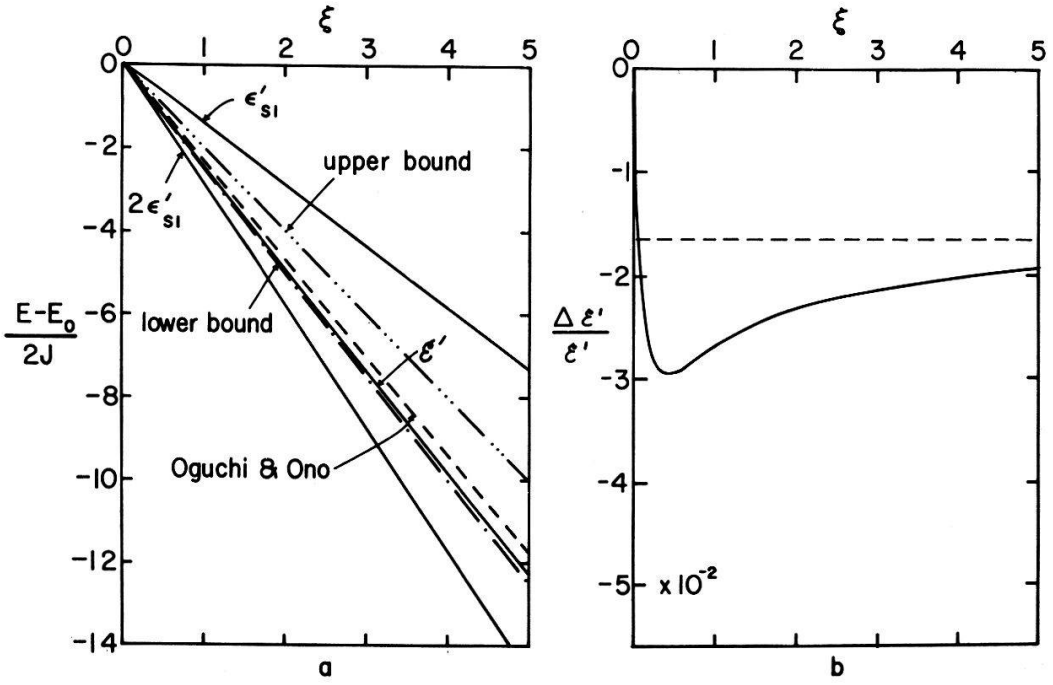

Figure 6

Part a: Energy $\mathscr{E}' = (E - E_0)/2SJ$ of the double spin deviation state of lowest energy as a function of ξ, for $\eta = 0$, $S = 1$, $S_0 = 1$, $K = 0$, $K_0 = 0$ along with upper and lower bounds. The energy ϵ'_{S1} of the lowest single spin deviation state, $2\epsilon'_{S1}$, and the energy of Oguchi and Ono's [12] double spin deviation state are shown for comparison. *Part b*: Fractional energy change $\Delta\mathscr{E}'/\mathscr{E}'$ of the lowest lying double spin deviation state due to the band state perturbation, as a function of ξ, for $\eta = 0$, $S = 1$, $K = 0$, $K_0 = 0$, $S_0 = 1$. The dashed line is the asymptote as $\xi \to \infty$. The parameters are defined in equations (2.1) and (2.13), and E_0 is the energy of the bottom of the spin wave band.

in Figures 6–9. In Figures 6a and 8a, we see that $\mathscr{E}'$ is considerably lower in energy than the lowest single spin deviation energy ϵ'_{S1}, but is raised above the lowest two spin deviation energy $2\epsilon'_{S1}$ as given by the linear spin wave theory, by interaction between the various $\hat{b}$-type magnons present. Since $\Delta\mathscr{E}'_l < 0$ for the lowest lying discrete state, the net interaction of this state with the band magnons lowers its energy, but the

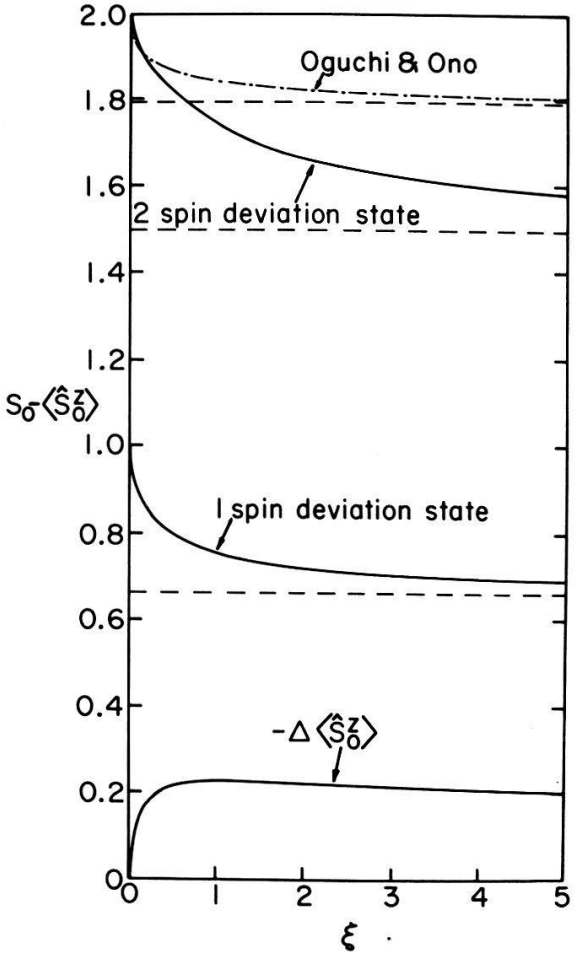

Figure 7
Spin deviation $S_0 - \langle \hat{S}^z_0 \rangle$ of the impurity ion corresponding to the energy $\mathscr{E}'$ of the lowest lying double spin deviation state as a function of ξ, for $\eta = 0$, $S = 1$, $S_0 = 1$, $K = 0$, $K_0 = 0$, along with the change $-\Delta\langle \hat{S}^z_0 \rangle$ in spin deviation due to the band state perturbation. The spin deviation of the $S1$ single spin deviation state are shown for comparison. Dashed lines are asymptotes as $\xi \to \infty$. The parameters are defined in equations (2.1) and (2.13).

larger interaction (of opposite sign) with the discrete state magnons causes a net increase of $\mathscr{E}$ above $2\epsilon'_{S1}$. The largest interaction between discrete state magnons occurs between two $S1$ magnons and is positive, as given by equation (9.6), and the lowest lying discrete state has a large $|S1, S1)$ component x_{S1S1}. The remaining discrete states have a relatively minor, but measurable, effect on $\mathscr{E}'$.

It is also seen in Figures 6a and 8a, that the energy $\mathscr{E}'$ computed for the double spin deviation eigenstate approaches the upper bound as $\xi \to 0$, and approaches the lower bound as $\xi \to \infty$. The fractional difference between $\mathscr{E}'$ and the lower bound approaches zero as $\xi \to \infty$. This is the behavior required of the true lowest eigenstate (see Appendix). Although the energy of the Oguchi and Ono state correctly approaches the upper bound as $\xi \to 0$, as $\xi \to \infty$ it approaches -2.366ξ for host spin $S = 1$ instead of the correct -2.5ξ, and -2.618ξ for $S = \frac{1}{2}$ instead of the correct -3ξ (see Appendix, equations (A.1) and (A.8)).

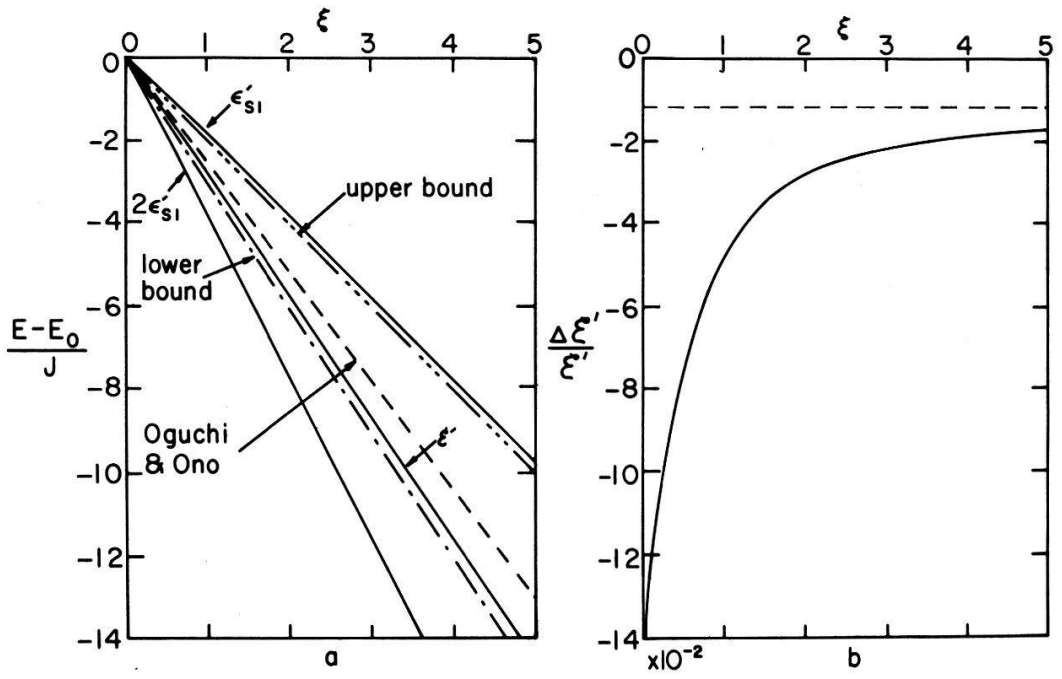

Figure 8a and b:
Same as Figure 6, except $S = \frac{1}{2}$.

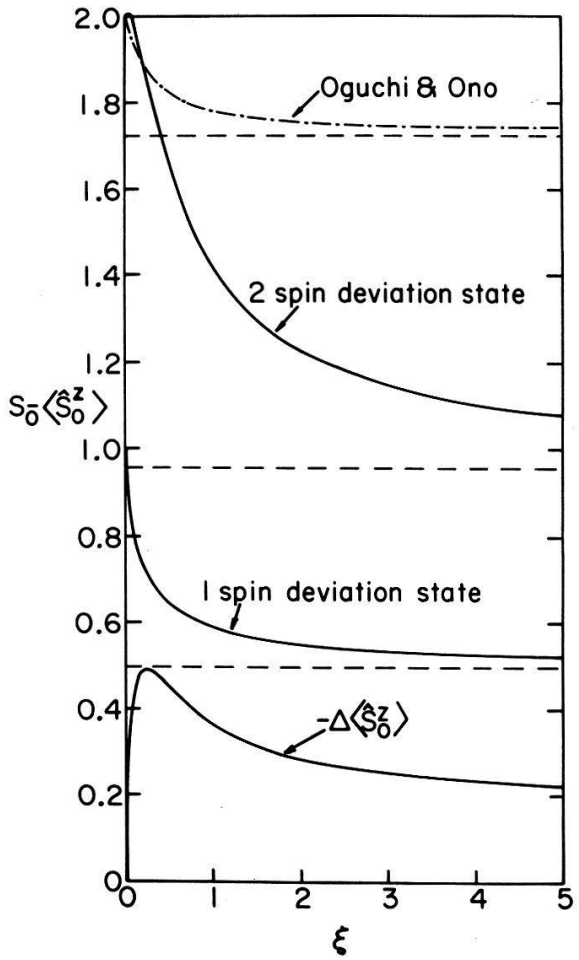

Figure 9:
Same as Figure 7, except $S = \frac{1}{2}$.

Figures 6b and 8b display the fractional change in the energy $\Delta\mathscr{E}'/\mathscr{E}'$ of the lowest lying double spin deviation state due to the band state perturbation, which is measure of the degree of satisfaction of Condition (10.8). We must have $|\Delta\mathscr{E}'/\mathscr{E}'| \ll 1$ in order that equations (10.15) and (10.16) give accurately the effect of the band states on the energy and spin deviation. A characteristic of $\Delta\mathscr{E}'/\mathscr{E}'$ is an asymptotic approach as $\xi \to \infty$ to a small constant which is smaller for larger host spin S, a dip which is smaller and occurs at larger ξ for larger S, and an upswing to zero at $\xi = 0$ (the dip occurs at $\xi \sim 0.001$, $\Delta\mathscr{E}'/\mathscr{E}' \sim 0.25$, for $S = \frac{1}{2}$ in Figure 8b and so cannot be shown). This behavior occurs because the matrix elements in equation (10.15) are of order ξ as $\xi \to \infty$ as are the energy denominators, but approach zero faster than ξ as $\xi \to 0$ while the energy denominators are again of order ξ. The contribution of the band perturbation to the impurity spin deviation has behavior similar to $\Delta\mathscr{E}'/\mathscr{E}'$, as seen in Figure 7 and 9, as might be expected from the form of equation (10.16).

Since $|\Delta\mathscr{E}'/\mathscr{E}'| \ll 1$ for all ξ and S (except near $\xi = 0.001$ for $S = \frac{1}{2}$) for the lowest lying discrete double spin deviation state, one might say that the first order perturbation on the wave function gives an accurate result for all ξ and S. However, Condition (10.14) must be satisfied for the perturbation expansion to converge. The largest contribution to $\Delta\mathscr{E}'$ is for $|b)) = |Sb, Sb')$ in equations (10.9) and (10.15), because for this set of band states, $|\mathscr{H}_{bl}| \gg |\mathscr{H}_{b'l}|$, $b' \neq b$. If further, $|b'))$ is a partially local band state such as $|S1, Sb)$, it is found that $\mathscr{H}_{bb'} \to 0$ is $\xi \to 0$ at least as fast as $\mathscr{E}' \to 0$, so that Condition (10.14) is well satisfied for this case for all ξ. If $|b)) = |Sb, Sb')$ and $|b'))$ is also a non-local band state, $|Ab'', Ab''')$, the evaluation of $\mathscr{H}_{bb'}$ is tedious and has not been carried out. However, in this case, both $|b))$ and $|b'))$ spread the spin deviation almost equally over all lattice sites and are very similar to the set of states $|k, k'\rangle$ containing free pure host magnons of momentum k and k', the spin deviation at each lattice site being of order N^{-1}. The pure host two spin deviation problem has been solved exactly [23–27], and it is found that the two magnon interaction is small, except for the creation of bands of two magnon bound states. Our approach is incapable of yielding these bound magnons, since the perturbation expansion must then be carried to infinite order, but since there are only $\sim N$ such states as compared to $\sim N^2$ unbound magnon states, and since no bound magnons have energies below that of the fully aligned state, we presume their effect on the lowest lying discrete state is small, except, perhaps for $\xi \sim 0$. Certainly Condition (10.14) is satisfied for large ξ, but it may be that $\mathscr{E}'$ approaches zero as $\xi \to 0$ with a somewhat different functional dependence than that given by our first order perturbation on the wave function.

In Figures 7 and 9 is shown the spin deviation of the impurity ion in the lowest lying discrete double spin deviation state, as calculated from equations (9.4) and (10.16) and also as given by the Oguchi and Ono approximation. The impurity spin deviation should approach 2 as $\xi \to 0$ and as $\xi \to \infty$ should approach the spin deviation characteristic of the lowest state of the system composed of the impurity and its nearest neighbors (see Appendix). Both our approximation and that of Oguchi and Ono give the correct result for $\xi = 0$. For $\xi \to \infty$ and $S = 1$, our calculation yields the correct result, 1.5 (see Appendix, equation (A.10)), within numerical error, but the Oguchi and Ono result is 1.79. For $\xi \to \infty$ and $S = \frac{1}{2}$, our approximation gives 0.96, close to the correct result, 1.0, while that of Oguchi and Ono gives 1.72. But also for $S = \frac{1}{2}$, our approximation slightly violates the kinematic restriction that the spin deviation be no greater than 2 (for $S_0 = 1$) for small ξ, reaching a peak spin deviation of 2.009. It may be that inclusion of higher order perturbation terms

would yield better results for $S = \frac{1}{2}$ in our scheme, but the violation of the kinematic restriction may be at least partly due to the unphysical states $|j, j\rangle$, $j \neq 0$. Even though $S_0 = 1$, we see in equations (8.2) and (8.3) that these states contribute spin deviation to the impurity ion when our basis states $|m, n)$ are used.

We see that our approximation yields results generally superior to that used by Oguchi and Ono, for the lowest lying double spin deviation eigenstate. We also see that the inclusion of the band perturbation is essential to obtain correct behavior of the energy and spin deviation. It is perhaps surprising that the band states, which have non-local components, give a non-vanishing contribution as $\xi \to \infty$, since the spin deviation in this limit must be fully concentrated on the impurity and its neighbors (see Appendix). This may be understood by noting that the non-local components strongly 'interfere' for sites farther away from the impurity, such that the sums in equation (10.16) decrease rapidly with distance from the impurity, and the spin deviation perturbation vanishes for sites other than the impurity and its nearest neighbors in the limit $\xi \to \infty$ [17].

2. *The metamagnetic transition*

When a magnetic field is applied, the energy $\mathscr{E}$ of the lowest double spin deviation state is raised relative to the energy ϵ_{S1} of the lowest single spin deviation state, which is itself raised relative to the fully aligned state. A metamagnetic transition from two spin deviations to one or zero spin deviations can occur at a critical field h_{c2}, below which the ground state contains two spin deviations. A metamagnetic transition from one spin deviation to the fully aligned state can occur at a critical field h_{c1}, above which the ground state contains no spin deviations. Since ϵ_{S1} is measured relative to the fully aligned state, h_{c1} is given by $\epsilon_{S1} = 0$, or

$$gh_{c1} = -\epsilon'_{S1}, \tag{11.1}$$

provided $h_{c1} > 0$ ($\epsilon'_{S1} < 0$). The ground state contains at least one spin deviation if $h < h_{c1}$. If $\epsilon'_{S1} > 0$, the ground state is the fully aligned state. Then h_{c2} is given by $\mathscr{E} = \epsilon_{S1}$, or

$$gh_{c2} = \epsilon'_{S1} - \mathscr{E}', \tag{11.2}$$

if $h_{c1} < h_{c2}$, provided $h_{c2} > 0$ ($\epsilon'_{S1} > \mathscr{E}'$). If $\mathscr{E}' > \epsilon'_{S1}$, the ground state cannot contain two spin deviations.

The condition $h_{c1} < h_{c2}$ implies from equations (11.1) and (11.2) that $\mathscr{E}' > 2\epsilon'_{S1}$, that is, the net interaction of two $S1$ magnons with each other and the other magnons must be repulsive. If the net interaction is attractive, or $\mathscr{E}' < 2\epsilon'_{S1}$ ($h_{c1} > h_{c2}$), the metamagnetic transition occurs directly from two spin deviations to the fully aligned state, and h_{c1} is meaningless. If $\mathscr{E}' < 2\epsilon'_{S1}$, the phase transition is given by $\mathscr{E} = 0$, or

$$gh_{c2} = -\mathscr{E}'/2. \tag{11.3}$$

If $\mathscr{E}' < 2\epsilon'_{S1}$, the ground state contains two spin deviations if $h < h_{c2}$, none if $h > h_{c2}$.

Since $\mathscr{E}'$ and ϵ'_{S1} are independent of gh, it is convenient to fix S, ξ, and η and determine h_{c1} and h_{c2} from equations (11.1–11.3). Then, for given S and ξ, the value of g_0/g at which h_c occurs is given for each value of η by [36]

$$g_0/g = 1 + \eta/gh_c, \tag{11.4}$$

where h_c is h_{c1} or h_{c2}. The results of this calculation are displayed in Figure 10 as phase diagrams of h vs. g_0/g for $\xi = 10$, 1, and 0.1, and for $S = 1$ and $\frac{1}{2}$, wherein

h_{c1} and h_{c2} phase lines are shown. A general feature of the h_{c2} phase lines is that they are similar to the h_{c1} phase lines, and like h_{c1}, $h_{c2} \to \xi/g_0$ as $g_0/g \to 0$ and $h_{c2} \to -\epsilon'_{A1}/g$ as $g_0/g \to \infty$.

Also shown in Figure 10 is a curve, denoted by $h_{c2'}$, which represents the two spin deviation phase line without including the band perturbation $\Delta\mathscr{E}$. The net interaction of two $S1$ magnons with each other and the other discrete magnon states is repulsive, so that $h_{c2'} < h_{c1}$. It is found that if $\eta < 0$, or if $\eta > 0$ and not too large, the interaction between two $S1$ magnons is dominant. Since $\Delta\mathscr{E}' < 0$, we have $h_{c2} > h_{c2'}$, but, with one exception, $\Delta\mathscr{E}'$ is small enough in Figure 10 that h_{c2} is close to $h_{c2'}$ and

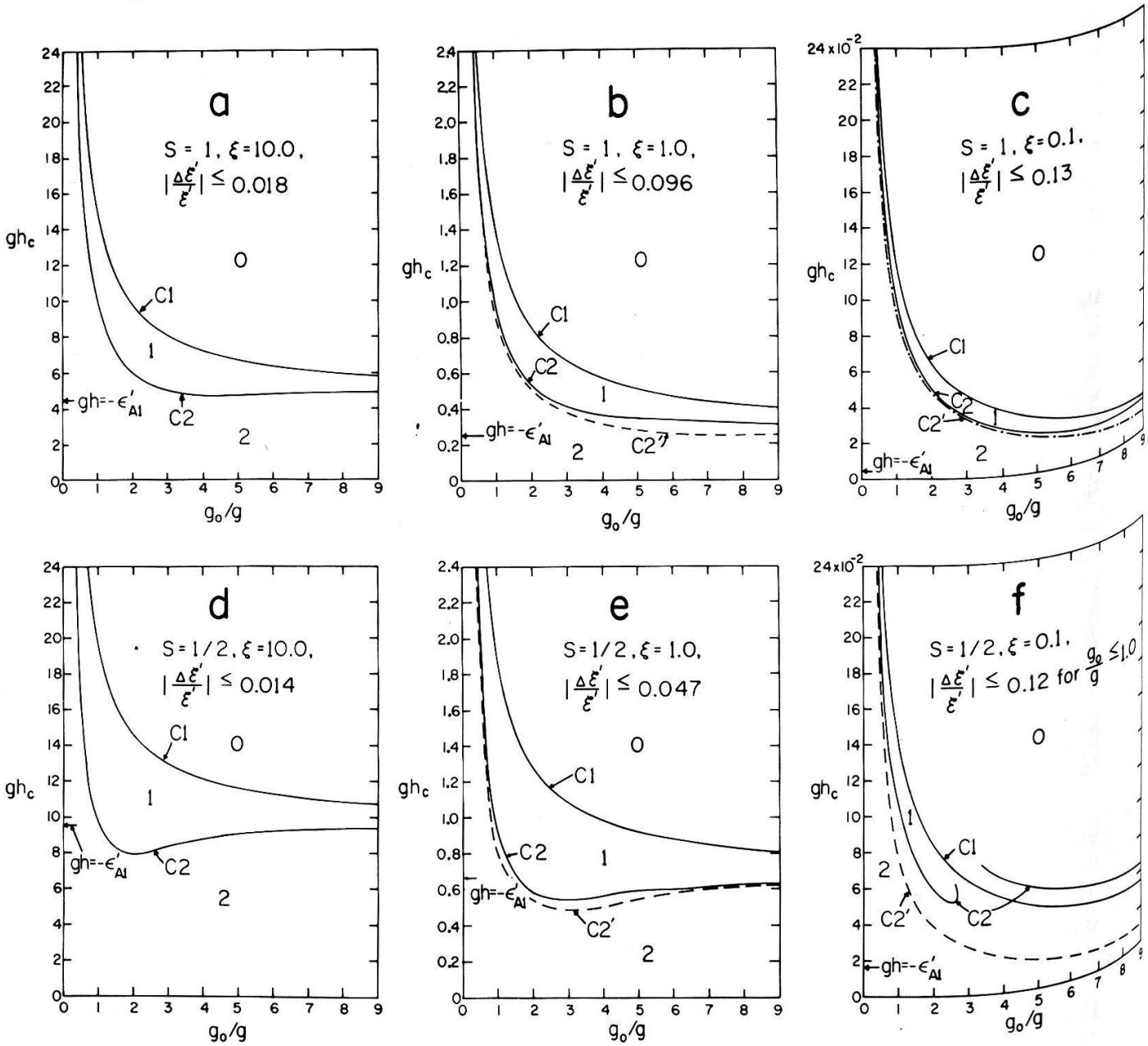

Figure 10
Critical field h_c for the metamagnetic transition to the ground state containing one (curve $c1$) and two (curve $c2$) spin deviations as a function of g_0/g, for different values of the parameter S and ξ, for $K = K_0 = 0$. Labels '0', '1' and '2' designate regions where the ground state contains 0 (fully aligned state), 1 and 2 spin deviations, respectively. In Figures a and d the magnitude of band perturbation $\Delta\mathscr{E}'$ is too small to have noticeable effect on curve $c2$. In Figures b, c, e and f, the curves $c2'$ is obtained from $c2$ by neglecting $\Delta\mathscr{E}'$. The parameters are defined in equations (2.1) and (2.13), $\Delta\mathscr{E}'$ and $\mathscr{E}'$ are introduced in Section XI.

$h_{c2} < h_{c1}$, so that the net interaction of two $S1$ magnons with all magnon states, including band magnons, is repulsive. Note that in Figure 10, for $\xi = 0.1$ and $S = \frac{1}{2}$, we have $h_{c2} > h_{c1}$ for $g_0/g \gtrsim 2.7$. In this range of parameters, we also have $|\Delta\mathscr{E}'/\mathscr{E}'| > 1$, so that the first order perturbation calculation is invalid. This becomes clear from Figures 6b and 8b which show that $|\Delta\mathscr{E}'/\mathscr{E}'|$ is largest for $S = \frac{1}{2}$ when ξ is small, for $\eta = 0$. In the broken part of the curve which represents h_{c2} for $\xi = 0.1$ and $S = \frac{1}{2}$, a simple pole appears [17] in the first sum in equation (10.15) for $\Delta\mathscr{E}$, because $\mathscr{E}'_l > \mathscr{E}'_{S1}$. Even for $\xi = 0.1$ and $S = \frac{1}{2}$, our approximation appears valid for $g_0/g \leqslant 1$, since $|\Delta\mathscr{E}'/\mathscr{E}'|$ is then small.

For $S = \frac{1}{2}$, the spin deviation at all lattice sites j is less than $2S_j$ in the vicinity of the h_{c2} phase line in our calculation of the lowest lying discrete double spin deviation eigenstate, except for the region described in the preceding paragraph where $|\Delta\mathscr{E}'/\mathscr{E}'| > 1$. The unphysical states $|j, j\rangle, j \neq 0$, appear to have no obvious effect in cases where the band perturbation theory is valid.

The question arises, whether for $S_0 = 1$, a region of parameter values exist for which the ground state contains more than two spin deviations. We answer this question by paraphrasing a more general result found in the Appendix to suit the case at hand of zero anisotropy. It is expected that for $S \geqslant 1$, $g_0/g > 2S - 1$, a region exists for $\xi \gg 1$ and $gh \ll \xi/2S$ where the ground state contains $4S$ spin deviations. But in all other parameter regions, it is expected that the ground state contains no more than two spin deviations.

XII. Conclusion

We have developed an approximation scheme for finding the discrete double spin deviation eigenstates of the infinite chain Heisenberg ferromagnet containing a substituted impurity. Taking the double spin deviation states as given by linear spin wave theory in terms of the exact single spin deviation eigenstates as basis, the Hamiltonian was diagonalized with respect to the discrete states, and the band states were treated as a perturbation on the resulting wave functions. The approximation was applied to the lowest lying discrete double spin deviation state for impurity spin one and appears to yield accurate results for the eigenenergy, wave function, and metamagnetic transition over a wide range of parameters.

This approximation scheme was compared to the Oguchi and Ono approximation [12] in which one spin deviation is required to remain on the impurity ion, for the case of zero field, zero anistropy, and antiferromagnetic impurity–host exchange. For this case, the spin deviation is rather well localized about the impurity ion, and the Oguchi and Ono approximation should yield its most favorable result. Our approximation scheme in this case definitely gives the more accurate result over a wider range of impurity–host exchange. The Oguchi and Ono approximation is particularly poor for large antiferromagnetic impurity–host exchange, but has been improved upon by Ono and Endo [29] by allowing the second spin deviation to be shared by the impurity with its neighbors, and their result is much closer to ours for this case. However, Ono and Endo's basis states are not orthogonal, and are approximately orthogonal only when the spin deviation is rather well localized about the impurity, so that their approximation is still rather restricted. Our basis states are exactly orthogonal, regardless of the localization of the spin deviation, and our method appears to give good results for parameter ranges for which the spin deviation is not well localized.

Our approximation scheme should give even better results for the discrete double

spin deviation states of the three-dimensional ferromagnet containing an impurity, because Condition (10.14), which is required for convergence of the band perturbation results, should be better satisfied than in the one-dimensional case. This is because in three dimensions the propagating band magnons can avoid each other so that their interaction should be less. This reasoning is found to be valid when comparing the two spin deviation solutions for the one- and three-dimensional cases in the pure host [23–27]. Extension of our calculation to the three-dimensional case is possible. The increase in number of neighbors to the impurity ion results in a larger matrix to be solved for the single spin deviation problem and results in more discrete states to be included in the diagonalization as well as more band states of different symmetry [16] to be accounted for in the band perturbation, which will also involve integrals over the three-dimensional density of states. The three-dimensional pure host Green functions cannot be given a closed form, but there are viable approximations for them [37, 38].

The extension of our approximation scheme to more than two spin deviations, while simple in principle, becomes tedious and intractable for a large number of spin deviations. For the n-spin deviation problem, $\sim N^n$ different states must be formed from one spin deviation eigenstates, resulting in $\sim 3^n$ discrete states to be included in the diagonalization and, more importantly, resulting in $\sim n$ dimensional integrals to be evaluated in the band perturbation calculation. Higher order terms in the perturbation expansion, very difficult to calculate, may be required as well.

A crude but simple estimate of the energy of the lowest lying n-spin deviation state can be made when the excitation energy ϵ_{S1} of the lowest lying single spin deviation eigenstate is negative, in the region of parameter values (see Section XI) for which the dominant interaction for two magnons is the interaction energy between two $S1$ magnons, given by v in equation (9.6). If we include only two body interactions, then the energy of the n-spin deviation state in the interacting magnon picture is

$$\mathscr{E}(n) \sim n\epsilon_{S1} + n(n-1)v/2, \tag{12.1}$$

$n(n-1)/2$ being the number of two body interaction bonds for n magnons of type $S1$. If $\epsilon_{S1} < 0$, the ground state is obtained by exciting $S1$ magnons out of the fully aligned state until either the addition of another magnon would raise the energy ($\mathscr{E}(n+1) > \mathscr{E}(n) < \mathscr{E}(n-1)$) or until the spin deviation at some lattice site j would be greater than $2S_j$, whichever happens first. Thus the number of spin deviations n_g in the ground state is given by

$$-\epsilon_{S1}v^{-1} < n_g < 1 - \epsilon_{S1}v^{-1}, \qquad \epsilon_{S1} < 0, \qquad v > 0, \tag{12.2}$$

provided the spin deviation at each lattice site j is no greater than $2S_j$. We see that n_g is large when $|\epsilon_{S1}| \gg v$. If $v < 0$, no minimum energy is reached as a function of n, and the ground state then contains the maximum number of spin deviations consistent with the kinematic restriction on the spin deviation. However, for all parameter ranges considered in Section XI, it is found that $v > 0$. We note that Wortis [23] has developed a Green-function method to find the double spin deviation states, in particular, the bound states of two magnons, for a pure ferromagnet. We have extended his method to the impurity problem [39]. It was found, that his method leads to great mathematical complications already in the one spin deviation case, without leading to any new results. Application of Wortis' method to the two spin deviation impurity

problem led to even greater complexities, and no approximation scheme was found which would be as transparent as the one used here.

Appendix

The problem of the exact multiple spin deviation ground state for the infinite chain Heisenberg ferromagnet with impurity remains unsolved. Since our calculation is an approximation, useful upper and lower bounds on the ground state energy will be obtained by a method similar to the one used by Anderson [40] for the pure antiferromagnet and by Oguchi and Ono [12] for magnetic impurity problems. An estimate will also be given for the number of spin deviations contained in the ground state.

Since we are interested only in cases for which the fully aligned state is not the ground state, we shall assume throughout the appendix that the impurity–host exchange is antiferromagnetic ($J_0 < 0$). To simplify the analysis, we shall also assume that both impurity and host anisotropies are of the easy axis type ($K \geqslant 0$, $K_0 \geqslant 0$).

1. Bounds on the ground state energy

The energy of the 'Neél state' ($2S_0$ spin deviations on the impurity ion, none on the host ions) is an upper energy bound to the ground state of the Hamiltonian given by equation (2.1), according to the Ritz variational principle. From equation (2.1), we see that the upper energy bound E_U is given by

$$E_U = E_0 + 4J_0S_0S + 2\mu g_0HS_0, \tag{A.1}$$

where E_0 is the energy of the fully aligned state.

Since the ground state energy of a Hamiltonian can be no smaller than the sum of the lowest eigenenergies of its parts [12, 39], a lower energy bound may be obtained as a sum of the lowest eigenenergies of

$$\hat{\mathscr{H}}_{3J} = -J_0\hat{S}_0\cdot(\hat{S}_1 + \hat{S}_{N-1}) = -\tfrac{1}{2}J_0(\hat{\mathscr{S}}^2 - \hat{S}'^2 - \hat{S}_0^2) \tag{A.2}$$

and the remainder $\hat{\mathscr{H}}_{N-3J} = \hat{\mathscr{H}} - \hat{\mathscr{H}}_{3J}$. Here $\hat{S}' = \hat{S}_1 + \hat{S}_{N-1}$, is the sum of the spins of the neighbors to the impurity and $\hat{\mathscr{S}} = \hat{S}' + \hat{S}_0$ is the total spin of the three spin system. From equation (A.2) we see that H_{3J} commutes with $\hat{\mathscr{S}}^2$, $\hat{\mathscr{S}}^z$, $\hat{S}'^2$, and $\hat{S}_0^2$, so that the angular momentum vector coupling coefficients can be used to form the eigenstates of $\hat{\mathscr{H}}_{3J}$. We denote the eigenvalues of $\hat{\mathscr{S}}^2$, $\hat{\mathscr{S}}^z$, $\hat{S}'^2$ by $\mathscr{S}(\mathscr{S}+1)$, M and $S'(S'+1)$ respectively. The eigenstates $|\mathscr{S}, S', M\}$ are a complete orthonormal set of eigenstates of $\mathscr{H}_{3J}$, with eigenenergies

$$E_{3J}(\mathscr{S}, S', M) = -J_0[\mathscr{S}(\mathscr{S}+1) - S'(S'+1) - S_0(S_0+1)]/2. \tag{A.3}$$

Since in the fully aligned state $M = 2S + S_0$, the number of spin deviations m of the three-spin system is related to M as

$$m = 2S + S_0 - M. \tag{A.4}$$

From equation (A.3), we see that for each $\mathscr{S}$ and S' there is a degeneracy of $2\mathscr{S}+1$, since E_{3J} does not depend on M. This is because $\hat{\mathscr{H}}_{3J}$ is isotropic and independent of $\mathscr{S}^z$. Thus the eigenenergies of $\hat{\mathscr{H}}_{3J}$ are independent of the number of spin deviations.

Since $J_0 < 0$, the minimum of E_{3J} is obtained when $S' = 2S$ and $\mathscr{S} = |S' - S_0|$. Then equation (A.3) yields

$$E_{3J}(2S - S_0, 2S, M) = J_0(2S_0S + S_0) \tag{A.5}$$

and

$$E_{3J}(S_0 - 2S, 2S, M) = J_0(2S_0S + 2S), \tag{A.6}$$

Equation (A.5) gives the minimum when $S_0 \leqslant 2S$, equation (A.6), when $S_0 \geqslant 2S$. The lowest eigenstate of $\hat{\mathscr{H}}_{N-3J}$ is fully aligned, with energy

$$E_{N-3J} = E_0 + 2J_0S_0S. \tag{A.7}$$

Then from equations (A.5–A.7), we find the lower energy bound E_L, as

$$E_L = E_0 + 4J_0S_0S + \begin{cases} J_0S_0, & \text{for } S_0 \leqslant 2S \\ 2J_0,S, & \text{for } S_0 \geqslant 2S \end{cases} \tag{A.8}$$

2. Spin deviations in the ground state

Next we discuss the number of spin deviations contained in the ground state, for $H = K = K_0 = 0$. The ground state approaches the 'Neél state' with energy E_U (equation (A.1)) in the limit $J_0/J \to 0_-$, since in this limit the antiferromagnetic impurity–host exchange has no effect on the host spin alignment, and the energy is minimized by full alignment of the host spins and antiparallel alignment of the impurity spin. The ground state approaches the state with energy E_L (equation (A.8)) in the limit $J_0/J \to -\infty$, since in this limit the host–host exchange has no effect on the alignment of the impurity and its neighbors. The energy is now minimized by full alignment of the host spins, excluding the impurity and its two neighbors. The cluster consisting of the spins of the impurity and its neighbors assumes the spin state whose energy is E_{3J}, given by equations (A.5–A.6). Hence, for $J_0/J \to -\infty$, the ground state is degenerate unless $S_0 = 2S$, as seen from equations (A.4–A.6), containing $2S_0$ through $4S$ spin deviations. But states having other than $2S_0$ spin deviations are higher in energy for $J_0/J \to 0$, the lowest such state having the character of a spin wave. For values of J_0/J between 0 and $-\infty$, it is possible that an eigenstate of $\hat{\mathscr{H}}$ having other than $2S_0$ spin deviations may cross over the $2S_0$ spin deviation eigenstate and become lower in energy than the latter, but this is thought to be unlikely, since it would imply a special significance for the particular value of J_0/J for which the two states become equal in energy. If such a 'crossover' does not occur, the ground state contains $2S_0$ spin deviations for $H = K = K_0 = 0$ except in the limit $J_0/J \to -\infty$, at which point the $2S_0$ spin deviation eigenstate becomes degenerate with those having $2S_0$ through $4S$ spin deviations.

A quantity of physical interest is the expectation value of the spin of the impurity ion in state $|\mathscr{S}, S', M\}$. Using formulae from the theory of coupling of angular momenta [17], we obtain

$$\{\hat{S}_0^z\} = [\mathscr{S}(\mathscr{S} + 1) - S'(S' + 1) + S_0(S_0 + 1)]M/2\mathscr{S}(\mathscr{S} + 1). \tag{A.9}$$

In the ground state of $\hat{\mathscr{H}}_{3J}$, for which $S' = 2S$ and $\mathscr{S} = |S' - S_0|$, we have for the expectation value $\{\hat{S}_0^z\}_g$ of the impurity spin,

$$\{\hat{S}_0^z\}_g = -MS_0(2S - S_0 + 1)^{-1}, \qquad \text{for } S_0 \leqslant 2S, \tag{A.10}$$

and

$$\{\hat{S}_0^z\}_g = -M(S_0 + 1)(S_0 - 2S + L)^{-1}, \qquad \text{for } S_0 \geqslant 2S. \tag{A.11}$$

Setting the number of spin deviations $m = 2S_0$ in equations (A.12–A.14) ($M = 2S - S_0$) we obtain the expected asymptotic value of the spin deviation $S_0 - \{\hat{S}_0^z\}_g$ of the impurity ion in the ground state of $\hat{\mathscr{H}}$ in the limit $J_0/J \to -\infty$, given as

$$S_0 - \{\hat{S}_0^z\}_g = 2S_0 - S_0(2S - S_0 + 1)^{-1}, \qquad \text{for } S_0 \leqslant 2S, \tag{A.13}$$

and

$$S_0 - \{\hat{S}_0^z\}_g = 2S_0 - 2S(S_0 - 2S + 1)^{-1}, \qquad \text{for } S_0 \geqslant 2S, \tag{A.13}$$

for $m = 2S_0$.

Now consider the case of a finite applied field H and finite anisotropy constants K and K_0. The partitioning of $\hat{\mathscr{H}}$ into $\hat{\mathscr{H}}_{3J}$ and $\hat{\mathscr{H}}_{N-3J}$ used in Section A.1 is then inappropriate for discussing the number of spin deviations contained in the ground state, because the spin deviations on the neighbors of the impurity are then different for the ground states of $\hat{\mathscr{H}}_{3J}$ and $\hat{\mathscr{H}}_{N-3J}$. Instead, we write $\hat{\mathscr{H}} = \hat{\mathscr{H}}_3 + \hat{\mathscr{H}}_{N-3}$, with

$$\hat{\mathscr{H}}_3 = \hat{\mathscr{H}}_{3J} + \hat{\mathscr{H}}_{3H} + \hat{\mathscr{H}}_{3K}, \tag{A.14}$$

where

$$\hat{\mathscr{H}}_{3H} = -\mu H(g_0\hat{S}_0^z + g\hat{S}'^z), \tag{A.15}$$

and

$$\hat{\mathscr{H}}_{3K} = -K_0(\hat{S}_0^z)^2 - K[(\hat{S}_1^z)^2 + (\hat{S}_{N-1}^z)^2], \tag{A.16}$$

and where $\hat{\mathscr{H}}_{N-3}$ contains no anisotropy or Zeeman terms for the impurity or its neighbors and always has a fully aligned ground state in the presence of an applied field or easy axis anisotropy, in the z direction. Since $\hat{\mathscr{H}}_{3H}$ does not commute with $\hat{\mathscr{S}}^2$ unless $g_0 = g$ and $\hat{\mathscr{H}}_{3K}$ does not commute with $\hat{\mathscr{S}}^2$ unless $K_0 = K$, it is inconvenient to obtain the exact ground state of $\hat{\mathscr{H}}_3$ for general values of g_0, K_0, S, and S_0. We will first consider $\hat{\mathscr{H}}_{3H}$ and $\hat{\mathscr{H}}_{3K}$ as perturbation on $\hat{\mathscr{H}}_{3J}$, and then will give arguments based on physical grounds for the number of spin deviations contained in the ground state of $\hat{\mathscr{H}}$ when H, K, and K_0 become large.

First we consider the case $H > 0$ and $K = K_0 = 0$. The first order energy perturbation effect of $\hat{\mathscr{H}}_{3H}$ on the energy of the ground state of $\hat{\mathscr{H}}_{3J}$ is given by

$$E_{3H} = -\mu H[g_0\{\hat{S}_0^z\}_g + g(M - \{\hat{S}_0^z\}_g)], \tag{A.17}$$

the off diagonal matrix elements of $\hat{\mathscr{H}}_{3H}$ with respect to the degenerate ground states of $\hat{\mathscr{H}}_{3J}$ being zero. The minimum separation of any two energy levels of $\hat{\mathscr{H}}_{3J}$ is $|J_0|/2$ [17], so that this result is valid for $E_{3H} \ll |J_0|/2$. From equations (A.10), (A.11), and (A.17) we find

$$E_{3H} = -\mu HM[(2S + 1)g - S_0g_0](2S - S_0 + 1)^{-1}, \qquad \text{for } S_0 \leqslant 2S, \tag{A.18}$$

and

$$E_{3H} = -\mu HM[(S_0 + 1)g_0 - 2Sg](S_0 - 2S + 1)^{-1}, \qquad \text{for } S_0 \geqslant 2S. \tag{A.19}$$

E_{3H} is minimized by the maximum of M, $M = |S_0 - 2S|$, or the minimum of M, $M = -|S_0 - 2S|$, depending on the sign of the square bracket in (A.18) or (A.19). This removes the degeneracy of the ground state of $\hat{\mathscr{H}}_{3J}$ with respect to M. From

equations (A.4), (A.18) and (A.19), we see that for $S_0 < 2S$, the ground state of $\hat{\mathscr{H}}_3$ contains $2S_0$ spin deviations if $g_0/g \leqslant (2S-1)/S_0$ and $4S$ spin deviations if $g_0/g > (2S-1)/S_0$. For $S_0 \geqslant 2S$, the ground state of $\hat{\mathscr{H}}_3$ contains $2S_0$ spin deviations if $g_0/g \leqslant 2S/(S_0+1)$ and $4S$ spin deviations if $g_0/g > 2S/(S_0+1)$.

Following closely our discussion about the number of spin deviations contained in the ground state of $\hat{\mathscr{H}}$ for the case of $H = K = K_0 = 0$, in the limit $J_0/J \to 0$, we can conclude that the ground state will contain no more than $2S_0$ spin deviations, since the lowest state containing more than $2S_0$ spin deviations would be given by a spin wave-like state superimposed by the 'Neél state' in this limit, with additional energy $\sim \mu g H$. It is expected from the form of equations (A.15) that an applied field $H \gtrsim |J_0|/\mu g_0$ reduces the number of spin deviations in the ground state of $\hat{\mathscr{H}}$. For $S_0 \geqslant 2S$, the ground state of $\hat{\mathscr{H}}$ will contain no more than $2S_0$ spin deviations in the limit $J_0/J \to -\infty$, since the ground state of $\hat{\mathscr{H}}_3$ contains no more than $2S_0$ spin deviations. Again assuming no 'crossover' for J_0/J between the limits 0 and $-\infty$, we expect that for $S_0 \geqslant 2S$, the ground state of $\hat{\mathscr{H}}$ will contain no more than $2S_0$ spin deviations for arbitrary J, J_0, g, g_0, and H. The same conclusion is reached for $S_0 < 2S$, if $g_0/g \leqslant (2S+1)/S_0$, since in this case the ground state of $\hat{\mathscr{H}}_3$ contains $2S_0$ spin deviations. However, for $S_0 < 2S$ and $g_0/g > (2S+1)/S_0$, the $\hat{\mathscr{H}}_3$ ground state contains $4S$ spin deviations, so that one can expect that, for $H \ll |J_0|/\mu g_0$ and $|J_0| \gg J$, the ground state of $\hat{\mathscr{H}}$ will contain $4S$ ($4S > 2S_0$) spin deviations. This means, that, as $|J_0|/J$ is varied, a crossover occurs from a ground state with $2S_0$ to a ground state with $4S$ spin deviations. The larger g_0/g is, the smaller the value of $|J_0|/J$ will be at which the crossover occurs.

Finally, we consider the case $H > 0$, $K_0 > 0$, $K > 0$. The first order energy perturbation of $\hat{\mathscr{H}}_{3K}$ on the $\hat{\mathscr{H}}_{3J}$ ground state is given by

$$E_{3K} = -K_0\{(\hat{S}_0^z)^2\}_g - K\{(\hat{S}_1^z)^2 + (\hat{S}_{N-1}^z)^2\}_g, \tag{A.20}$$

the off-diagonal matrix elements being zero. This result is valid if $E_{3K} \ll |J_0|/2$. The expectation values in equation (A.20) may be found in a manner similar to that used to obtain equations (A.9–A.11). It is found that the M-dependent terms in E_{3K} are proportional to M^2 with positive coefficients [17], so that E_{3K} is always a minimum when $M = \pm|S_0 - 2S|$. Since E_{3H} is minimized by these same M values, in the first order of perturbation the argument concerning the number of spin deviations in the ground state of $\hat{\mathscr{H}}$ is unchanged.

From the form of equation (A.20), we see that the effect of $\hat{\mathscr{H}}_{3K}$ on the ground state is crudely to decrease the spin deviation of a spin S_j if its spin deviation is less than S_j and to increase the spin deviation if it is more than S_j. When the $\hat{\mathscr{H}}_{3H}$ perturbation is small, it is easily shown from equations (A.10) and (A.11) that in the $\hat{\mathscr{H}}_3$ ground state the impurity ion spin deviation is more than S_0, except that it is less than S_0 when the ground state contains $4S$ spin deviations, and that the spin deviation of the impurity neighbors is less than S. Thus, if the $\hat{\mathscr{H}}_3$ ground state contains more than $2S_0$ spin deviations, a large K_0 will tend to lower the number of spin deviations to $2S_0$. The energy of a spin wave-like state of $\hat{\mathscr{H}}$ in the presence of anisotropy is raised by $\sim(2S-1)K$, and for $|J_0| \ll J$, we expect only the impurity to have significant spin deviation. We conclude that the argument set forth concerning the ground state behavior in the presence of an applied field is little altered by the presence of anistropy. The ground state of $\hat{\mathscr{H}}$ is expected to contain no more than $2S_0$ spin deviations, except that it is expected to contain $4S$ spin deviations for $S_0 < 2S$, $g_0/g > (2S-1)/S_0$, $H \ll |J_0|/\mu g_0$, $|J_0| \gg J$, and $K_0 \ll |J_0|$.

REFERENCES

[1] Y. IZYUMOV, Advanc. Phys. *14*, 569 (1965).
[2] A. MARADUDIN, Solid State Phys. *18*, 273 (1966).
[3] A. MARADUDIN et al., Rev. Mod. Phys. *30*, 175 (1958).
[4] R. COWLEY and W. BUYERS, Rev. Mod. Phys. *44*, 406 (1972).
[5] I. M. LIFSHITZ, Usp. Fiz. Nauk. *83*, 617 (1964) [English transl.: Soviet Phys. Uspekhi *7*, 549 (1965)].
[6] T. WOLFRAM and J. CALLAWAY, Phys. Rev. *130*, 2207 (1963).
[7] H. ISHII, J. KANAMORI and T. NAKAMURA, Progr. Theoret. Phys. (Kyoto) *33*, 795 (1965).
[8] Y. IZYUMOV and M. MEDVEDEV, J. Eksp. Teor. Fiz. *51*, 517 (1967) [English transl.: Soviet Phys.—JETP *24*, 347 (1967)].
[9] Y. WANG and H. CALLEN, Phys. Rev. *160*, 358 (1967).
[10] J. PARKINSON, Solid State Commun. *5*, 419 (1967).
[11] P. ERDÖS and E. RHODES, Helv. Phys. Acta *41*, 785 (1968). The following corrections should be made in this reference:
Eq. (2)—replace $\hat{a}_i^+{}_{+1}\hat{a}_{i-1}$ by $\hat{a}_i^+{}_{+1}\hat{a}_{i+1}$. Eq. (14)—replace $\xi = -J/J_0$ by $\xi = -J_0/J$. Following Eq. (26), the fourth sentence should be deleted; in the fifth sentence, instead of 'In both cases, the...', read 'The...', in the sixth sentence instead of 'In our case, for instance, the...', read 'The...', the seventh and eighth sentence should read 'For $\eta \leqslant 0$, $\xi > 0$ and for energies at the bottom of the spin wave band, $c_0 \to 1$. This appears to be due to the negligibly small effect of the antiparallel alignment of the impurity spin. This, in turn, is a consequence of the negligibly small antiferromagnetic exchange between the impurity and its neighbors in comparison with the ferromagnetic exchange between the host spins'. On page 791, in Fig. 3b, replace '$J_0/J \leqslant 0$' by '$J_0/J \geqslant 0$'. In the table, for curve No. 4 omit $J_0/J \to 0_-$; for curve No. 7 replace '$\epsilon \to \infty + gh$' by '$\epsilon \to -\infty + gh$'. Replace Eq. $\alpha = 1 + \gamma\xi$ by $\alpha = (1 + \gamma\xi)^{-1}$. On page 792, line 18, replace 2a, b, and c by 2b, c, and d.
[12] T. OGUCHI and I. ONO, J. Phys. Soc. Japan *26*, 32 (1969).
[13] T. OGUCHI and I. ONO, J. Phys. Soc. Japan *33*, 1533 (1972).
[14] M. STEINER and W. KRÜGER, Solid State Commun. *9*, 227 (1971).
[15] M. STEINER and H. DACHS, Solid State Commun. *9*, 1603 (1971).
[16] J. TORRANCE, JR. and M. TINKHAM, Phys. Rev. *187*, 587 (1969).
[17] E. RHODES, Ph.D. dissertation, Florida State University 1972 (unpublished).
[18] A. MARADUDIN, in *Phonons and Phonon Interactions*, edited by T. BAK (W. A. Benjamin, N.Y., 1964).
[19] J. KLAUDER, Ann. Phys. (N.Y.) *14*, 43 (1961).
[20] Y. IZYUMOV, Proc. Phys. Soc. (London) *87*, 505 (1966).
[21] R. C. JONES, J. Phys. C: Solid State Phys. *4*, 2903 (1971).
[22] E. SHILES, Phys. Rev. B*6*, 1801 (1972).
[23] M. WORTIS, Phys. Rev. *132*, 85 (1963).
[24] D. MATTIS, *The Theory of Magnetism* (Harper and Row, N.Y. 1965).
[25] R. SILBERGLITT and J. TORRANCE, Phys. Rev. B 2, 772 (1970).
[26] T. TONEGAWA, Progr. Theoret. Phys. (Kyoto) *46*, Supplement, 61 (1970).
[27] T. OGUCHI, J. Phys. Soc. Japan *31*, 394 (1971).
[28] In the Appendix to the paper, we consider only easy axis host and impurity anisotropy in analyzing the number of spin deviations contained in the ground state. In Ref. [3], the case of easy plane impurity anisotropy combined with easy axis host anisotropy is studied for a system of five ions, each of spin one. In this case, the ground state is very sensitive to the magnitude of the host anisotropy, so that the ground state for the five ions is not indicative of the ground state for a large number of host ions, unless the impurity anisotropy is large compared to the host anisotropy.
[29] I. ONO and Y. ENDO, Phys. Letters *41A*, 440 (1972).
[30] T. HOLSTEIN and H. PRIMAKOFF, Phys. Rev. *58*, 1098 (1940).
[31] Even N is assumed, for odd N, there are $(N + 1)/2$ symmetric and $(N - 1)/2$ antisymmetric single spin deviation eigenstates, from which $(N + 1)^2/4$ symmetric and $(N^2 - 1)/4$ antisymmetric distinguishable double spin deviation states can be formed.
[32] This symmetry implies $c_{m(N/2)+j} = \pm c_{m(N/2)-j}$ for even N, which is assumed in equations (8.8–8.10). For odd N, there is no essential change, since the limit $N \to \infty$ will be taken, but for completeness, $c_{m[(N+1)/2]+j} = \pm c_{m[(N-1)/2]-j}$.

[33] Non-local states are those for which the spin deviation at each lattice site is of order N^{-1} or less, including the impurity site.

[34] The condition equation (10.8) is not essential to the calculation, but simplifies the numerical analysis by obviating the need to solve an integral equation for $E_l^{\downarrow}$. If it is not satisfied, high order terms in the perturbation expansion are probably necessary.

[35] The Oguchi and Ono approximation, which is a simple modification of the single spin deviation problem, is discussed in detail in Ref. [17].

[36] This procedure is more complicated for h_{c2} if uniaxial host and impurity anisotropies κ and ϕ are allowed, because the double spin deviation basic states then depend on κ and ϕ so that $\mathscr{E}' = \mathscr{E} - 2gh - 2(2S-1)\kappa$ depends on κ and ϕ through the matrix elements. Then a, simple analytic relation such as equation(11.4) does not exist in terms of $\eta = (g_0 - g)h + (2\gamma S - 1)\phi - (2S-1)\kappa$.

[37] I. MANNARI and C. KAWABATA, Research Note 15, Dept. of Physics, Faculty of Science, Okayama Univ. 1964 (unpublished).

[38] J. OITMAA, Solid State Commun. *9*, 745 (1971).

[39] H. EVEQUOZ and P. ERDÖS, unpublished results.

[40] P. ANDERSON, Phys. Rev. *83*, 1260 (1951).