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Spin configurations of ferromagnetic magnons in a periodic loop*†

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A periodic loop is considered with two magnetic electrons per atom occupying two degenerate bands with negligible bandwidth. The Hamiltonian considered contains terms representing the intra-atomic Hund's-rule coupling and the assumed ferromagnetic nearest-neighbor Heisenberg exchange interaction between electrons of the same and of different bands. The competing effects of these terms are studied in the exact solution of the spin-1 ferromagnon problem using a method due to Bethe. Both the energy spectra and the associated wave functions are obtained. It is found that the lowest-lying spin-wave excitations are characterized by a side-by-side spin-1 triplet pair configuration dominating over the on-site triplet configuration (rigid z-spin 1 on the atom), no matter how large the Hund's rule coupling.

I. INTRODUCTION

It is well known that the intra-atomic Hund's-rule coupling plays a dominant role in the formation of atomic magnetic moments for isolated atoms. It was suggested years ago by Slater that the Hund's-rule coupling may also be the decisive factor that is responsible for the magnetic ordering in narrow-band transition metals.[1] This point of view has been adopted by a number of authors.[2] The question is how a magnetic effect on the atom can be transferred from atom to atom so as to produce long-range correlations. There is first of all direct Coulomb interaction between electrons on different sites, producing the ferromagnetic spin-spin interaction, for example. There are also indirect effects, the main one of which comes from a band-hopping term used as a perturbation. The second-order virtual hopping between say nearest neighbors coupled with a Coulomb repulsion between electrons on an atom is enough to give an effective spin-spin interaction between neighbors.

Unfortunately, there are very few exactly solvable problems in this area. One can solve the hydrogen molecule, and similar small systems, and also certain one-dimensional systems.[2] Another exactly solvable problem is the half-filled one-band situation with nearest-neighbor interactions. We investigated this problem[3] by a method due to Bethe, in the saturated-ferromagnetic limit, F_{sat}, and found the electron-hole excitations without approximation. The effective spin interaction between sites arising from virtual hopping appears in the solution [see Eq. (5.10) of Ref. 3] for the lowest-energy branch, but the F_{sat} state is nondegenerate. The excitation itself was found to be a bound electron hole, with electron-hole distance falling off exponentially in the wave function. The hopping, etc., had therefore the twofold effect of (i) altering the effective spin-spin interaction constant J, and (ii) replacing the rigid atomic spin (electron "over" hole) by a bound state in which the electron strays from the hole, but only exponentially with distance. It was also found that by a proper choice of constants, the F_{sat} state was stable relative to these excitations (i.e., the excitation energies were non-negative). However, if the spin-spin interaction between sites was turned off, the F_{sat} state was *not* stable relative to these excitations. Thus in the half-filled one-band problem, the intra-atomic interactions do not promote ferromagnetism, even though the effective exchange between sites through hopping did occur. The reason for this is that the indirect effect is antiferromagnetic. It is possible that this special exact result is a symptom of a more general result, namely, that for ferromagnetism (as opposed to antiferromagnetism) the direct interatomic exchange is essential, even when the band is only partially filled, and even with band degeneracy.[4]

In a subsequent paper,[5] we investigated the problem of two degenerate bands, each band half filled. This is another exactly solvable problem, the F_{sat} state being nondegenerate. The results were similar to those in Ref. 3: When the spin-spin interaction between sites was turned off, the F_{sat} state is not stable relative to spin-flip excitations.

In the present paper,[6] we consider two spin flips (two electrons, two holes) in the two-band case. The interest in this problem is to determine the structure of the ferromagnon excitation when there are the two competing possibilities of a wave in which the z components of each atom's two electrons flip as a unit (an "on-site z-spin-1 state"), and of a wave in which z components of neighboring electrons flip as a unit (a "side-by-side z-spin-1 state"). The analogous rigid-spin problem was studied by Wortis.[7] In general the lowest-lying levels will be combinations of all configurations. We find that the on-site z-spin-1 configuration does not ever dominate, even in the limit of large Hund's-rule coupling. The side-by-side configuration *always* dominates. The lowest-lying excitations relative to the F_{sat} state will in fact be the *single* spin flips (discussed in Ref. 5) no matter how large the Hund's-rule coupling.

II. HAMILTONIAN AND WAVE FUNCTION

In order to simplify the problem somewhat, we shall eliminate the band terms, and assume that their effect is, as stated above in Refs. 4 and 5, mainly to change the effective exchange constants, and to make each "spin-$\frac{1}{2}$" a bound electron-hole pair with electron-hole distance probability varying exponentially with distance. What is left is a spin Hamiltonian as follows:

$$H=H_0+H_1, \tag{2.1}$$

where

$$H_0=-J_{12}\sum_{i=1}^{N}\vec{S}_i^{(1)}\cdot\vec{S}_i^{(2)}, \tag{2.2}$$

$$\begin{aligned} H_1=&-J\sum_{i=1}^{N}(\vec{S}_i^{(1)}\cdot\vec{S}_{i+1}^{(1)}+\vec{S}_i^{(2)}\cdot\vec{S}_{i+1}^{(2)}) \\ &-J'\sum_{i=1}^{N}(\vec{S}_i^{(1)}\cdot\vec{S}_{i+1}^{(2)}+\vec{S}_i^{(2)}\cdot\vec{S}_{i+1}^{(1)}). \end{aligned} \tag{2.3}$$

Here $\vec{S}_i^{(\lambda)}$ is the spin operator characterized by band index λ ($\lambda=1, 2$) and Wannier index i. These spin operators are in turn defined in terms of second-quantized Fermion operators

$$\begin{aligned} (S_i^{(\lambda)})^+&=(C_{i,+}^{(\lambda)})^\dagger(C_{i,-}^{(\lambda)}), \\ (S_i^{(\lambda)})^-&=(C_{i,-}^{(\lambda)})^\dagger(C_{i,+}^{(\lambda)}), \\ (S_i^{(\lambda)})^z&=\tfrac{1}{2}(n_{i,+}^{(\lambda)}-n_{i,-}^{(\lambda)}). \end{aligned} \tag{2.4}$$

The H_0 term represents the intra-atomic Hund's-rule coupling, and the H_1 term is the nearest-neighbor-exchange term. The J term in H_1 represents the exchange interaction between electrons of the same band, and the J' term represents the exchange between electrons of different bands.

When $J=J'$, the nearest-neighbor-exchange Hamiltonian H_1 can be written as

$$H_1=-J\sum_{i=1}^{N}\vec{S}_i\cdot\vec{S}_{i+1}, \tag{2.5}$$

where

$$\vec{S}_i\equiv\vec{S}_i^{(1)}+\vec{S}_i^{(2)}. \tag{2.6}$$

The Hamiltonian in Eq. (2.5) is of the form of a standard Heisenberg Hamiltonian. Here $\vec{S}_i$ is the total-spin operator associated with the ith lattice site; it is a spin-1 operator if it operates only on the triplet states.

Here we must explain what we mean by triplet or singlet states. Let us for the moment ignore the lattice index and concentrate on a particular lattice site. There are altogether four spin configurations:

$$\binom{+}{+},\quad \tfrac{1}{2}\sqrt{2}\left[\binom{-}{+}+\binom{-}{+}\right],\quad \binom{-}{-},\quad \tfrac{1}{2}\sqrt{2}\left[\binom{+}{-}-\binom{-}{+}\right]. \tag{2.7}$$

Here the upper and lower components of the column matrix indicate the spin configurations of the electron in band 1 and band 2, respectively. The first three configurations listed above are triplet states, and the last one is a singlet state. When we reintroduce the lattice indices, we can then talk about triplet or singlet states associated with a lattice site in terms of the corresponding Wannier functions.

Let us now look at the spin-flip states relative to the saturated ferromagnetic state, $|F_{\mathrm{sat}}\rangle$, which is the state with all lattice sites in the $\binom{-}{-}$ configuration. The other spin configurations can be generated from the F_{sat} state by applying a proper combinations of the spin-raising operators. The eigenvalue of H associated with F_{sat} is called E_0.

The one-spin-flip states are generated by applying the spin-raising operator on $|F_{\mathrm{sat}}\rangle$ once. From the complete set in Eq. (2.7), there are two one-spin-flip states: one triplet (t) and the singlet (s),

$$\begin{aligned} \Phi_f^{(t)}&\equiv[(S_f^{(1)})^+ +(S_f^{(2)})^+]\,|F_{\mathrm{sat}}\rangle\equiv\left[\binom{+}{-}+\binom{-}{+}\right]_f, \\ \Phi_f^{(s)}&\equiv[(S_f^{(1)})^+ -(S_f^{(2)})^+]\,|F_{\mathrm{sat}}\rangle\equiv\left[\binom{+}{-}-\binom{-}{+}\right]_f, \end{aligned} \tag{2.8}$$

where $f=1, 2, \ldots, N$. Here the notation used in the right-hand side means that only the f site has a spin flip, and that the other sites are all in the $\binom{-}{-}$ configuration. It can be easily shown that the set of all singlets spans an invariant subspace under the operation of the Hamiltonian in Eq. (2.1); the same is also true for the triplets. So we can solve the energy eigenvalue problems of the triplets and singlets separately. The lowest-energy solutions are the spin waves. There are two types of them: one corresponding to the propagation of a flipped triplet and the other to that of a flipped singlet. The dispersion relations of spin waves of wave vector $k=\pi n/N$, $n=1, 2, \ldots, N$, are

$$E_k^{(t)}-E_0=(J+J')[1-\cos k], \tag{2.9}$$

$$E_k^{(s)}-E_0=J_{12}+2J'+(J-J')[1-\cos k]. \tag{2.10}$$

It is clear from the above expression that both the Hund's-rule coupling and the interband nearest-neighbor exchange interaction contribute to the energy difference between the triplet-spin-wave and the singlet-spin-wave spectra.

We next look at the two-spin-flip states. The simplest way to analyze them is in the following terms: (i) states with two flips at f and g in the 1 band, $\binom{+}{-}_f\binom{+}{-}_g$, of which there are $\frac{1}{2}N(N-1)$ because of symmetry between f and g and because the $g=f$ case is not a state of the system; (ii) states with two spin flips at f and g in the 2 band, $\binom{-}{+}_f\binom{-}{+}_g$, of which again there are $\frac{1}{2}N(N-1)$; and (iii) states with a spin flip at f in band 1 and g in band 2,

$(\substack{-\\+})_f(\substack{+\\-})_g$, of which there are N^2. The total number of independent states is therefore $2N^2 - N$.

It is then possible to reformulate the basic states in terms of singlets and triplets as follows:

triplet pairs

$$\Phi^{(tt)}_{f,g} = [(S^{(1)}_f)^+ + (S^{(2)}_f)^+][(S^{(1)}_g)^+ + (S^{(2)}_g)^+]\,|\,F_{\text{sat}}\rangle; \tag{2.11a}$$

singlet pairs ($f \neq g$)

$$\Phi^{(ss)}_{f,g} = [(S^{(1)}_f)^+ - (S^{(2)}_f)^+][S^{(1)}_g)^+ - (S^{(2)}_g)^+]\,|\,F_{\text{sat}}\rangle; \tag{2.11b}$$

singlet-triplet mixtures ($f \neq g$)

$$S^{(st)}_{f,g} = [(S^{(1)}_f)^+ - (S^{(2)}_f)^+][(S^{(1)}_g)^+ - (S^{(2)}_f)^+]\,|\,F_{\text{sat}}\rangle. \tag{2.11c}$$

The $f = g$ state of a triplet pair is the state where two spin flips occur at the same site $(\substack{+\\+})$. There would be an identical configuration arising from the $f = g$ state of the singlet pairs, so it is removed from the latter category to avoid duplication and because it has the significance of a triplet, not a singlet. Notice that there are then $\frac{1}{2}N(N+1)$ triplet pairs (because of symmetry and inclusion of $f = g$ states), $\frac{1}{2}N(N-1)$ singlet pairs, and $N^2 - N$ singlet-triplet mixtures. The latter number comes about because there is no symmetry in f and g, but the $g = f$ states must not be counted because they are not states of the system. The total number of states is then $2N^2 - N$, just the same number that was counted in the previous paragraph.

The advantage of the singlet-triplet description is threefold. First, under the operation of H, the singlet-triplet mixtures are closed whereas the singlet pairs and triplet pairs although coupled are separated from the singlet-triplet mixtures. Thus we get a preliminary separation into two branches of states. Second, the triplet pairs (and singlet pairs) refer to sites only, and have no band index. They represent two spin excitations at two sites, and therefore are closely related to the usual concept of a spin deviation in the ordinary treatment of a Heisenberg Hamiltonian. Thus the two-magnon problem solved by Wortis[7] should be related to our triplet-pair states. Note that for $f = g$ we get an "on-site z-spin-1 state" on the atom. We wish ultimately to determine to what extent the lowest-lying excitations are composed of these on-site configurations as opposed to the "nearest-neighbor" ones. And third, when $J = J'$, the triplet pairs and singlet pairs themselves separate and form two simple closed systems under the action of H.

III. SINGLET PAIRS AND TRIPLET PAIRS

In this section we shall solve the energy eigenvalue problem in the subspace of the singlet pairs and triplet pairs. Consider

$$H\Psi = E\Psi, \tag{3.1}$$

where

$$\Psi = \sum_{f,g=1}^{N} [X_{f,g}\Phi^{(tt)}_{f,g} + Y_{f,g}\Phi^{(ss)}_{f,g}], \tag{3.2}$$

and where H is the Hamiltonian defined in Eq. (2.1). Here $X_{f,g}$ and $Y_{f,g}$ are coefficients to be determined. These coefficients must satisfy the periodic boundary conditions

$$\begin{aligned} X_{f,g} &= X_{f,g+N} = X_{f+N,g}, \\ Y_{f,g} &= Y_{f,g+N} = Y_{f+N,g}. \end{aligned} \tag{3.3}$$

They also have to satisfy the symmetry requirements

$$\begin{aligned} X_{f,g} &= X_{g,f}, \\ Y_{f,g} &= Y_{g,f}, \end{aligned} \tag{3.4}$$

since the functions $\Phi^{(tt)}_{f,g}$ and $\Phi^{(ss)}_{f,g}$ are, by definition, symmetric with respect to the interchange of indices f and g. For the convenience of later treatments, we shall assume N to be odd.

Substituting Eq. (3.2) into Eq. (3.1) and taking matrix elements with respect to various $\Phi^{(tt)}_{f,g}$ and $\Phi^{(ss)}_{f,g}$, we obtain the following sets of equations:

$$\begin{aligned} &(E - E_0 - 4\tilde{J})X_{f,g} + \tilde{J}(X_{f,g+1} + X_{f,g-1} + X_{f+1,g} + X_{f-1,g}) \\ &\quad + (\delta_{f,g+1} + \delta_{f,g-1})(\tilde{J}X_{f,g} + J''Y_{f,g}) - \tfrac{1}{2}\tilde{J}(\delta_{f,g+1}X_{f,g+1} + \delta_{f,g}X_{f,g-1}) + \delta_{f+1,g}X_{f+1,g} + \delta_{f-1,g}X_{f-1,g}) = 0, \end{aligned} \tag{3.5}$$

$$\begin{aligned} &(E - E_0 - 4\tilde{J} - 2J_{12})Y_{f,g} + J''(Y_{f,g+1} + Y_{f,g-1} + Y_{f+1,g} + Y_{f-1,g}) \\ &\quad + (\delta_{f,g+1} + \delta_{f,g-1})(\tilde{J}Y_{f,g} + J''X_{f,g}) - \tfrac{1}{2}J''[\delta_{f,g-1}X_{f,g-1} + \delta_{f,g-1}X_{f,g-1} + \delta_{f+1,g}X_{f+1,g} + \delta_{f-1,g}X_{f-1,g}] = 0, \end{aligned} \tag{3.6}$$

where

$$\tilde{J} \equiv \tfrac{1}{2}(J + J'), \tag{3.7}$$

$$J'' \equiv \tfrac{1}{2}(J - J'), \tag{3.8}$$

and where f, $g = 1, 2, \ldots, N$.

To solve the set of coupled equations in Eq. (3.5) and Eq. (3.6), we first separate out the exponential factor associated with the motion of the center of positions of the two flipped spins at f and g, i.e., consider solutions of the form

$$X_{f,g}=e^{iK(f+g)}X_r\,,\qquad Y_{f,g}=e^{iK(f+g)}Y_r\,, \tag{3.9}$$

where

$$K=n\pi/N\,,\quad n=1,\,2,\,\ldots,\,N\qquad \pm r=0,\,1,\,\ldots,\,N-1\,. \tag{3.10}$$

The symmetry conditions of Eqs. (3.3) and (3.4) are maintained by stipulating

$$X_{-r}=X_r\,,\quad e^{iKN}X_{N-r}\equiv X_r\,,\qquad Y_{-r}=Y_r\,,\quad e^{iKN}Y_{N-r}\equiv Y_r\,. \tag{3.11}$$

Notice that there are N K's, N assumed odd for convenience. Further, the number of independent X_r's is $\frac{1}{2}(N+1)$ and the number of independent Y_r's is $\frac{1}{2}(N-1)$. The total number of states is again N^2 in the singlet-pair triplet-pair branch.

Substituting Eq. (3.10) into (3.5) and (3.6), we obtain a pair of coupled equations,

$$(E-E_0-4\tilde{J})X_r+2\tilde{J}\cos K(X_{r+1}+X_{r-1}) +(\delta_{r+1,0}+\delta_{r-1,0})[\tilde{J}X_r+J''Y_r-\tilde{J}\cos K X_0)]=0\,, \tag{3.12a}$$

$$(E-E_0-4\tilde{J}-2J_{12})Y_r+2J''\cos K(Y_{r+1}+Y_{r-1}) +(\delta_{r+1,0}+\delta_{r-1,0})[J''X_r+\tilde{J}Y_r-J''\cos K X_0]=0\,. \tag{3.12b}$$

Equations (3.12a) are $\frac{1}{2}(N+1)$ independent equations in the $\frac{1}{2}(N+1)$ independent X_r, and Eqs. (3.12b) are $\frac{1}{2}(N-1)$ independent equations in the $\frac{1}{2}(N-1)$ unknowns Y_r, with coupling between them.

The most significant thing about these equations is that the Hund's-rule coupling coefficient J_{12} enters the singlet equation (3.12b), but not the triplet one. Thus in the lowest-lying states dominated by triplet configurations, J_{12} will not enter the energy directly. In fact when $J=J'$, the combination $J''=0$: Triplet states are completely separated from the singlets, and there will be *no* appearance by J_{12} in the lowest-lying excitations. This is therefore a special case of considerable interest; it is simple to handle, and contains the qualitative features of the general solution. It is treated in Sec. IV.

The fact that J_{12}'s direct appearance in the energy spectrum for the triplet-pair states is minimal does not mean that J_{12} has had no effect. The H_0 term in the Hamiltonian promotes triplets as opposed to singlets, and the fact that the triplets have lower energy is the effect of H_0. But H_0 does not distinguish between the three members of the triplet family. Thus $\Phi_{ff}^{(tt)}$ is an eigenstate of H_0 with the same energy, E_{00}, say, that F_{sat} has. But $\Phi_{fg}^{(tt)}$ is also an eigenstate of H_0 with this same energy. The first function $\Phi_{ff}^{(tt)}$ has spin 1 and z component 1; the second function $\Phi_{fg}^{(tt)}$ also has spin 1 but z component $\frac{1}{2}$ relative to F_{sat}. H_0 does not make a distinction between these states energywise.

This may seem strange, but it stems from the fact that H_0 is isotropic. If H_0 were replaced by the Ising interaction H_0, $z=-J_{12}\sum S_{i,z}S_{i+1,z}$, which does distinguish the z components, then indeed the on-site z-spin-1 state would be preferred. Both $\Phi_{ff}^{(tt)}$ and $\Phi_{fg}^{(tt)}$ are eigenstates of this operator, but the eigenvalue of the *fg* state is J_{12} higher than that of the other. So long as the Hund's-rule term is in isotropic Heisenberg form, as opposed to Ising form, it can make no distinction between a side-by-side triplet pair $\Phi_{fg}^{(tt)}$ $(f\neq g)$ and an on-site triplet $\Phi_{ff}^{(tt)}$. This is the reason that the triplet-pair spin waves are not much influenced by the Hund's-rule coupling.

IV. SPECIAL CASE $J=J'$

The case $J=J'$ is especially simple to handle, since H_0 commutes with H_1, and since the singlet-pair branch separates from the triple-pair branch. This is immediately visible from Eq. (3.12) in which we now must set $J''=0$:

$$(E-E_0-4J)X_r+2J\cos K(X_{r+1}+X_{r-1}) +(\delta_{r+1,0}+\delta_{r-1,0})J(X_r-X_0)\cos K)=0\,, \tag{4.1a}$$

$$(E-E_0-2J_{12}-4J)Y_r+(\delta_{r+1,0}+\delta_{r-1,0})JY_r=0\,. \tag{4.1b}$$

The two sets of equations are independent.

To solve Eq. (4.1b) first, we note that there are solutions of the type

$$E=E_0+2J_{12}+4J\,,\qquad Y_1=Y_{N-1}=0,\quad X_i=0\ldots\ (\text{all } i)\,, \tag{4.2}$$

and bound state solutions of the type

$$E=E_0+2J_{12}+3J\,,\qquad Y_i=0(i=2,\,3,\,\ldots,\,N-2),\ X_i=0(i=0,\,\ldots,\,N-1). \tag{4.3}$$

In Eq. (4.2) the $Y_2Y_3\ldots Y_{N-2}$ may be nonzero. Since there are $\frac{1}{2}(N-3)$ independent such Y's, there are $\frac{1}{2}(N-3)$ independent orthogonalizable states in this subbranch. In Eq. (4.3), only Y_1 and Y_{N-1} are nonzero. Only one of these is independent. There is one state of this type therefore. The result is that the singlet states number $\frac{1}{2}(N-1)$ as expected, and have energies as in Eqs. (4.2) and (4.3).

To solve Eq. (4.1a) we try, following Bethe,[8] the form

$$X_r=e^{ikr}+\beta e^{-ikr}\,,\quad r=1,\,2,\,\ldots,\,N-1\qquad X_0=\text{unspecified}\,. \tag{4.4}$$

Substitution into the $r=2,\,3,\,\ldots,\,N-2$ equations of (4.1a) we get the dispersion relation

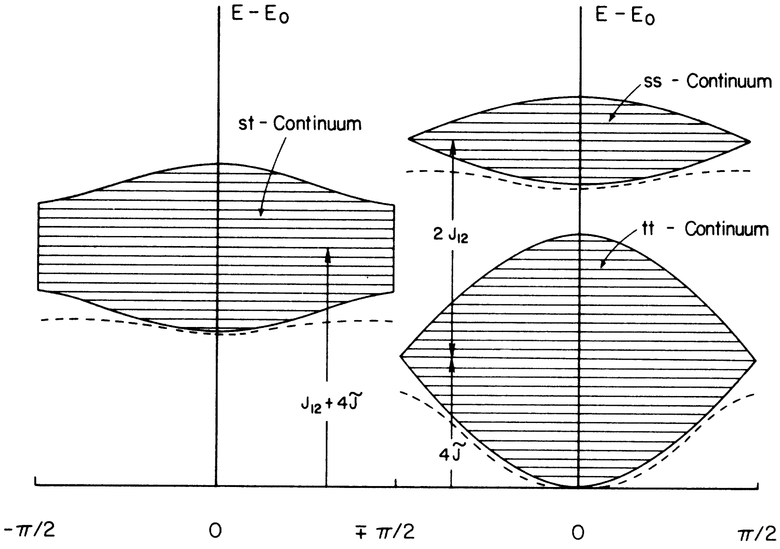

FIG. 1. This figure shows the structure of various continua and the associated bound states of all the two spin states. The *tt* continuum stands for the triplet-pair continuum, *ss* for singlet pairs, and *st* for singlet-triplet mixtures.

$$E_k = E_0 + 4J(1 - \cos K \cos k)\,. \tag{4.5}$$

There are three unknowns, X_0, β, and k, left to determine from the three remaining equations at $r = 1$, 0, $N-1$. These equations are the boundary conditions. Using the symmetry conditions $X_1 = e^{iKN}X_{N-1}$ and Eqs. (4.4) and (4.5), these three equations become two,

$$X_1 - X_0 \cos k = 0\,, \tag{4.6}$$

$$X_1 + X_0 \cos K - 2(1+\beta)\cos K = 0\,. \tag{4.7}$$

Further, in order for $X_{N-r} = e^{iKN}X_r$, Eq. (3.11), β is completely determined from (4.4) as

$$\beta = e^{i(k+K)N}\,. \tag{4.8}$$

Substitution of this into Eqs. (4.6) and (4.7), and eliminating X_0, gives an equation for k that can be written

$$\cot[\tfrac{1}{2}(k+K)N] = -\tan k \frac{\cos k + \cos K}{\cos k - \cos K}\,. \tag{4.9}$$

To proceed we allow k to be complex,

$$k = k_1 + i k_2\,. \tag{4.10}$$

where k_1 and k_2 are real. Then

$$\cos k = \cos k_1 \cosh k_2 - i \sin k_1 \sinh k_2\,. \tag{4.11}$$

From (4.5) we see that $\cos k$ must be real. Therefore either

$$\sin k_1 = 0\,, \tag{4.12a}$$

i.e., $k_1 = 0$ and k is pure imaginary or

$$\sinh k_2 = 0\,, \tag{4.12b}$$

i.e., $k_2 = 0$ and k is pure real. [The possibility $k_1 = \pi$ in (4.12a) can be shown not to be a solution.] Consider the latter case. This obviously represents the unbound continuum states. Examination of Eq. (4.9) shows $\frac{1}{2}(N+1)$ real roots, one of which is at $k = \pi$. However the value $k = \pi$ is not a state of the system, as can be seen from the fact that all the X_r's in Eq. (4.4) are zero (remember we have always been assuming N is odd), and X_0 is also zero from Eq. (4.6). Thus the real-k states available number $\frac{1}{2}(N-1)$. To complete the spectrum, there is the case of k pure imaginary, Eq. (4.12a). The total number is then $\frac{1}{2}(N+1)$.

To find this last state, we first of all notice that for it, $|\cos k| > 1$, and the energy of this state lies outside the continuum of wave states with real k as seen from (4.5). The energy lies, in fact, below the continuum of triplet states, and below the states in Eq. (4.2). For $J_{12} > J$ (reasonable physically), the energy lies below the one in (4.3) for all K.

The wave coefficients are

$$X_r = e^{-k_2 r} + e^{-k_2(N-r)} e^{iKN}\,. \tag{4.13}$$

Substitution of (4.3) into (4.9) gives after adjustment

$$e^{iKN} e^{-k_2 N} = -\frac{1 + e^{-2k_2} - 2e^{+k_2}\cos K}{1 + e^{+2k_2} - 2e^{-k_2}\cos K}\,. \tag{4.14}$$

There is no solution for negative k_2, since the right-hand side can never catch up to the left-hand side as k_2 increases. There is one solution for positve k_2. It can be determined in the limit as N goes to infinity, and the left-hand side vanishes. The equation is then related to the equation for two-magnon bound states using the Heisenberg Hamiltonian with spin 1.[9]

We recall from Eq. (3.2) that X_0 is the amplitude associated with the (↕) configuration, whereas X_1 is the largest amplitude associated with the side-by-side triplet. Thus

$$R_0 = |X_0/X_1|^2 \qquad (4.15)$$

gives the relative probability of these two configurations in the state found above. We notice immediately that it is independent of J_{12} [see below Eq. (3.12) for a discussion of this], and it is less than 1:

$$R_0 \cong 1/\cosh^2 k_2 < 1 \,. \qquad (4.16)$$

This means that the side-by-side configuration always dominates the wave function and the more firmly bound. This is a rather surprising result. We anticipate that it will be approximately the case even when $J \neq J'$, since the insignificance of J_{12} resulted from the fact that $\Phi_{fg}^{(tt)}$ and $\Phi_{ff}^{(tt)}$ are both eigenstates of H_0 with the same eigenvalue, a result still valid when $J \neq J'$.

It should be noted that the energy of the lowest triplet-pair branch given by $E_k = E_0 + 4J(1 - \cos K \cosh k_2)$ is higher at each K than the single triplet branch in Eq. (2.9). One might have expected that strong Hund's-rule coupling in the atom would make a flipped z-spin 1 on the atom cheaper energy wise than a flipped z-spin-$\frac{1}{2}$. But this is not the case. All H_0 does is promote triplets, but it does not distinguish z components.

V. GENERAL SOLUTION OF THE TRIPLET PAIRS AND SINGLET PAIRS

Now let us consider the general solution of Eqs. (3.12a) and (3.12b). We try solutions of the form[10]

$$X_r = a_x e^{ikr} + b_x e^{-ikr} \,, \qquad (5.1a)$$

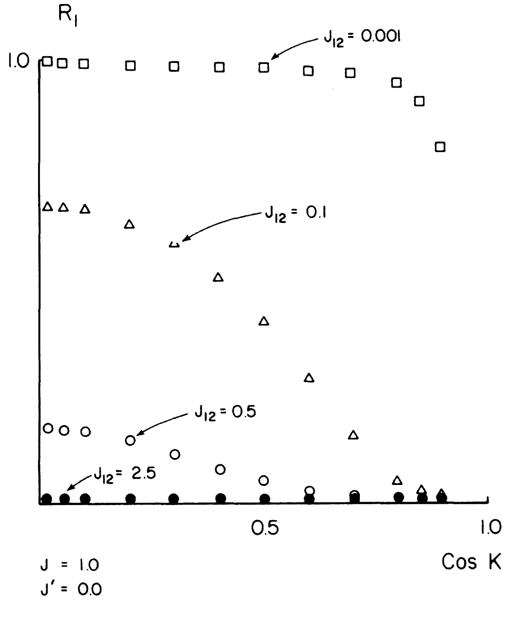

FIG. 2. This figure shows the dependence of the R_1 vs cos K curve on the value of J_{12}, with J and J' fixed.

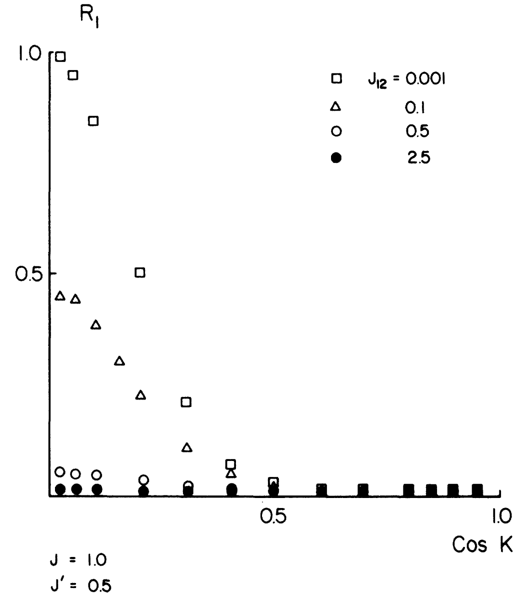

FIG. 3. This figure is similar to Fig. 2, except that the value of J' has increased.

$$Y_r = a_y e^{iqr} + b_y e^{-iqr} \,, \qquad (5.1b)$$

where $r = 1, 2, \ldots, (N-1)$. The corresponding dispersion relations are obtained from Eqs. (5.1) and (3.12), $r = 2, 3, \ldots, N-2$, as

$$E - E_0 + 4\tilde{J} + 4\tilde{J} \cos K \cos k = 0 \,, \qquad (5.2a)$$

$$E - E_0 - 4\tilde{J} - 2J_{12} + 4J'' \cos K \cos q = 0 \,. \qquad (5.2b)$$

It should be noticed that the wave numbers k and q are not independent. They are related through the dispersion relations in Eqs. (5.2a) and (5.2b),

$$\cos q = (\cos k + \lambda)/\mu \,, \qquad (5.3)$$

where

$$\lambda = J_{12}/2\tilde{J} \cos K \,, \quad \mu = J''/\tilde{J} < 1 \,. \qquad (5.4)$$

There are seven unknowns to be determined. They are k, q, a_x, a_y, b_x, b_y, and X_0. From Eq. (3.11), we get

$$b_x = a_x e^{ikN} e^{iKN} \qquad (5.5a)$$

and

$$b_y = a_y e^{iqN} e^{iKN} \,. \qquad (5.5b)$$

These are the analogs of Eq. (4.8). They eliminate two of the unknowns. There are five left, one of which is determined from the normalization of the wave function. A second condition is the relation (5.3) between k and q.

Thus we need three independent equations. One of these is the $r = 0$ equation of (3.12a). Using (5.3)-(5.5), we find

$$X_1 = X_0 \cos k \,. \qquad (5.6)$$

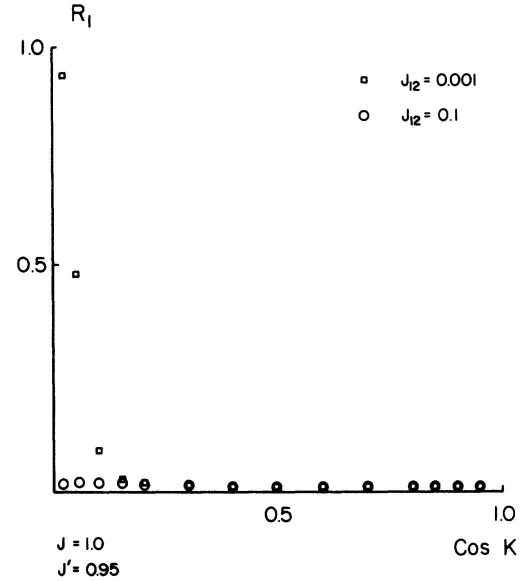

FIG. 4. This is a set of R_1 vs cos K curves, where $J-J'$ is very small; i.e., the nearest-neighbor part of the Hamiltonian is close to a standard Heisenberg Hamiltonian. Here we see that R_1 is almost negligible for all K even when J_{12} is very small.

The other two are the $r=1$ equations of Eqs. (3.12a) and (3.12b). They become

$$a_x[2\cos k\cos K(1+e^{iKN}e^{ikN}) - (\cos k+\cos K) \times (e^{ik}+e^{iKN}e^{ik(N-1)})] - a_y\mu\cos k(e^{iq}+e^{iKN}e^{iq(N-1)}) = 0\,, \quad (5.7a)$$

$$a_x\mu(\cos K-\cos k)(e^{ik}+e^{iKN}e^{ik(N-1)}) + a_y[2\mu\cos k\cos K(1+e^{iKN}e^{iqN}) - \cos k(e^{iq}+e^{iKN}e^{iq(N-1)})] = 0\,. \quad (5.7b)$$

The determinantal equation here is the generalization of Eq. (4.9), and reduces to that equation when $\mu = 0$.

Equation (5.7) contains the complete set of solutions to Eqs. (3.12). Let us examine the nature of the possible solutions. In general, for a given solution, both k and q can either be real or complex. For the sake of clarity, let us assume that

$$\lambda = J_{12}/2J\cos K > 2\,. \quad (5.8)$$

Under this restriction, it can be easily seen from Eq. (5.3) that k and q cannot be both real. We have three types of states.

(i) The wave number q is real, and hence k must be complex (pure imaginary). From Eq. (5.2b) we see that the energy of such a state lies within the envelope

$$|E - E_0 - 4\tilde{J} - 2J_{12}| \leq 4J''\cos K\,. \quad (5.9)$$

In this case the singlet-pair part of the wave function, the Y_r's, forms a simple wave with wave vector q, and the triplet part, the X_r's, decreases exponentially as r increases ($r/K \ll 1$). Such states obviously have some continuum character. We shall give them the name "singlet-pair continuum states." They are the generalization of Eq. (4.2).

(ii) The wave number k is real, and hence q must be complex. This energy of such a state lies within the envelope

$$|E - E_0 - 4\tilde{J}| \leq 4\tilde{J}\cos K\,. \quad (5.10)$$

In this case the triplet-pair part of the wave function is a simple wave, and the singlet-pair part of the wave function decreases exponentially as r increases. These are the generalization of the real-k solutions of Eq. (4.9).

(iii) Both k and q are complex. Let $k = k_1 + ik_2$, $q = q_1 + iq_2$, where k_1, k_2, q_1, q_2 are real numbers; then the location of the energy level of such a state is determined by the signs of $\cos k_1$ and $\cos q_1$. The energy level may lie below the "triplet-pair continuum" which is specified by the envelope in Eq. (5.10). In this case both $\cos k_1$ and $\cos q_1$ are positive. The energy level may also lie above the triplet-pair continuum but below the singlet-pair continuum. In this case both $\cos q_1$ is positive, but $\cos k_1$ is negative. In both cases X_r and Y_r decrease exponentially as r increases ($r/N \ll 1$, and $N \to \infty$). Such states are obviously bound states. (Actual calculations show that there is no state lying above

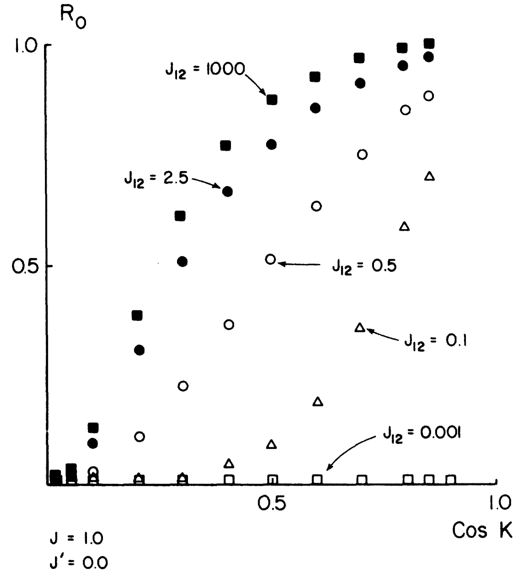

FIG. 5. This figure shows the dependence of the R_0 vs cos K curve on J_{12}. Notice that this set of curves approaches a "saturation curve" when J_{12} increases.

the singlet-pair continuum.) In Fig. 1, a schematic diagram of all the continua and bound-state spectra is given.

We are mainly interested in the properties of the bound states, in particular the lowest ones lying below the triplet-pair continuum. For actual calculation of the bound-state spectrum, we use the limit $N \rightarrow \infty$ such that the exponential factors e^{ikN} and e^{iqN} both vanish. The determinantal equation associated with Eqs. (5.7) then reduces to

$$\begin{vmatrix} 2\cos K\cos k - e^{ik}(\cos k + \cos K) & -\mu\cos k\, e^{iq} \\ \mu(\cos K - \cos k)e^{ik} & 2\mu\cos K\cos k - \cos k\, e^{iq} \end{vmatrix} = 0 . \tag{5.11}$$

Recall that k and q are related to the energy eigenvalue $(E - E_0)$ through Eqs. (5.2), so the energy eigenvalues can be directly obtained from Eq. (5.11).

In addition to the energy eigenvalues, we can also obtain the wave functions by finding a_y/a_x. There are two quantities of interest; they are

$$R_1 \equiv |Y_1/X_1|^2 , \tag{5.12}$$

$$R_0 \equiv |(X_0/X_1)|^2 . \tag{5.13}$$

The quantity R_1 is a measure of the mixing of the singlet pairs to the triplet pairs; a state with a small value of R_1 means that it is composed predominantly of the triplet pairs. The quantity R_0 has been introduced in previous discussions. It is the relative probability of the $(\substack{+\\+})$ configuration to the configuration which has two one-spin-flip triplets sitting side by side.

Both R_0 and R_1 can be expressed in terms of k and q through Eqs. (5.6) and (5.1). The explicit expressions are

$$R_0 = 1/\cos^2 k , \tag{5.14}$$

$$R_1 = |e^{iq}a_y/e^{ik}a_x|^2 , \tag{5.15}$$

where

$$\frac{a_y}{a_x} = \frac{2\cos K\cos k - e^{ik}(\cos k + \cos K)}{\mu(\cos k)e^{iq}} . \tag{5.16}$$

For a given set of parameters $\{J, J', J_{12}\}$, and a given K, Eq. (5.11) can be solved with the aid of a computer to obtain the energy eigenvalues. The corresponding R_0 and R_1 can also be calculated. The bound-state solutions are discussed in the summary, and the results of numerical calculations for certain values of the parameters are shown in Figs. 2-7.

It can be seen immediately for complex k and q that $\cos^2 k > 1$ whence $R_0 < 1$. Thus the side-by-side triplet configurations still dominate the on-site triplet configuration in the lowest levels.

VI. SINGLET-TRIPLET MIXTURE

In this section we shall solve the eigenvalue problem of two-spin-flip excitations in the subspace of the singlet-triplet mixtures defined in Eq. (1.11); i.e., consider

$$H\Phi = E\Phi , \tag{6.1}$$

where

$$\Phi = \sum_{f,g} Z_{f,g}\Phi^{(st)}_{f,g} \cdots Z_{f,f} = 0 \tag{6.2}$$

and where the $Z_{f,g}$'s are coefficients to be determined. These coefficients must satisfy the periodic boundary conditions,

$$Z_{f,g+N} - Z_{f+N,g} = Z_{f,g} , \quad f,\ g = 1, 2, \ldots, N . \tag{6.3}$$

To solve this problem we calculate $H\Phi^{(st)}_{f,g}$ and then substitute the result into Eq. (6.1). Shifting indices under the summation sign and noticing the linear independence of various $\Phi^{(st)}_{f,g}$'s, we arrive at

$$\begin{aligned}(E - E_0 - J_{12} - 4\tilde{J})Z_{f,g} &+ \tilde{J}(Z_{f,g+1} + Z_{f,g-1}) \\ &+ \tilde{J}''(Z_{f+1,g} + Z_{f-1,g}) \\ &+ (\delta_{f,g+1} + \delta_{f,g-1})(\tilde{J}Z_{f,g} + J''Z_{g,f}) = 0 .\end{aligned} \tag{6.4}$$

To solve Eq. (6.4), we again consider solutions of the form

$$\begin{aligned} &Z_{f,g} = e^{iK(f+g)}Z_r , \quad r \equiv f - g \\ &Z_{r\pm N} = Z_r e^{-iKN} , \\ &Z_0 = 0 ,\end{aligned} \tag{6.5}$$

where $K = \pi n/N$, $n = 1, 2, \ldots, N$. Substitute Eq. (6.5) into Eq. (6.4) and simplify the result. We then get

$$\begin{aligned}(E - E_0 - J_{12} - 4\tilde{J})Z_r &+ J\cos K[Z_{r+1} + Z_{r-1}] \\ &- iJ'\sin K[Z_{r+1} - Z_{r-1}] \\ &+ (\delta_{r+1,0} + \delta_{r-1,0})[\tilde{J}Z_r + J''Z_{N-r}] = 0 , \\ r = 1, 2, \ldots, N - 1 . &\end{aligned} \tag{6.6}$$

Equation (6.6) can again be solved by Bethe's method. For $r \neq 1$, $(N - 1)$, Eq. (6.6) can be satisfied by solutions of the form

$$Z_r = e^{i(\varphi+k)r} + \gamma e^{i(\varphi-k)r} , \quad r = 1, 2, \ldots, (N-1) , \tag{6.7}$$

with corresponding dispersion relation

$$E - E_0 - J_{12} - 4\tilde{J} + \xi\cos k = 0 , \tag{6.8}$$

where

$$\xi \equiv 2[J^2\cos^2 K + J'^2\sin^2 K]^{1/2} \tag{6.9}$$

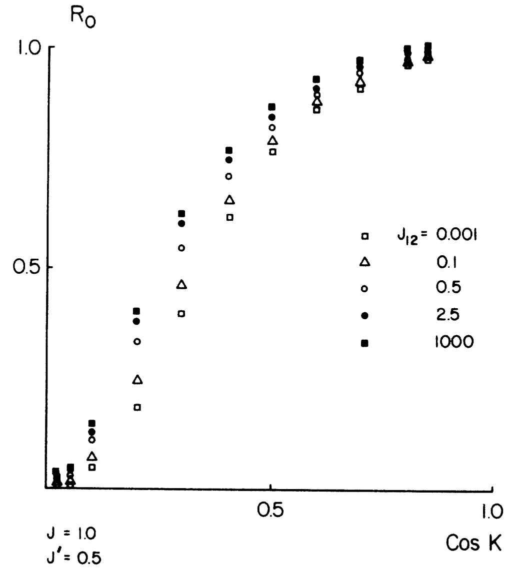

FIG. 6. This figure is similar to Fig. 5 except that the value of J' has increased. The result is a faster saturation.

and where φ is specified by

$$\cos\varphi \equiv 2J\cos K/\xi\,, \quad \sin\varphi \equiv 2J'\sin K/\xi\,. \tag{6.10}$$

The wave vector k and the coefficient γ are then determined by the boundary equations $r=1,\ N-1$. Setting $r=1$ and $r=(N-1)$ in Eq. (6.6) and simplifying the results, we obtain

$$\xi e^{i\varphi}(1+\gamma) - 2(\tilde{J}Z_1 + J''Z_{N-1}) = 0\,, \tag{6.11}$$

$$\xi e^{i(N-1)\varphi}(e^{ikN} + \gamma e^{-ikN}) - 2(\tilde{J}Z_{N-1} + J''Z_1) = 0\,. \tag{6.12}$$

From Eq. (6.8) we see that "singlet-triplet continuum" is specified by the envelope

$$|E - E_0 - J_{12} - 4\tilde{J}\,| \le 2[J^2\cos^2 K + J'^2\sin^2 K]^{1/2}\,. \tag{6.13}$$

Thus the singlet-triplet continuum lies right in between the single-pair continuum and the triplet-pair continuum discussed in Sec. V.

A schemetic diagram of the energy spectrum of the singlet-triplet mixture is also contained in Fig. 1. Bound states can again be obtained by considering the complex wave vector in the limit N approaches infinity as was done in Sec. V. We shall not repeat the details here.

VII. SUMMARY

Using the Hamiltonian in Eq. (2.1), we have solved the eigenvalue problem of the two-spin-flip excitations relative to the F_{sat} state exactly. There are three types of two-spin-flip states: the triplet pairs, the singlet pairs, and the triplet-singlet mixtures. Under the operation of the Hamiltonian considered, the singlet-triplet mixtures can be solved separately from the other two types of states.

The eigenvalue problem in the subspace of the singlet-triplet mixtures is solved in Sec. VI. The solutions contain both continuum and bound states. The energy gap between the center of the continuum and the E_0 level is $(J_{12}+2J+2J')$. The envelope of the continuum is

$$|E - E_0 - J_{12} - 4\tilde{J}\,| \le 2[J^2\cos^2 K + J'^2\sin^2 K]^{1/2}\,. \tag{7.1}$$

Bound states are attached to the bottom of this continuum at $K=0$. The half-width of the continuum at $K=0$ is $2J$, so there is a gap of at least $(J_{12}+2J')$ between the entire spectrum and the E_0 level.

The coupled eigenvalue problem of the singlet pairs and triplet pairs is solved in some detail in Secs. III–V. The results show that there are two distinct continua. One of these is centered at a level $(2J_{12}+4\tilde{J})$ from E_0. This one is called the singlet-pair continuum, because the singlet-pairs part of the wave functions of the states lying in this continuum are simple waves. The envelope of this continuum is given by

$$|E - E_0 - (2J_{12}+4\tilde{J})| \le |4J''\cos K|\,. \tag{7.2}$$

There are bound states associated with this continuum. The other continuum, the triplet continuum, is centered at a level $4\tilde{J}$ from E_0. The envelope of this continuum is given by

$$|E - E_0 - 4\tilde{J}\,| \le |4\tilde{J}\cos K|\,, \tag{7.3}$$

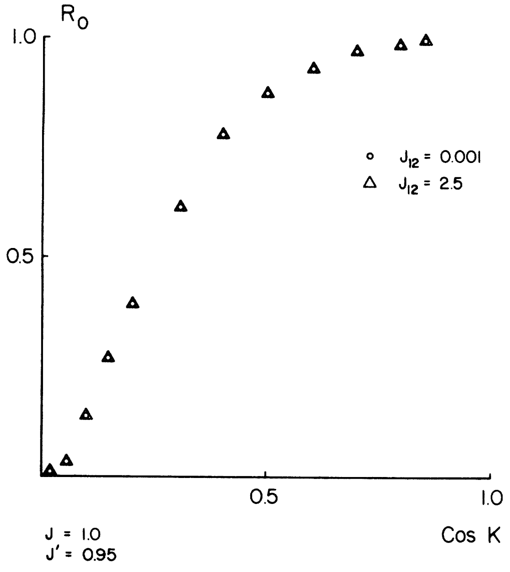

FIG. 7. This figure shows the R_0 vs cos K curves when $J-J'$ is very small. Notice that the curves are almost independent of the value of J_{12}.

So the bottom of this continuum touches the E_0 level at $K=0$. The bound states lying below the triplet-pair continuum are the lowest-lying states. These are the states we pay particular attention to. Both the energy spectrum and the wave functions of the lowest-lying bound states can be found. It is the wave function that reveals much of the physical information. We studied two quantities of interest: $R_1 \equiv |Y_1/X_1|^2$ and $R_0 \equiv |X_0/X_1|^2$, defined in Eqs. (5.12) and (5.13). The ratio R_1 is a measure of the mixing ratio of the singlet pairs relative to the triplet pairs. For a given state, the greater R_1 is, the greater is the singlet-pair contribution to the composition of this state. The ratio R_0 is the relative probability of the $(^{\uparrow}_{\downarrow})$ spin configuration to the spin configuration that represents two one-spin-flip triplets sitting side by side. The knowledge of R_0 gives information concerning the contribution of the $(^{\uparrow}_{\downarrow})$ configuration to the two-magnon bound states.

To discuss the behavior in some more detail, we reduce the results for the lowest-lying spin-wave branch. From Eqs. (5.11) and (5.3) we can determine $k\,(=ik_2)$ and $q\,(=iq_2)$, and then

$$R_0 = (1/\cosh k_2)^2\,, \tag{7.4}$$

$$R_1 = \mu^2 \frac{[1-\cos K/\cosh k_2]^2}{[2J_{12}\tilde{J}^{-1} + 2\cos K(2\cosh k_2 - \mu e^{-q_2}) - 1]^2}\,. \tag{7.5}$$

The equations determining k_2 and q are

$$2\mu \cosh q_2 = 2\cosh k_2 + \lambda\,, \tag{7.6}$$

$$(e^{2k_2}\cos K - \cosh k_2)(2\mu e^{q_2}\cos K - 1) = \mu^2(\cosh k_2 - \cos K)\,. \tag{7.7}$$

[The branch with $k=\pi + ik_2$ gives Eq. (7.7) but with $\cosh k_2$ replaced by $-\cosh k_2$.]

From Eq. (7.7) we see that in the $\mu = 0$ limit the right-hand side is zero. In this limit there are two branches of roots to the left-hand side. The first factor gives the triplet-pair bound state of Eq. (4.14), the second factor gives the singlet-pair bound state[10] of Eq. (4.3).

Another limit of interest is $J_{12}\to\infty$. This implies that $\lambda\to\infty$, and, from Eq. (7.6), that $q_2\to\infty$. From Eq. (7.7) this gives back again the triplet-pair bound-state condition that was found when $\mu=0$: $e^{2k_2}\cos K - \cosh k_2 = 0$. Thus the solution for k_2 as a function of K saturates as J_{12} gets large. Further, the saturated values of k_2 for $K=0$ is $k_2=0$, and the value for $K=\pi/2$ is $k_2=\infty$. The quantity $\cos K \cosh k_2$ that appears in the energy, Eq. (5.2a), varies from 1 at $K=0$ to $\frac{1}{4}$ at $K=\pi/2$.

By substituting these results in Eqs. (7.4) and (7.5), we see that as J_{12} gets large, R_0 also saturates, the saturated value going from 1 to 0 as K goes from 0 to $\pi/2$. Similarly, R_1 vanishes in the limit. Thus for large J_{12} and arbitrary μ the lowest-lying branch has no singlet contribution and is dominated by the side-by-side triplet-pair configuration rather than by the on-site configuration.

As J_{12} gets smaller, down to the order of $\tilde{J}$, both R_0 and R_1 remain always less than 1 but the shapes of the curves as functions of K vary somewhat.

As μ gets smaller and finally vanishes, we see from Eq. (7.5) that R_1 vanishes. This is consistent with the discussion apropos of Eq. (3.12), in which it was seen that when $J''=0$, the coupling between singlet and triplet pairs disappears. The decoupling of singlet and triplet pairs occurs if either $J_{12}\to\infty$ or $J=J'$.

The results for intermediate values of the parameters are exhibited in Figs. 2–7. Thus the main result of this calculation is that in the model considered, the lowest-lying spin-wave branch of excitations is dominated by side-by-side triplet-pair configurations. The model depends on there being two bands, each half full, and the on-site interaction being of the isotropic Heisenberg type. If the interaction is not isotropic, the above-derived results are not necessarily valid although the method used here is applicable. And in the case where there are say three degenerate bands, but only two electrons per atoms, it is hard to see how to apply the method used in this paper, since the ferromagnetic reference state is degenerate. It would be interesting to see if the result found here for the one-electron-per-atom-per-band model is carried over to the general case of more than one band per electron per atom, but a different approach would have to be used.

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