Multimagnon excitations in alternating spin/bond chains

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The nature of multimagnon excitations in alternating spin/bond ferromagnetic chains are studied using a combination of scaling methods and the recursion method. Two-magnon excitations for a Heisenberg chain with either alternating spin magnitudes, S and S', or alternating nearest-neighbor couplings, J_1 and J_2 can be studied using real-space rescaling techniques. These results are then used to investigate three-magnon excitations using the recursion method. [S0163-1829(98)02038-6]

I. INTRODUCTION

In recent years various quasi-one-dimensional magnetic systems have been synthesized. These systems include chains in which metal ions alternate regularly with organic radicals,[1] ordered bimetallic chains,[2] and donor-acceptor electron-transfer salts.[3] Much of the interest has been focused on systems with antiferromagnetic ground states[4] and the nature of their excitation spectra. Many of these materials can be described in terms of isotropic Heisenberg exchange interactions which may alternate in strength and sign along the chain. The chains are often composed of two sublattices which have unequal spin magnitudes S and S' and magnetic measurements[5] indicate that these materials can be antiferromagnets, ferrimagnets or ferromagnets. In a previous paper,[6] we have studied the nature of one- and two-magnon excitations in an alternating bond/spin ferromagnetic chain for the case of general isotropic near-neighbor interactions. Both a direct analytic method and second method based on a scaling transformation were used to study the relationship of bound state branches to the scattering state continua. In the present work we extend our study to three-magnon excitations in alternating systems which can be described by an isotropic Heisenberg exchange interaction between near neighbors. Although the one- and two-magnon problems can be reduced to single-particle problems,[7,8] this is not possible for three-magnon excitations. The general m-magnon problem in the case of the uniform bond spin $S=\frac{1}{2}$ chain has been solved exactly using the Bethe ansatz[9,10] but this method cannot generally be used for higher spin or for alternating systems except in very special cases.[11–15] We use the recursion method[16] to transform the three-magnon equations into a tridiagonal form which then provides a continued fraction representation for the calculation of the density of states. The spin and/or bond alternation is responsible for gaps in the three-magnon continuum and this fact makes the termination of the continued fraction more complicated than for uniform chains. However, knowledge of the one- and two-magnon spectra can be used to predict the location of the gap edges and this is all that is needed to implement the termination procedure.

In Sec. II we describe the model and discuss the two-magnon excitations. In Sec. III we derive the three-magnon equations in a basis of three-spin deviations, and in Sec. IV we describe the recursion method and the termination procedure for the resulting continued fraction representation of the Green's function. Section V describes our results and Sec. VI summarizes our findings.

II. THE MODEL

We consider a chain composed of two nonidentical sublattices, each being uniform and homogeneous. The Hamiltonian for these two nonidentical one-dimensional sublattices with alternating spin magnitudes, S and S', and alternating nearest-neighbor interactions can be expressed as

$$\hat{H}=-\sum_{n=1}^{N/2}\left[J_1(\vec{S}'_{2n}\cdot\vec{S}_{2n+1})+J_2(\vec{S}_{2n+1}\cdot\vec{S}'_{2n+2})\right], \quad (1)$$

where the total number of sites of the chain N is even and J_1, J_2 represent the interactions which alternate in strength along the chain. $\vec{S}'_{2n}$ and $\vec{S}_{2n+1}$ are quantum spin operators at the even and odd sites, respectively, and they satisfy the usual commutation relations. We impose periodic boundary conditions to ensure translational invariance within sublattices and thus the total wave vector K is a good quantum number and restricted to the range $|K|\leqslant\pi/2a$ where a is the lattice spacing which will be set equal to unity in what follows. Both the total spin $\vec{S}^2_{\rm tot}$ and the component in the z direction $S^z_{\rm tot}$ are constants of motion. The ferromagnetic state in which all spins are aligned along some arbitrary direction is an exact eigenstate of $\hat{H}$ with eigenvalue $E_0=-(N/2)SS'(J_1+J_2)$. We denote this state which has a z component of total spin $S^z_{\rm tot}=(N/2)(S+S')$ by $|0\rangle$ and we take this to be our reference state.

The excitations relative to this reference state can be classified according to the total amount of reduction in the z component of the total spin, $S^z_{\rm tot}=(N/2)(S+S')-m$, and such a state is called a m-magnon excitation. The general problem is to solve the Schrödinger equation

$$\hat{H}|\psi_m\rangle=E_m(K)|\psi_m\rangle \quad (2)$$

for the excitation energies $E_m(K)$ of the m-magnon state as a function of total wave vector K.

A. One-magnon excitations

A general one-magnon state can be written as

$$|\psi_1\rangle=\sum_{n=1}^{N/2}[a_{2n}|2n\rangle+a_{2n+1}|2n+1\rangle], \tag{3}$$

where the ket $|n\rangle$ represents the state with the z component of the nth spin reduced by one unit relative to the reference state. The Schrödinger equation $\hat{H}|\psi_1\rangle=E_1|\psi_1\rangle$ results in equations relating the neigboring amplitudes a_r

$$\begin{aligned}[E_1-S(J_1+J_2)]a_{2n}&=-\sqrt{SS'}(J_1a_{2n+1}+J_2a_{2n-1}),\\ [E_1-S'(J_1+J_2)]a_{2n+1}&=-\sqrt{SS'}(J_1a_{2n}+J_2a_{2n+2}),\end{aligned} \tag{4}$$

where E_1 is measured relative to the reference state energy E_0. The eigenvalue E_1 can be written as

$$E_K^{\mu}=B+\frac{1}{2}\mu\sqrt{\chi^2+4|\nu_K|^2}, \tag{5}$$

where

$$\nu_K=\sqrt{SS'}(J_1e^{iK}+J_2e^{-iK}), \tag{6}$$

$$B=\frac{1}{2}(S+S')(J_1+J_2), \tag{7}$$

and

$$\chi=(S-S')(J_1+J_2). \tag{8}$$

The solutions for these excitations are characterized by real wave vectors. The index $\mu=\pm1$ labels the two branches which by convention are referred to as ''optic'' for the upper branch and ''acoustic'' for the lower branch and the dimensionless wave vector K lies in the range $0-\pi/2$. In general, there is a nonzero gap between the two branches at the Brillouin-zone boundary ($K=\pi/2$)

$$E_{\rm gap}=2\sqrt{B^2-4SS'J_1J_2}. \tag{9}$$

This gap vanishes only in the uniform case where $S=S'$ *and* $J_1=J_2$. Hence, an important difference between uniform and nonuniform ferromagnetic chains is the presence of gaps in the excitation spectrum. Note that the gap discussed here has nothing to do with the Haldane gap[17,18] between the ground and excited states which also appears in some alternating chains.

B. Two-magnon excitations

The two-magnon states $|\psi_2\rangle$ can be written as

$$\begin{aligned}|\psi_2\rangle=\sum_{n\leqslant m}[&a_{2n,2m}|2n,2m\rangle+a_{2n,2m+1}|2n,2m+1\rangle\\ &+a_{2n-1,2m}|2n-1,2m\rangle+a_{2n+1,2m+1}|2n+1,2m+1\rangle],\end{aligned} \tag{10}$$

where the ket $|r,s\rangle$ with $r<s$ represents the state with single deviations on the rth and sth spins relative to the reference state $|0\rangle$, while the ket $|r,r\rangle$ represents the state with two spin deviations on the same (rth) site. As in the one-magnon problem, we consider the two-magnon Schrödinger equation $\hat{H}|\psi_2\rangle=E_2|\psi_2\rangle$, where E_2 is the two-magnon excitation energy measured relative to the reference state energy E_0. The equations relating the various amplitudes are obtained by applying the Hamiltonian (1) to the general form of the wave function (10) and then equating the coefficients of each basis ket. The resulting equations can be artificially grouped into two sets. One set involves amplitudes with spin deviations separated by at least two sites ($m>n$) that we will refer as the ''noninteracting equations.'' The other set will be called the ''interacting equations'' and will involve amplitudes with spin deviations on the same or neighboring sites. It can easily be shown[6,19,20] that the energy eigenvalues are simply the sum of the energy of two noninteracting magnons

$$E_2(k_1,k_2)=E_{K,q}^{\mu_1,\mu_2}=E_{k_1}^{\mu_1}+E_{k_2}^{\mu_2}, \tag{11}$$

where k_1 and k_2 are the wave vectors of the individual magnons, μ_1 and μ_2 label the branches of the single magnon dispersion curves. The total wave vector $K=k_1+k_2$ and the relative wave vector $q=(k_1-k_2)/2$ can also be used to label the energies. Translational invariance requires K to be real but k_1 and k_2 can be complex.

For real values of k_1 and k_2, or equivalently, for real values of K and q, there are three energy regions which form three different energy continua due to the gap in the one magnon dispersion curve. Depending on the values used for μ_1 and μ_2, they can be identified as ''acoustic-acoustic,'' ($\mu_1=\mu_2=-1$) ''optic-optic,'' ($\mu_1=\mu_2=+1$) or ''mixed-mode'' ($\mu_1=-\mu_2=\pm1$).

The knowledge of the solution of the one-magnon problem allows us to determine the regions in the $E_2(K)$ versus K plane where scattering state solutions corresponding to two free magnons are found. These continua correspond to solutions in which both individual wave vectors k_1 and k_2 are real. However, by considering the possibility of solutions with q complex, we also find the existence of ''bound state'' solutions outside of these regions. These solutions can be found numerically by solving a 4×4 eigenvalue problem or using a real space rescaling procedure.[6,19] The energy regions where the scattering states are located are determined by the one-magnon spectrum and the bound states are found outside these continua. Similarly, the complete two-magnon spectrum, scattering plus bound states, will determine the location of the continua for the three-magnon problem.

III. THREE-MAGNON EXCITATIONS

A general three-magnon state can be written as

$$|\psi_3\rangle=\sum_{i\leqslant j\leqslant k}a_{ijk}|ijk\rangle, \tag{12}$$

where we define an orthonormal set of three spin deviation states

$$|i,j,k\rangle=C_{ijk}S_i^-S_j^-S_k^-|0\rangle \tag{13}$$

with C_{ijk} being the coefficients normalizing these states and satisfying

$$C_{2i,2j,2k}=\begin{cases}\dfrac{1}{\sqrt{8S'^3}}, & i\neq j\neq k,\\ \dfrac{1}{\sqrt{8S'^2(2S'-1)}}, & \text{two of } i,j,k \text{ equal},\\ \dfrac{1}{\sqrt{24S'(2S'-1)(S'-1)}}, & i=j=k,\end{cases} \tag{14}$$

$$C_{2i,2j,2k+1}=\begin{cases}\dfrac{1}{\sqrt{8S'^2S}}, & i\neq j,\\ \dfrac{1}{\sqrt{8SS'(2S'-1)}}, & i=j.\end{cases} \tag{15}$$

In order to obtain the coefficients with two odd indices and one even index or with three odd indices, we need only to exchange S with S'.

We consider the effect of the Hamiltonian on the complete set of states in the $m=3$ basis in the same way as was done for $m=1$ and $m=2$. The translational invariance property of the Hamiltonian can be taken into account by first transforming the states $|i,j,k\rangle$ into center of mass and relative coordinates

$$|i,j,k\rangle\equiv|j;x,y\rangle, \tag{16}$$

where

$$x=j-i\geqslant 0,$$
$$y=k-j\geqslant 0. \tag{17}$$

There are two groups of kets $|j;x,y\rangle$ for any pair of values (x,y) corresponding to each sublattice or equivalently to odd or even values of j. We define the Fourier transforms with respect to the center of mass of each group as follows:

$$|e,K;x,y\rangle=\sqrt{\frac{2}{N}}\sum_{r=0}^{N/2-1}e^{-\imath K(2r-x/3+y/3)}|2r;x,y\rangle,$$
$$|o,K;x,y\rangle=\sqrt{\frac{2}{N}}\sum_{s=0}^{N/2-1}e^{-\imath K(2s+1-x/3+y/3)}|2s+1;x,y\rangle, \tag{18}$$

where e and o stand for the ''even'' and ''odd'' sublattice, respectively.

The effect of the Hamiltonian on these states can be summarized by the following two equations:

$$(E_3-\varepsilon^{K,e}_{x,y})|e,K;x,y\rangle+\kappa^e_{x,y}|e,K;x,y+1\rangle+\rho^e_{x,y}|e,K;x+1,y\rangle+\vartheta^e_{x,y}|e,K;x-1,y\rangle+\sigma^e_{x,y}|e,K;x,y-1\rangle$$
$$+\tau^e_{x,y}|o,K;x-1,y+1\rangle+\lambda^e_{x,y}|o,K;x+1,y-1\rangle=0, \tag{19}$$

$$(E_3-\varepsilon^{K,o}_{x,y})|o,K;x,y\rangle+\kappa^o_{x,y}|o,K;x,y+1\rangle+\rho^o_{x,y}|o,K;x+1,y\rangle+\vartheta^o_{x,y}|o,K;x-1,y\rangle+\sigma^o_{x,y}|o,K;x,y-1\rangle$$
$$+\tau^o_{x,y}|e,K;x-1,y+1\rangle+\lambda^o_{x,y}|e,K;x+1,y-1\rangle=0, \tag{20}$$

where the first equation represents the action of the Hamiltonian on an ''even'' ket and the second equation corresponds to an ''odd'' ket. In this representation, the even and odd kets can be visualized as a semi-infinite lattice of points in the first quadrant of the xy plane. Three distinct groups of kets can be identified according to whether they lie in the ''bulk'' $(x,y\geqslant 2)$, on a ''surface'' $(x\geqslant 2,y\leqslant 1$ or $x\leqslant 1,y\geqslant 2)$ or on both surfaces $(x,y\leqslant 1)$.

When the Hamiltonian acts on a ket in the bulk (both $x,y\geqslant 2$), we have a set of equations that we shall refer to as ''noninteracting'' and the coefficients in Eq. (19) have the form shown in Table I. The notation used for the coefficients in this table and for those that will follow, allow for a straightforward transfer of an equation for an even ket [with general form (19)] to an equation for an odd ket [with general form (20)]. The coefficients are defined as follows:

$$\varepsilon=S(J_1+J_2), \tag{21}$$
$$\varepsilon'=S'(J_1+J_2),$$
$$\varepsilon_0=\varepsilon_3=\varepsilon_6=3\varepsilon,$$
$$\varepsilon_1=2\varepsilon+\varepsilon'-2J_1,$$
$$\varepsilon_2=\varepsilon+2\varepsilon'-J_1-J_2,$$
$$\varepsilon_4=2\varepsilon+\varepsilon',$$
$$\varepsilon_5=2\varepsilon+\varepsilon'-J_2,$$
$$w_0=-\kappa J_1\sqrt{3S(S'-1)},$$

TABLE I. Coefficients when the Hamiltonian is applied to an even ket with both $x,y\geqslant 2$.

	Noninteracting coefficients						
x,y	$\varepsilon^{K,e}_{x,y}$	$\kappa^e_{x,y}$	$\rho^e_{x,y}$	$\vartheta^e_{x,y}$	$\sigma^e_{x,y}$	$\tau^e_{x,y}$	$\lambda^e_{x,y}$
$2l,2m$	ε_6	w	$\bar{w}^*$	w	$\bar{w}^*$	$\bar{w}^*$	w
$2l,2m+1$	ε_4	$\bar{w}$	$\bar{w}^*$	w	w^*	$\bar{w}^*$	w
$2l+1,2m$	ε_4	w	w^*	$\bar{w}$	$\bar{w}^*$	$\bar{w}^*$	w
$2l+1,2m+1$	ε'_4	$\bar{w}$	w^*	$\bar{w}$	w^*	$\bar{w}^*$	w

TABLE II. Coefficients when the Hamiltonian is applied to an even ket. One of the values of x and y is $\leqslant 1$ while the other is $\geqslant 2$.

Two-bound and one-free magnon coefficients							
x,y	$\varepsilon^{K,e}_{x,y}$	$\kappa^{e}_{x,y}$	$\rho^{e}_{x,y}$	$\vartheta^{e}_{x,y}$	$\sigma^{e}_{x,y}$	$\tau^{e}_{x,y}$	$\lambda^{e}_{x,y}$
$0,2m$	ε_3	w	$\overline{w_2^*}$		$\overline{w}^*$		w_2
$0,2m+1$	ε_4	$\overline{w}$	$\overline{w_2^*}$		w^*		w_2
$2l,0$	ε_3	w_2	$\overline{w}^*$	w		$\overline{w_2^*}$	
$2l+1,0$	ε_4	w_2	w^*	$\overline{w}$		$\overline{w_2^*}$	
$1,2m$	ε_5	w	w^*	$\overline{w_2}$	$\overline{w}^*$	$\overline{w_2'}^*$	w
$1,2m+1$	ε_5'	$\overline{w}$	w^*	$\overline{w_2}$	w^*	$\overline{w_2'}^*$	w
$2l,1$	$\overline{\varepsilon_5}$	$\overline{w}$	$\overline{w}^*$	w	w_2^*	$\overline{w}^*$	w_2'
$2l+1,1$	$\overline{\varepsilon_5'}$	$\overline{w}$	w^*	$\overline{w}$	w_2^*	$\overline{w}^*$	w_2'

$$w_2 = -\kappa J_1 \sqrt{S(2S'-1)},$$

$$w_4 = -\kappa J_1 \sqrt{(2S-1)(2S'-1)},$$

$$w = -\kappa J_1 \sqrt{SS'},$$

and $\kappa = e^{iK/3}$. The presence of a prime on a coefficient indicates that S should be replaced by S' and vice versa, whereas the presence of a bar indicates that J_1 should be replaced by J_2 and vice versa in relation to the corresponding coefficient without the bar or the prime. As usual, an asterisk (*) indicates that the complex conjugate of the coefficient should have been taken. For example, the coefficient ε_4' (appearing in Table I) is equal to $2\varepsilon' + \varepsilon$ once the proper replacements of S and S' are made in the definition of ε_4. Similarly, $\overline{w} = -\kappa J_2 \sqrt{SS'}$ once J_1 is replaced by J_2 in the definition for w. The coefficients corresponding to the action of the Hamiltonian on the odd kets are easily obtained by taking the even coefficients and then adding or removing their primes or bars depending whether they are present or not. This is a direct result of the fact that one sublattice has the same equations as the other, only with the spins S,S' and bonds J_1,J_2 exchanged. As an example, the coefficients obtained when the Hamiltonian acts on the odd ket $|o,K;2l,2m\rangle$ are $(\varepsilon_6', \overline{w}', w'^*, \overline{w}', w'^*, w'^*, \overline{w}')$, where we have applied these rules to the first row of coefficients in Table I.

The second group of equations corresponds to the case where either x or y is $\leqslant 1$ while the other is $\geqslant 2$ and we will refer to this group as the two-bound one-free magnon group. The coefficients are given in Table II. The final group of equations is obtained when the Hamiltonian is applied to a ket $|e,K;x,y\rangle$ with both $x,y \leqslant 1$, i.e., for the case of three magnons on the same site or on nearest-neighbor sites. We will refer to this group as the three-bound magnon group. Table III gives the coefficients.

These three groups of equations are of course coupled and the complete solution of the three-magnon problem involves finding the solutions of all of these groups together. There are two types of scattering state solutions to the complete set of equations. The first type has energy eigenvalues that can be written as the sum of the energy of three noninteracting magnons

TABLE III. Coefficients when the Hamiltonian is applied to an even ket with both $x,y \leqslant 1$.

Three-bound magnon coefficients							
x,y	$\varepsilon^{K,e}_{x,y}$	$\kappa^{e}_{x,y}$	$\rho^{e}_{x,y}$	$\vartheta^{e}_{x,y}$	$\sigma^{e}_{x,y}$	$\tau^{e}_{x,y}$	$\lambda^{e}_{x,y}$
$0,0$	ε_0	w_0	$\overline{w_0^*}$				
$0,1$	ε_1	$\overline{w}$	$\overline{w_2^*}$		w_0^*		w_4
$1,0$	$\overline{\varepsilon_1}$	w_2	w^*	$\overline{w_0}$		$\overline{w_4^*}$	
$1,1$	ε_2	$\overline{w}$	w^*	$\overline{w_2}$	w_2^*	$\overline{w_2'}^*$	w_2'

$$E_3(K) = E^{\mu_1}_{k_1} + E^{\mu_2}_{k_2} + E^{\mu_3}_{k_3}, \tag{22}$$

where k_1,k_2,k_3 are all real and $K = k_1 + k_2 + k_3$. Figure 1(a) shows the one-magnon excitation energies obtained from Eq. (5) for the case $S = 2S' = 1$ and $J_1 = 2J_2 = 1$ and Fig. 1(b) shows the corresponding three-free continua. The gaps are a consequence of the presence of the energy gap between the optic and acoustic branches of the one-magnon dispersion curve for all values of the wave vector. The lowest continuum is due to the combination of three acoustic (A) branches ($\mu_1 = \mu_2 = \mu_3 = -1$) and is labeled as ''AAA''; the highest continuum is due to the combination of three optic (O) branches ($\mu_1 = \mu_2 = \mu_3 = +1$) and is labeled as ''OOO.'' The other two continua are mixed combinations

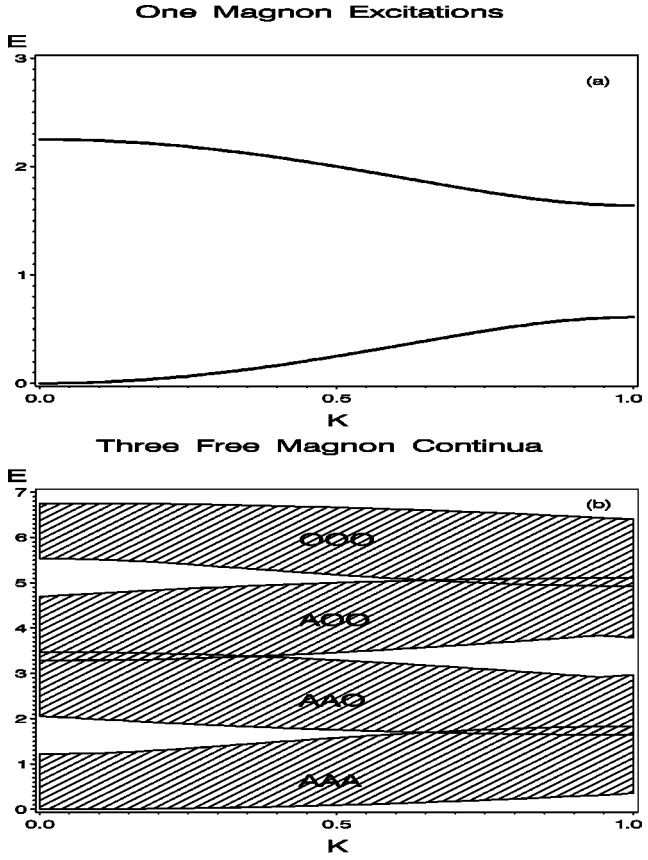

FIG. 1. (a) One magnon excitation branches (solid curves) and (b) the corresponding three-free magnon scattering state continua (shaded regions) for an alternating ferromagnetic chain with $S = 2S' = 1$ and $J_1 = 2J_2 = 1$. The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

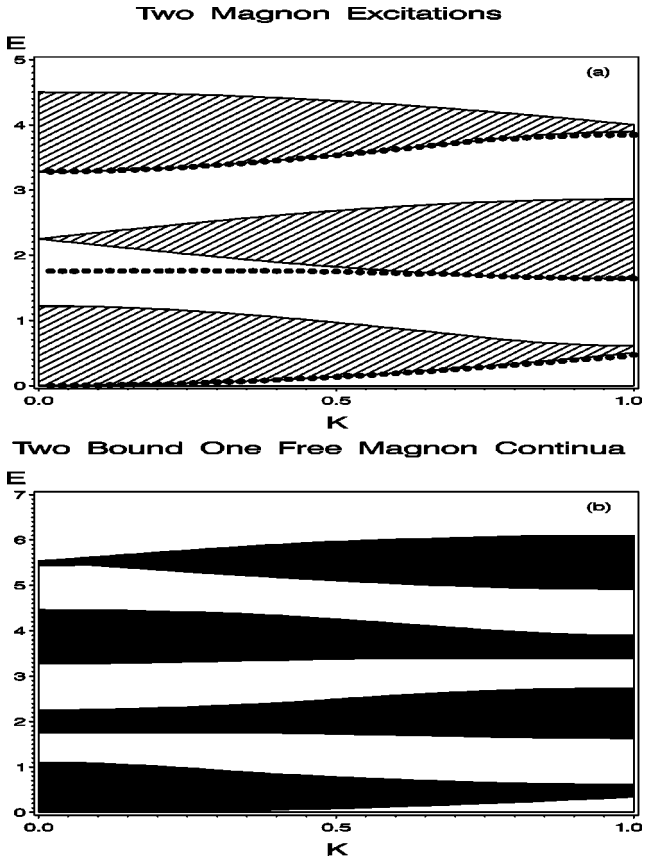

FIG. 2. (a) Two-bound magnon state branches (solid dots) with the two magnon scattering state continua (shaded regions) and (b) the two-bound one-free magnon scattering state continua (solid regions) for an alternating ferromagnetic chain with $S=2S'=1$ and $J_1=2J_2=1$. The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

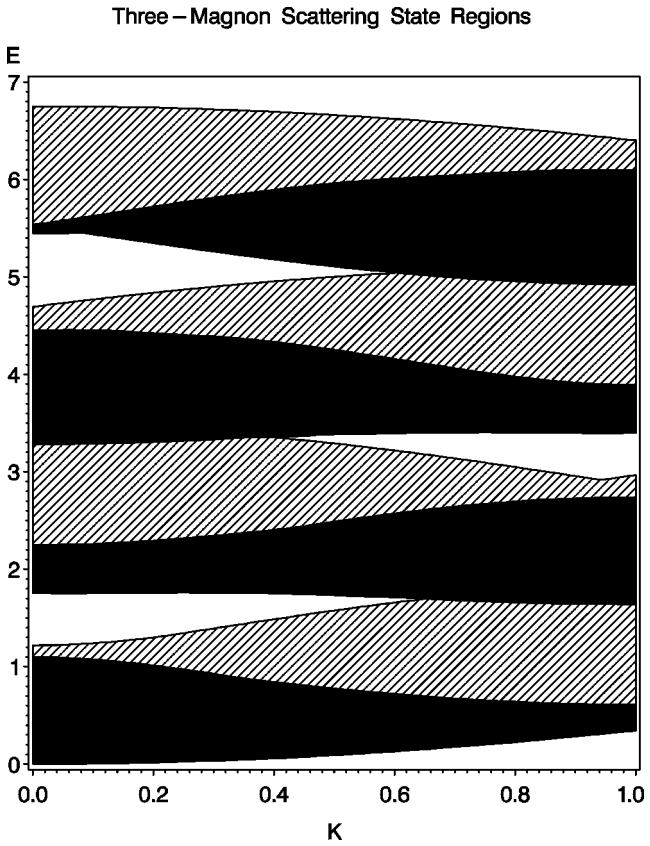

FIG. 3. Three-magnon scattering state continua for an alternating ferromagnetic chain with $S=2S'=1$ and $J_1=2J_2=1$. The results of the three-free (shaded regions) and two-bound one-free continua (solid regions) are superimposed. The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

and are labeled as ''AAO'' and ''AOO.'' As the differences between S and S' or J_1 and J_2 become larger, the gaps between the continua increase in magnitude and width.

The second type of continuum solution has eigenvalues that can be written as

$$E_3(K)=E_2(k_1,k_2)+E_{k_3}^{\mu_3}. \tag{23}$$

The term $E_2(k_1,k_2)$ corresponds to the energy of two bound magnons at the real wave vector k_1+k_2 and hence these solutions have complex values of k_1 and k_2 but a real value of k_3. Still, these eigenstates correspond to scattering states in which one magnon is free and two are bound. They form continua which overlap with the three-free magnon scattering state continua described above. Figure 2(a) shows the two-magnon excitation spectrum for the same case ($S=2S'=1, J_1=2J_2=1$) obtained using the methods described by Medved, Southern, and Lavis[6] and Fig. 2(b) shows the corresponding two-bound one-free magnon continua. Figure 3 shows the superposition of the three-free and two-bound one-free scattering state continua. In this example case, there are two gaps at small values of K, three gaps at intermediate values of K and one gap for values of K near the Brillouin-zone boundary. At any fixed value of the total wave vector K, the combined continua exhibit a varying number of gaps. The number of gaps depends on the specific values of S, S', J_1, and J_2. However, the energies of the gap edges are completely determined by the corresponding one- and two-magnon spectra.

In addition to these scattering state solutions, there are also three-magnon bound state energies which are discrete and are found outside the continua. It is only in certain special integrable cases[8] that the groups completely decouple from one another and the bound states then have special features. In the two-magnon subspace,[6] the problem of solving the combined set of equations (scattering plus bound states) can be reduced to a numeric implementation of a 4×4 matrix eigenvalue problem. A similar approach to resolving the three-magnon problem is not possible, as we would have to find the eigenvalues of an infinite matrix. In the next section, we will present a different approach which will give us a direct method of identifying bound states for the three-magnon problem.

IV. RECURSION METHOD

The recursion method[16,21,22] can be used to obtain spectral information about any Hamiltonian. The basic idea is to transform the Hamiltonian to a tridiagonal form so that a continued fraction representation of the local Green's function can be obtained. The local Green's function is defined by

$$G_j(E+\imath\delta)=\langle j|(E-\hat{H}+\imath\delta)^{-1}|j\rangle \tag{24}$$

where δ is a small positive imaginary part in the energy E and $|j\rangle$ is an arbitrary ket in the three-magnon basis. We will start with the basis kets $\{|e,K;x,y\rangle,|o,K;x,y\rangle\}$ and construct a new basis, $\{|\boldsymbol{v}_n\rangle\}$, in which the Hamiltonian assumes a tridiagonal form. The complete orthonormal set of states $\{|\boldsymbol{v}_n\rangle\}$ satisfies a three-term recursion relation

$$\hat{H}|\boldsymbol{v}_n\rangle = a_n|\boldsymbol{v}_n\rangle + b_{n+1}|\boldsymbol{v}_{n+1}\rangle + b_n|\boldsymbol{v}_{n-1}\rangle, \tag{25}$$

where $a_n, b_n \in \mathrm{Re}$. Hence the new matrix representation of the Hamiltonian in this basis is

$$\hat{H} = \begin{pmatrix} a_0 & b_1 & & & \\ b_1 & a_1 & b_2 & & 0 \\ & b_2 & a_2 & b_3 & \\ 0 & & & \ddots & \end{pmatrix} \tag{26}$$

and the Green's function for the initial ket $\{|\boldsymbol{v}_0\rangle\}$ is given by the continued fraction[16]

$$G_0(E) = \cfrac{1}{E - a_0 - \cfrac{b_1^2}{E - a_1 - \cfrac{b_2^2}{E - a_2 - \cfrac{b_3^2}{\ddots}}}}. \tag{27}$$

In carrying out the procedure described above, we choose an initial ket (or linear combination of kets) in the three-magnon basis and generate the coefficients a_n, b_{n+1} up to some maximum value of $n = n_{\max}$. The asymptotic behavior of these coefficients as function of n depends upon the scattering state spectrum. If at a particular value of total wave vector K, there are no gaps, then the coefficients will approach constant values asymptotically. However, if there are one or more gaps present, then the asymptotic behavior is oscillatory.[22] In practice, we need only to calculate the coefficients up to some suitable value of $n_{\max}$ and terminate the continued fraction using our knowledge of the scattering state spectrum as follows:

$$G_0(E) = \cfrac{1}{E - a_0 - \cfrac{b_1^2}{\vdots \cfrac{}{E - a_{n_{\max}} - b_{n_{\max}+1}^2 G_0^{\infty}(E)}}}, \tag{28}$$

where $G_0^{\infty}(E)$ is the terminator which represents the asymptotic terms. In the case of no gaps in the scattering state continuum, the coefficients a_n and b_n converge to constant values,

$$\left.\begin{aligned} a_n &= a \\ b_{n+1} &= b \end{aligned}\right\} \quad \text{for} \quad n > n_{\max} \tag{29}$$

and the terminator of the continued fraction satisfies

$$G_0^{\infty}(E) = \cfrac{1}{E - a - \cfrac{b^2}{E - a - \cfrac{b^2}{\ddots}}} = \frac{1}{E - a - b^2 G_0^{\infty}(E)}. \tag{30}$$

Solving for G_0^{∞}, we have

$$G_0^{\infty}(E) = \frac{E - a \pm \sqrt{(E-a)^2 - 4b^2}}{2b^2}. \tag{31}$$

This is known as the square-root terminator and the choice of the positive or negative square root depends on whether E is less than or greater than a. The terminator determines the analytic properties of G_0. For example, G_0 is complex in the region of E where the argument of the square root is negative and this corresponds to the scattering state continuum. However, G_0 can also have isolated poles outside this energy region due to the other terms in the continued fraction and these energies are the bound states.

In the case of the square-root terminator, $G_0^{\infty}(E)$ is complex when

$$E_{\min} = a - 2b \leqslant E \leqslant a + 2b = E_{\max}. \tag{32}$$

Hence the asymptotic values of a_n and b_n(a and b, respectively) are related to the minimum energy $E_{\min}$ and maximum energy $E_{\max}$ of the continuum by

$$a = \frac{1}{2}(E_{\min} + E_{\max}),$$

$$b = \frac{1}{4}(E_{\max} - E_{\min}). \tag{33}$$

If, at a particular value of K, the three-magnon continuum has no gaps, we can terminate the continued fraction using Eqs. (31) and (33). However, the coefficients (a_n, b_{n+1}) of the continued fraction exhibit undamped oscillations if gaps are present. Turchi *et al.*[22] generalized the termination procedure for the calculation of the tail of the continued fraction when the continuum has multiple gaps. In the present work, the spin/bond alternation leads to several gaps in the three-magnon continuum depending on the spin magnitudes. For given values of S, S', J_1 and J_2, we use the one-magnon results to determine the three-free continua in the energy (E_3) versus wave vector (K) plane. We then use the two-bound magnon energies and the one-magnon excitation energy to determine the two-bound one-free continua in this plane. These continua are then superimposed to determine the number of gaps in the energy continua and the gap edges at any value of the total wave vector K so that the appropriate terminator can be used to study the density of states for the three-magnon problem.

V. RESULTS

Bell *et al.*[20] studied the two-magnon spectrum of the alternating bond $S = \frac{1}{2}$ Heisenberg chain and found a total of

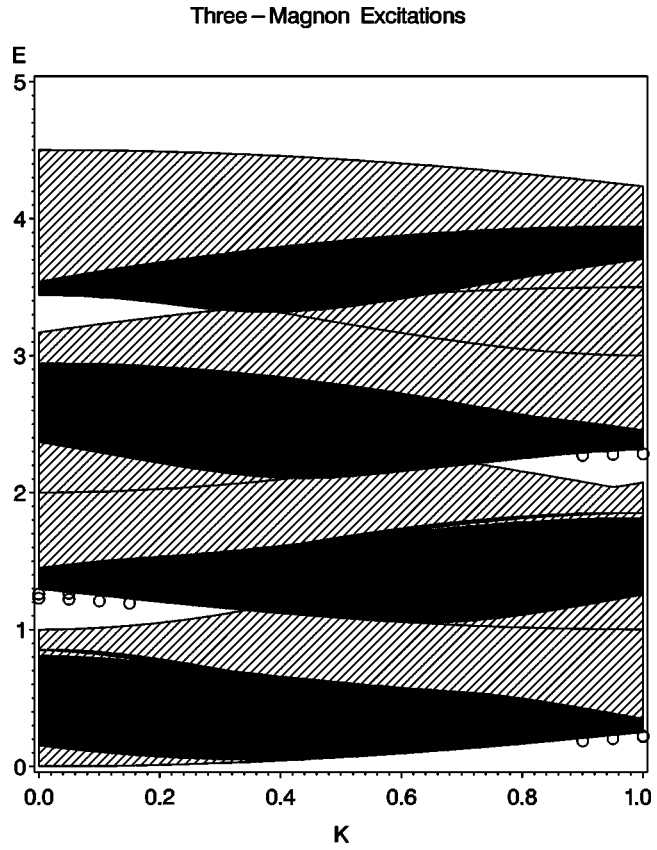

FIG. 4. The three-magnon continua and the three-magnon bound states (open circles) of the uniform spin $S=S'=\frac{1}{2}$ chain with alternating bonds ($J_1=2J_2=1$). The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

four two-magnon bound state branches compared to the uniform chain which only has one branch. Medved *et al.*[6] studied higher spin models and found that an additional bound state branch appears when the spin magnitude is increased to $S=1$. Here we extend these studies to the three-magnon excitations in alternating bond or spin chains. The uniform $S=\frac{1}{2}$ Heisenberg chain[9,10] has a single three-magnon bound state branch below the continuum throughout the first Brillouin zone $|K|\leqslant\pi$ but additional bound states[8] can appear near the Brillouin-zone boundary for higher values of S. When an alternation of the spins or the bonds is introduced, a folded Brillouin zone restricted to $|K|\leqslant\pi/2$ becomes the natural one, as it corresponds to two sites per primitive cell. In the folded representation, the uniform $S=\frac{1}{2}$ chain has a single bound state branch which folds back at $K=\pi/2$ and overlaps with the folded continuum. When alternation is introduced, the ''acoustic'' branch of the three-magnon bound state will remain with a gap between it and the ''optic'' branch at the new Brillouin-zone boundary. This ''optic'' branch may or may not lie entirely within the continuum. If it is below the continuum, it should only be present near the Brillouin-zone boundary and should become a resonant state when it enters the continuum region at smaller K unless it emerges again inside a continuum gap.

We first present some results for the case $S=S'=\frac{1}{2}$ with bond alternation. As the ratio J_2/J_1 is decreased from unity, a gap first appears in the continuum near $K=\pi/2$ followed by two additional gaps near $K=0$. Eventually these gaps extend across the entire zone and four distinct continua are formed. We have used the recursion method described above to calculate a local Green's function corresponding to one of the kets $\{|e,K;x,y\rangle,|o,K;x,y\rangle\}$ as a function of energy. The three magnon bound states are identified from the imaginary part of the Green's function. The bound state branch below the lowest continuum develops a gap at the zone boundary and the upper branch only exists for a small region of K before it becomes a resonance inside the continuum. As the ratio of J_2/J_1 decreases further, additional bound state branches are found in the two lowest gaps. However, we do not find any bound states in the highest gap. Figure 4 shows the spectrum for $2J_2=J_1$. The lowest gap contains two bound state branches with a small gap between them at $K=0$ and the second gap has a single branch near $K=\pi/2$. These branches are in addition to the usual two branches below the lowest continuum which have a small gap at $K=\pi/2$.

Exact results can be obtained in the case where either J_1 or J_2 is zero for all values of S and S'. We will discuss this case next as it provides a useful reference for comparing our results with both J_1 and J_2 nonzero. This limit will also help understand why there are no bound states in the highest gap for the case $S=S'=\frac{1}{2}$.

In the $J_2\rightarrow 0$ limit, the Hamiltonian (1) becomes

$$\hat{H}=\sum_{n=1}^{N/2}\hat{H}_{2n}^{b}, \tag{34}$$

TABLE IV. Three-magnon excitations for chains of isolated blocks in units of J_1 and where $\Xi=J_1(S+S')$. The degeneracy of the levels due to the three-free (3f), two-bound one-free (2b1f), and three-bound (3b) states has been indicated.

	Three-magnon excitation energies in the $J_2\rightarrow 0$ limit						
S,S'	0	Ξ	$2\Xi-J_1$	2Ξ	$3\Xi-3J_1$	$3\Xi-J_1$	3Ξ
Degeneracy	3f, 2b1f, 3b	3f, 2b1f, 3b	2b1f, 3b	3f, 2b1f	3b	2b1f	3f
1/2, 1/2	0,0,−	1,1,−	−,−	2,−			3
1, 1/2	0,0,0	1.5,1.5,−	−,−	3,3			4.5
3/2, 1/2	0,0,0	2,2,2	3,−	4,4		5	6
1, 1	0,0,0	2,2,2	3,−	4,4		5	6
3/2, 1	0,0,0	2.5,2.5,2.5	4,4	5,5		6.5	7.5
3/2, 3/2	0,0,0	3,3,3	5,5	6,6	6	8	9
2, 3/2	0,0,0	3.5,3.5,3.5	6,6	7,7	7.5	9.5	10.5

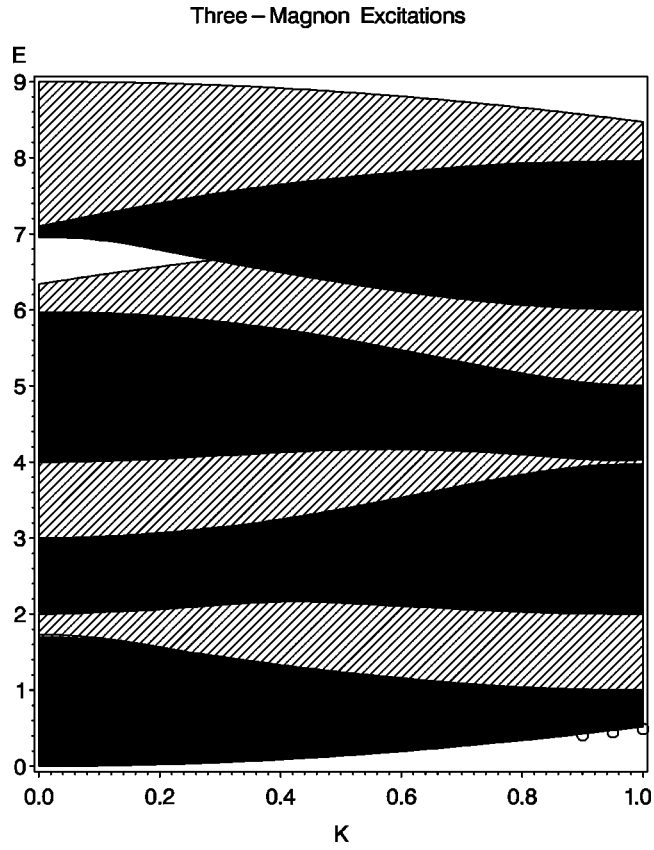

FIG. 5. The three-magnon continua for the $S=2S'=1$ uniform bond chain. The open circles represent the three magnon bound states. The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

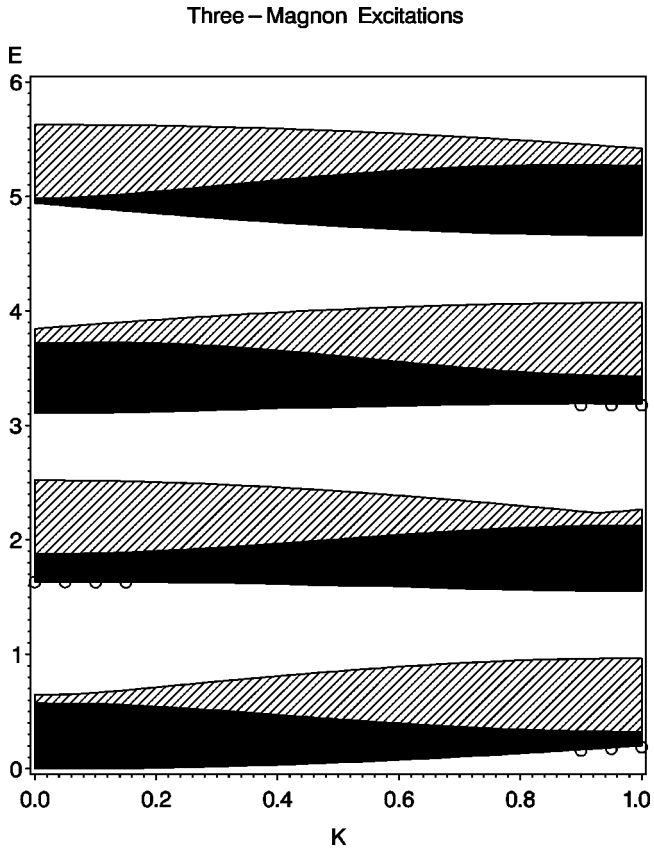

FIG. 6. The three-magnon continua for the $S=2S'=1$, $J_1=4J_2=1$ chain. The open circles represent the three magnon bound states. The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

where

$$\hat{H}^b_{2n}=-J_1\vec{S}'_{2n}\cdot\vec{S}_{2n+1} \tag{35}$$

and the energy can be expressed in terms of the total spin J of each noninteracting block. When the system is in our reference state $|0\rangle$, all the blocks have a total spin $J=S+S'$ and the total energy is $E_0=-J_1SS'(N/2)$. One-magnon excitations correspond to having a single deviation in any block and the energy transitions for the one-magnon excitations are

$$\Delta E_{J,J_z=+J\to J-1,J_z=+J-1}=J_1(S+S'),$$

$$\Delta E_{J,J_z=+J\to J,J_z=+J-1}=0, \tag{36}$$

which agree with Eq. (5) when evaluated for $J_2=0$. The two-magnon excitations can have either two single deviations in different blocks (2f) or in the same block (2b). When in different blocks, the energy is simply the sum of two single excitations and correspond to the excitations for two-free magnons. Two deviations in the same block involve a transition $J_z=+J\to J_z=+J-2$. A state with $J_z=+J-2$ could correspond to states with total spin equal to J, $J-1$ or $J-2$, and the excitation energies are given by

$$\Delta E_{2b}=\begin{cases}0\\ J_1(S+S') \quad \text{if} \quad S+S'\geqslant\frac{3}{2},\\ 2J_1(S+S')-J_1 \quad \text{if} \quad S+S'\geqslant 2.\end{cases} \tag{37}$$

Combining the transitions for two deviations in different blocks or in the same block, we obtain the following two-magnon states:

$$\Delta E=\begin{cases}0 \quad 2f,2b,\\ J_1(S+S') \quad 2f,(2b \ \text{if} \ S+S'\geqslant\frac{3}{2}),\\ 2J_1(S+S')-J_1 \quad (2b \ \text{if} \ S+S'\geqslant 2),\\ 2J_1(S+S') \quad 2f.\end{cases} \tag{38}$$

Similarly, the three-magnon states are the result of combining three deviations that could be in three separate blocks (3f), two in one block and the other in a different block (2b1f), or the three in the same block (3b). These three cases can be summarized as follows:

$$\Delta E=\begin{cases} 0 \quad 3f,2b1f,(3b \text{ if } S+S'\geqslant\frac{3}{2}) \\ J_1(S+S') \quad 3f,2b1f,(3b \text{ if } S+S'\geqslant 2) \\ 2J_1(S+S')-J_1 \quad (2b1f \text{ if } S+S'\geqslant 2),(3b \text{ if } S+S'\geqslant\frac{5}{2}) \\ 2J_1(S+S') \quad 3f,(2b1f \text{ if } S+S'\geqslant\frac{3}{2}) \\ 3J_1(S+S')-3J_1 \quad (3b \text{ if } S+S'\geqslant 3) \\ 3J_1(S+S')-J_1 \quad (2b1f \text{ if } S+S'\geqslant 2) \\ 3J_1(S+S') \quad 3f. \end{cases} \tag{39}$$

In Table IV, we have tabulated the three-magnon excitation energies derived from the $J_2\rightarrow 0$ limit for different S,S' chains indicating whether the excitation corresponds to three deviations in separate blocks (3f), two in one block (2b1f) or all three in the same block (3b). The first observation is that the number of levels (not considering degeneracy) increases from a minimum of 4 to a maximum of 7 as the total spin $(S+S')$ of the chain increases. Also, these energy levels separate as the total spin gets larger and this increases the possibility that gaps will appear even in the case of homogeneous bonds.

The degeneracy of each three-magnon level in the $J_2\rightarrow 0$ limit allows us to predict where three-magnon bound states can be found when J_2 is nonzero. Using the levels which correspond to three deviations in the same block (3b) or two in the same block (2b1f) as indicators, we expect to find three-magnon bound states below the continua associated with the levels 0, Ξ, $2\Xi-J_1$, 2Ξ, $3\Xi-3J_1$, and $3\Xi-J_1$, where $\Xi=J_1(S+S')$. Of course, all three types of the levels are bound when $J_2=0$. However, the bound states should split from the rest of the levels once J_2 is different from zero. For the special case of $S=S'=\frac{1}{2}$, bound state branches should only appear below the lowest continuum and in the first gap if we use the indicators above. This prediction agrees with the results shown in Fig. 4 except for the appearance of an additional bound state branch in the first gap at small K and another branch in the second gap near the Brillouin-zone boundary. If we associate these additional branches with the 3f states, then bound states only seem to appear below the three-free continua which have a contribution from at least one acoustic free magnon state. The ''OOO'' three-free continuum does not seem to have an associated bound state branch. If one of the spin magnitudes is increased to unity, we might expect an additional bound state branch below the lowest continuum. However, if such a state appears it will only be easily visible near the Brillouin-zone boundary. As the total spin $S+S'$ increases further, additional bound states in the gaps should be present. It is clear that the $2\Xi-J_1$ and $3\Xi-J_1$ levels will only appear for chains with $S+S'\geqslant 2$ and the $3\Xi-3J_1$ level for chains with $S+S'\geqslant 3$. Hence we do not expect to observe any bound states in the highest gap for the cases $S=S'=\frac{1}{2}$ and $S=2S'=1$.

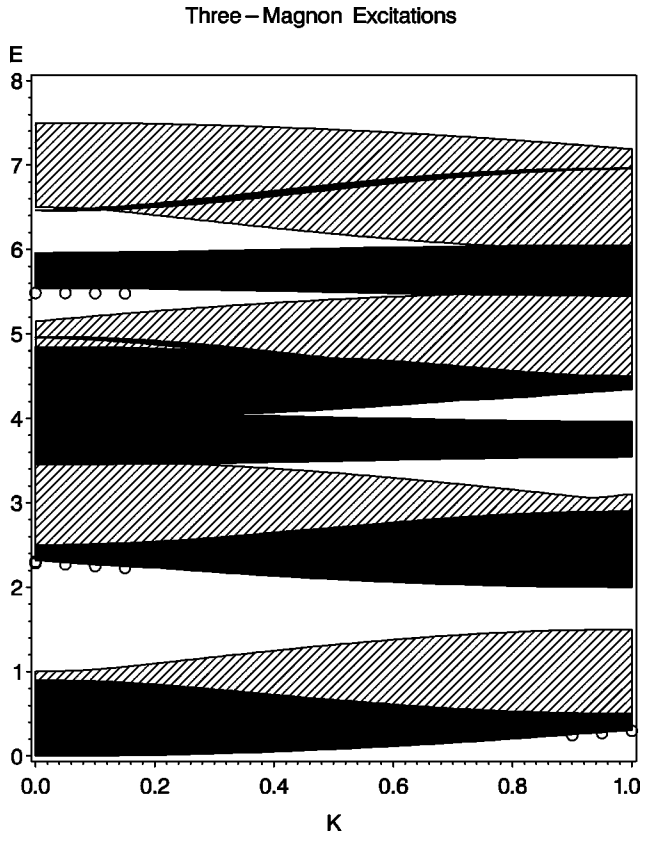

FIG. 7. The three-magnon continua for the $S=S'=1$, $J_1=4J_2=1$ chain. The open circles represent the three magnon bound states. The energy is in units of J_1 and the total wave vector K is in units of $\pi/2$.

We will now briefly discuss some results for the alternating $S=2S'=1$ chain. The $J_2=0$ limit predicts that wider gaps appear due to the fact that the levels separate as the total spin of a block increases, even in the case of no bond alternation. As can be seen in Fig. 5 one gap is present for small values of K as a consequence of the spin alternation. As J_2 decreases, more gaps start to appear and widen as shown in Fig. 6. Using the recursion method, we find bound states below the lower edge of the three-magnon continuum and in the first two gaps as expected from the $J_2\rightarrow 0$ analysis. In general, the results for a $S=2S'=1$ alternating bond chain look quite similar to the $S=S'=\frac{1}{2}$ alternating bond chain.

As a final example we consider the $S=S'=1$ alternating bond chain with $4J_2=J_1$. Figure 7 indicates that additional continua and gaps appear due to the larger value of the total spin. In this case we find bound state branches below the lowest continuum as well as in the first and fourth gaps. The procedure described here can be easily carried out for any values of S, S', J_1, and J_2.

VI. SUMMARY

In this paper we have described a simple method for studying the spectrum of three-magnon excitations in alternating spin or bond chains. The method can be used for both integrable and nonintegrable models. The recursion method is used to calculate a local Green's function in the three-magnon basis for fixed values of the spin and bond magnitudes and the total wave vector K. The energy values of the continuum edges at fixed K can be obtained from the knowledge of the corresponding one and two magnon excitation spectrum. This information is then used to terminate the continued fraction representation of the Green's function using the approach described by Turchi *et al.*[22]

In general, we find the usual bound state branch below the lower edge of the three magnon continuum for every case studied. Comparison of our results to the known analytical results of Bethe[9,10] for the $S=S'=1/2$ chain with homogeneous bonds indicate complete agreement. We extended the study of Bell *et al.*[20] of the alternating bond $S=1/2$ two-magnon spectrum to the three-magnon problem. When a bond alternation is introduced, gaps immediately appear and additional bound states are found. As J_2 decreases, the continua start to collapse into very predictable levels.

In conclusion, we have presented a direct and relatively simple procedure to search for bound states in alternating spin/bond chains. The recursion method was used very effectively to achieve this purpose and the generalization to an m-magnon problem would be straightforward.

ACKNOWLEDGMENTS

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