TWO-MAGNON BOUND STATES IN THE SIMPLE CUBIC HEISENBERG FERROMAGNET WITH NEXT NEAREST NEIGHBOUR INTERACTIONS

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Two-magnon bound states are examined for the simple cubic Heisenberg ferromagnet with next nearest neighbour exchange interactions using the Green function formalism. Analytic expressions for the two-magnon propagators are obtained for total pair wavevector at the Brillouin Zone corner. The Lattice Green's Functions that enter into these expressions in this case are shown to be related to those of the nearest neighbour FCC case which are much more easily calculated. The range of values of η (where η is the ratio of the next nearest to nearest neighbour interaction) for which bound states exist are calculated and compared with other results. A universal curve for the bound state energy as a function of η and valid for any spin is given.

1. *Introduction*

Most of the earlier work on two-magnon interactions and bound states has concentrated on the Heisenberg model with nearest neighbour (nn) interactions only. The results showed that bound states are usually formed for large total pair wavevectors [1–3] while for lower values of total pair wavevector some of the bound states enter the two-magnon continuum and become resonances [4]. The inclusion of single-ion or Ising anisotropy, or biquadratic exchange usually favors the formation of bound states [5–10]. Majumdar [11] obtained the bound states for a linear chain with next nearest neighbour (nnn) interactions and Loly et al. [12] examined the effect of nnn interactions on the resonances in the two-magnon Raman spectra in the FCC lattice.

Recently Krompiewski [13] using a Schrodinger scattering approach obtained the two-magnon bound states in the simple cubic (SC) lattice with nnn interactions. In this communication we present an alternate method based on the propagator or Green's function formalism. The advantage of this approach is that, in addition to bound states,

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the spectral functions within the two-magnon band can be directly obtained. In this paper we derive analytic expressions for the two-magnon propagators for total pair wavevector at the Brillouin Zone (BZ) corner. We also show that the Lattice Green's Functions (LGF) that enter into these expressions are given in terms of those for the nn FCC case. The advantage of this is that following Inoue [14], the latter LGF's are expressed in terms of elliptic integrals and easily evaluated both inside and outside the band using the arithmetic-geometric mean method [15]. The bound state conditions are obtained in a simple form and the range of values of η (where η is the ratio of the nnn to nn exchange interaction) calculated and compared to the results of a simple Ising analysis [10, 16] and those of Krompiewski [13]. A universal curve for the bound state energy as a function of η valid for any spin is presented.

The plan of the rest of the paper is as follows: in Section 2 the two-magnon propagator is defined and an exact equation for it obtained at absolute zero temperature. In Section 3 this equation is solved analytically for total pair wavevector at the BZ corner and then, in Section 4, these are used to obtain the bound state conditions and bound state energies. The conclusions are given in Section 5.

2. *The two-magnon propagator*

We consider the Heisenberg Hamiltonian

$$H = -\sum_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \tag{1}$$

where i and j stand for the position vectors $\boldsymbol{R}_i$ and $\boldsymbol{R}_j$ respectively and J_{ij} is the exchange interaction between the spins at sites i and j. The propagator for the scattering of two magnons with initial wavevectors $\boldsymbol{k}_1'$, $\boldsymbol{k}_2'$ and final wavevectors $\boldsymbol{k}_1$, $\boldsymbol{k}_2$ is

$$\begin{aligned} G(\boldsymbol{k}_1\boldsymbol{k}_2, \boldsymbol{k}_1'\boldsymbol{k}_2', t) &= -i\theta(t) \langle 0| [S^-_{\boldsymbol{k}_1}(t)S^-_{\boldsymbol{k}_2}(t), S^+_{\boldsymbol{k}_1'}(0)S^+_{\boldsymbol{k}_2'}(0)] |0\rangle \\ &= \langle\!\langle S^-_{\boldsymbol{k}_1}(t)S^-_{\boldsymbol{k}_2}(t) | S^+_{\boldsymbol{k}_1'}(0)S^+_{\boldsymbol{k}_2'}(0)\rangle\!\rangle, \end{aligned} \tag{2}$$

where $S^\alpha_{\boldsymbol{k}}$ is the Fourier transform of S^α_i and $|0\rangle$ is the fully aligned ground state at $T = 0$ K defined such that $S^z_i|0\rangle = -S|0\rangle$ and $S^-_i|0\rangle = 0$. The equation of motion for the Green's function in (2) has the standard form

$$\begin{aligned} \omega G(\boldsymbol{k}_1\boldsymbol{k}_2, \boldsymbol{k}_1'\boldsymbol{k}_2', \omega) &= \langle 0| [S^-_{\boldsymbol{k}_1}S^-_{\boldsymbol{k}_2}, S^+_{\boldsymbol{k}_1'}S^+_{\boldsymbol{k}_2'}] |0\rangle \\ &+ \langle\!\langle [S^-_{\boldsymbol{k}_1}(t)S^-_{\boldsymbol{k}_2}(t), H] | S^+_{\boldsymbol{k}_1'}(0)S^+_{\boldsymbol{k}_2'}(0)\rangle\!\rangle_\omega, \end{aligned} \tag{3}$$

where $G(\ldots, \omega)$ and $\langle\!\langle \ldots \rangle\!\rangle_\omega$ represent Fourier transforms. Introducing the total and relative wavevectors

$$\begin{aligned} \boldsymbol{K} &= \boldsymbol{k}_1 + \boldsymbol{k}_2, \quad 2\boldsymbol{k} = \boldsymbol{k}_1 - \boldsymbol{k}_2 \\ \boldsymbol{K}' &= \boldsymbol{k}_1' + \boldsymbol{k}_2', \quad 2\boldsymbol{k}' = \boldsymbol{k}_1' - \boldsymbol{k}_2' \end{aligned} \tag{4}$$

and evaluating the commutators in (3), it is straightforward to show that

$$\{\omega-2S[2J(0)-J(\tfrac{1}{2}\boldsymbol{K}+\boldsymbol{k})-J(\tfrac{1}{2}\boldsymbol{K}-\boldsymbol{k})]\}G(\boldsymbol{K}\boldsymbol{k},\boldsymbol{K}'\boldsymbol{k}',\omega)$$

$$= 4S^2\delta_{\boldsymbol{K}-\boldsymbol{K}'}\left(\delta_{\boldsymbol{k}+\boldsymbol{k}'}+\delta_{\boldsymbol{k}-\boldsymbol{k}'}-\frac{1}{NS}\right)$$

$$-\frac{2}{N}\sum_{\boldsymbol{q}}[J(\boldsymbol{q})-J(\tfrac{1}{2}\boldsymbol{K}-\boldsymbol{k}-\boldsymbol{q})]G(\boldsymbol{K}\boldsymbol{k}+\boldsymbol{q},\boldsymbol{K}'\boldsymbol{k}',\omega), \tag{5}$$

where $J(\boldsymbol{q})$ is the Fourier transform of J_{ij} and N is the number of spins. Taking $\boldsymbol{K}' = \boldsymbol{K}$ (conservation of total momentum) and introducing the partial Fourier transform

$$G(ij,\boldsymbol{K},\omega) = \frac{1}{N}\sum_{\boldsymbol{k}\boldsymbol{k}'} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}_i}e^{i\boldsymbol{k}'\cdot\boldsymbol{R}_j}G(\boldsymbol{K}\boldsymbol{k},\boldsymbol{K}\boldsymbol{k}',\omega), \tag{6}$$

equation (5) reduces to

$$G(ij,\boldsymbol{K},\omega) = 8S^2\left(1-\frac{\delta_{j0}}{2S}\right)\Lambda(ij,\boldsymbol{K},\omega)+2\sum_{l}J_l\tilde{\Lambda}(il,\boldsymbol{K},\omega)G(lj,\boldsymbol{K},\omega), \tag{7}$$

where

$$\Lambda(ij,\boldsymbol{K},\omega) = \frac{1}{N}\sum_{\boldsymbol{k}}\frac{\cos\boldsymbol{k}\cdot\boldsymbol{R}_i\cos\boldsymbol{k}\cdot\boldsymbol{R}_j}{\omega-\Omega(\boldsymbol{K},\boldsymbol{k})}, \tag{8}$$

$$\tilde{\Lambda}(ij,\boldsymbol{K},\omega) = \frac{1}{N}\sum_{\boldsymbol{k}}\frac{\cos\boldsymbol{k}\cdot\boldsymbol{R}_i(\cos\tfrac{1}{2}\boldsymbol{K}\cdot\boldsymbol{R}_j-\cos\boldsymbol{k}\cdot\boldsymbol{R}_j)}{\omega-\Omega(\boldsymbol{K},\boldsymbol{k})}, \tag{9}$$

and

$$\Omega(\boldsymbol{K},\boldsymbol{k}) = 2S[2J(0)-J(\tfrac{1}{2}\boldsymbol{K}+\boldsymbol{k})-J(\tfrac{1}{2}\boldsymbol{K}-\boldsymbol{k})] \tag{10}$$

is the total energy of two non-interacting spin waves with wavevectors $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$. We note that for nn interactions only, the Green's function equation in (7) is the same as that given by Wortis [1].

From equation (6), the diagonal part $G(ii,\boldsymbol{K},\omega)$ of the Green's function represents the propagator for processes involving the creation of two spin deviations separated by a distance $\boldsymbol{R}_i$. Below we consider a SC lattice and examine processes where two spin deviations are created on the same site ($\boldsymbol{R}_0 = (0, 0, 0)$), 1st neighbour sites ($\boldsymbol{R}_1 = (1, 0, 0)a$) and 2nd neighbour sites ($\boldsymbol{R}_2 = (1, 1, 0)a$) where a is the lattice constant. In shorthand notation these sites are indicated by $i = 0$, 1 and 2 respectively.

Equation (7) represents a set of coupled equations involving various Green's functions which, in general, is quite complex. For the SC nnn problem with $\boldsymbol{K}$ in the (111)-direction, these equations can be written in matrix forms which have to be inverted numerically.

Another drawback for the nnn problem is the great effort required to obtain the functions Λ and $\tilde{\Lambda}$ that appear in (7). These functions can be expressed in terms of the so-called Lattice Green's Functions (LGF). The LGF for site i is defined as

$$L_i(\omega) = \frac{1}{N}\sum_{k} \frac{e^{i\boldsymbol{k}\cdot\boldsymbol{R}_i}}{\omega-\Omega(\boldsymbol{K},\boldsymbol{k})}. \tag{11}$$

At present these LGF's can only be obtained by evaluating the sums in (11) numerically. However, at the zone corner ($\boldsymbol{K} = (1, 1, 1)\pi/a$) a considerable simplification results which enables us to obtain analytic expressions for $G(00, \boldsymbol{K}, \omega)$, $G(11, \boldsymbol{K}, \omega)$ and $G(22, \boldsymbol{K}, \omega)$. Furthermore, the LGF's in this case can be written in terms of those for the nn FCC case which Inoue [14] has shown to be expressible in terms of elliptic integrals of the first and second kinds. These integrals are easily evaluated both inside and outside the band using the arithmetic-geometric mean method [15]. These solutions are discussed in the next section.

3. Solution at the zone corner

When the total pair wavevector is at the zone corner ($\boldsymbol{K} = (1, 1, 1)\pi/a$) equation (7) can be solved explicitly. After some algebra we find

$$G(00, \boldsymbol{K}, \omega) = \frac{S}{3J_1}\left(1-\frac{1}{2S}\right)\left(L_{000}-\eta\frac{L_{110}(L_{000}+L_{110})}{SD_1}\right), \tag{12}$$

$$G(11, \boldsymbol{K}, \omega) = \frac{S}{6J_1}\frac{(L_{000}+L_{200})}{D_2}, \tag{13}$$

$$G(22, \boldsymbol{K}, \omega) = \frac{S}{3J_1}\frac{[P(L_{000}+L_{220})+Q(L_{211}-L_{110})+TL_{200}]}{D_1D_3D_4}, \tag{14}$$

where

$$D_1 = 1+\frac{\eta}{12S}(8L_{110}-2L_{200}+L_{220}+L_{000}+4L_{211}), \tag{15}$$

$$D_2 = 1+\frac{(L_{000}+L_{200})}{12S}, \tag{16}$$

$$D_3 = 1+\frac{\eta}{12S}(L_{000}+L_{220}+2L_{200}), \tag{17}$$

$$D_4 = 1+\frac{\eta}{12S}(L_{000}+2L_{110}+L_{220}-2L_{211}-2L_{200}), \tag{18}$$

$$P = \tfrac{1}{2}[A(A+2B-C)-4B^2], \tag{19}$$

$$Q = -2B(A+C), \tag{20}$$

$$T = -C(A+2B-C)-4B^2, \tag{21}$$

$$A = 1+\frac{\eta}{12S}(L_{000}+2L_{110}+L_{220}), \tag{22}$$

$$B = \frac{\eta}{12S}(L_{110}+L_{211}), \tag{23}$$

$$C = \frac{\eta}{6S}(L_{200}-L_{110}). \tag{24}$$

In the above $\eta = J_2/J_1$ where J_1 and J_2 are the nearest and next nearest neighbour exchange constants respectively and L_{lmn} is the normalized LGF for $\boldsymbol{R}_i = (l, m, n)\, a$ given by

$$L_{lmn}(\hat{\omega}) = \frac{1}{(2\pi)^3}\iiint\limits_{-\pi}^{\pi} dxdydz\, \frac{\cos(lx+my+nz)}{\hat{\omega}-\hat{\Omega}}, \tag{25}$$

where $\hat{\omega} = (\omega/24SJ_1)-2\eta$ and

$$\hat{\Omega} = (\Omega/24SJ_1)-2\eta = 1-\frac{2\eta}{3}(\sin x\sin y+\sin y\sin z+\sin z\sin x). \tag{26}$$

The only LGF's involved are L_{000}, L_{110}, L_{200}, L_{211} and L_{220}. However not all of these are independent and the following identities connecting the LGF's can be derived

$$(\hat{\omega}-1)L_{000}-2\eta L_{110} = 1, \tag{27}$$

$$(\hat{\omega}-1)L_{110}+\frac{\eta}{6}(2L_{200}-L_{220}-L_{000}+4L_{110}-4L_{211}) = 0. \tag{28}$$

We use these identities to eliminate L_{110} and L_{211}. The remaining LGF's are expressed in terms of those of the nn FCC case defined as

$$L^{\mathrm{FCC}}_{lmn}(t) = \frac{1}{\pi^3}\iiint\limits_{0}^{\pi} \frac{dxdydz\cos lx\cos my\cos nz}{t-(\cos x\cos y+\cos y\cos z+\cos z\cos x)}. \tag{29}$$

It is straightforward to show that

$$L_{lmn}(\hat{\omega}) = \frac{-3}{2\eta}(-1)^{\frac{l+m+n}{2}} L^{\mathrm{FCC}}_{lmn}\left(\frac{3}{2\eta}(1-\hat{\omega})\right). \tag{30}$$

Following Inoue [14], L^{FCC}_{lmn} are then expressed in terms of elliptic integrals of the 1st and 2nd kinds which can be easily evaluated by the arithmetic-geometric mean method [15] or other methods. Finally, we note that (25) can be written as

$$L_{lmn}(\hat{\omega}, \eta) = \frac{1}{\eta}L_{lmn}(u, 1), \tag{31}$$

where

$$u = 1 - \frac{1}{\eta}(1 - \hat{\omega}). \tag{32}$$

Thus we need only obtain L_{lmn} for one value of η viz $\eta = 1$ and use (31) and (32) to find L_{lmn} for any other value of η.

4. *Bound states at the zone corner*

The two-magnon bound states for single-site, 1st and 2nd neighbour excitations are given by the poles of $G(ii, \boldsymbol{K}, \omega)$ for $i = 0, 1$ and 2 respectively. Thus the bound state conditions are $D_1 = 0$ for $G(00, \boldsymbol{K}, \omega)$, $D_2 = 0$ for $G(11, \boldsymbol{K}, \omega)$ and $D_1 = 0$, $D_3 = 0$ and $D_4 = 0$ for $G(22, \boldsymbol{K}, \omega)$. It is straightforward to show that the bound state conditions thus obtained are the same as the determinental equations given by Krompiewski [13] for $\boldsymbol{K}$ at the Brillouin Zone corner. To be specific Krompiewski's $\det(F_1) = 0$ and $\det(F_2) = 0$ (his equations (29a) and (29b)) reduce to $D_1 D_2 D_3 = 0$ and $D_2 D_3 D_4 = 0$ respectively. The advantage of our results is that it gives us the bound state conditions for the three excitation processes mentioned above separately and hence enables us to determine which of these can form a bound state. Our numerical calculations show that $G(00, \boldsymbol{K}, \omega)$ and $G(22, \boldsymbol{K}, \omega)$ do not have poles outside the band and only $G(11, \boldsymbol{K}, \omega)$ can have a bound state. This is consistent with the conclusions of the Ising-type analysis [10, 16] although, as we shall see later, the predicted range of η values for which a bound state exists is different. From (13), the bound state condition is given by $D_2 = 0$ and using (31) and (32) this can be written as

$$L_{000}(u, 1) + L_{200}(u, 1) = -12S\eta. \tag{33}$$

The reason for writing the bound state condition this way is that the left hand side when plotted as a function of u is a universal curve and can be used to find the bound state energies for any value of η and S.

We look for bound states outside the two-magnon band, which from (26), extends from $\hat{\omega} = 1 - 2\eta$ to $\hat{\omega} = 1 + 2\eta/3$. Thus for $\eta > 0$ the bottom of the band is at $1 - 2\eta$ and for $\eta < 0$ it is at $1 + 2\eta/3$. In terms of u, the band extends from -1 to $5/3$ and the bottom of the band corresponds to -1 for $\eta > 0$ and $5/3$ for $\eta < 0$ i.e. $u < -1$ corresponds to energies below the band for $\eta > 0$ and $u > 5/3$ corresponds to energies below the band for $\eta < 0$. The left hand side of (33) is shown in Fig. 1 outside the two-magnon band. We note that at $u = -1$ it is finite and has its minimum value, while at $u = 5/3$ it has a logarithmic divergence. For a given η and S, the bound state is given by the intersection of this curve with the horizontal line corresponding to $(-12S\eta)$. This determines the value of u for the bound state and the bound state energy is then given by (32). We note from Fig. 1 that for $\eta < 0$ the bound state occurs at $u > 5/3$ and for $\eta > 0$ it occurs at $u < -1$ i.e. the energy of the bound state is always below the two-magnon band.

It can be seen from Fig 1 that a bound state exists for all negative values of η but for $\eta > 0$ it only exists below a critical value η_c given by the minimum value of the curve which occurs at $u = -1$. At this value of u, using the expressions for the LGF's given by Inoue [14], the left hand side of (33) reduces to a simple expression involving $K(k_{\pm})$ and $E(k_{\pm})$ where K and E are elliptic integrals of the 1st and 2nd kinds respectively and $k_{\pm}^2 = \frac{1}{2}\left(1 \mp \frac{\sqrt{3}}{2}\right)$. The resultant value of η_c is then

$$\eta_c = \frac{2}{\pi^2 S}\left[k_-^2 K(k_+)E(k_-) + k_+^2 K(k_-)E(k_+) - E(k_+)E(k_-)\right] = \frac{0.0464469}{S}. \qquad (34)$$

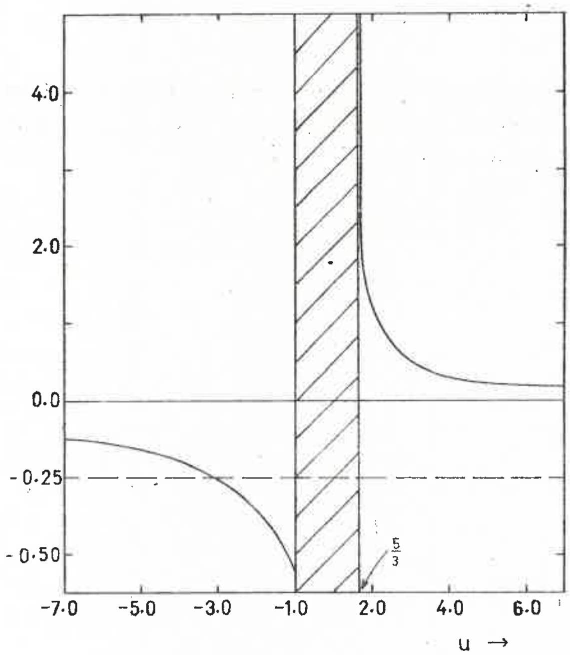

Fig. 1. The two-magnon bound state condition for $G(11, K, \omega)$. The solid curve represents the universal function given by the left hand side of equation (33) and the horizontal broken line represents the right hand side (for $\eta = 1/48$ and $S = 1$). The intersection of these two gives the bound state energy. The shaded region indicates the two-magnon continuum. Note the difference in scales for positive and negative values in the vertical axis

This agrees well with the value of 0.05 given by Krompiewski [13] for $S = 1$. The value of η_c given in (34) is also close to the result of a simple Ising analysis [10, 16] which gives $\eta_c = 1/24S = 0.04167/S$. However, the Ising analysis predicts a lower bound for $\eta(= -1/8S)$ for which a bound state exists while the present rigorous results show that bound states are possible for all negative η.

The bound state energy for any value of η and S can be found from the universal curve in Fig. 1. As can be seen from this figure, the divergence as u approaches 5/3 makes the graphical solution more difficult for larger negative η. However, in this region an asymptotic expansion for the elliptic integrals and hence for L_{000} and L_{200} enables us to obtain an analytic expression for the bound state energy. For $u = 5/3+\delta$, where δ is small and positive it is straightforward to show that

$$L_{000}(u, 1)+L_{200}(u, 1) = \frac{6}{\pi^2}\left[\ln\left(\frac{32}{3\delta}\right)-1\right]. \tag{35}$$

Equating this to $-12S\eta$ gives δ and hence u as a function of η. Thus using (32) we find that the bound state energy is given by

$$\hat{\omega} = 1+\frac{2\eta}{3}\left[1+16e^{-(1-2\pi^2 S\eta)}\right]. \tag{36}$$

The range of η for which (36) is a good approximation is measured by the accuracy of the expansion in (35) which is better than 1% for $\eta = -1/3S$, better than 0.1% for $\eta = -1/2S$ and gets better for larger negative η.

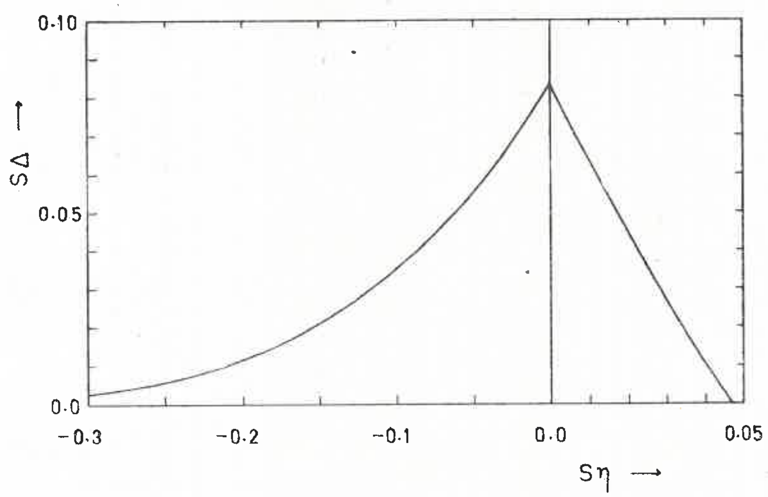

Fig. 2. Two-magnon bound state energy as a function of η. $\Delta = \hat{\omega}_0-\hat{\omega}_{BS}$ where $\hat{\omega}_0$ is the energy of the bottom of the two-magnon band and $\hat{\omega}_{BS}$ is the bound state energy. This is a universal curve valid for any spin S. Note the difference in scales for positive and negative values in the horizontal axis

The variation of the bound state energy with η for any value of the spin S is shown by the universal curve in figure 2 where Δ is defined as the binding energy $\hat{\omega}_0-\hat{\omega}_{BS}$ where $\hat{\omega}_0$ is the energy of the bottom of the band and $\hat{\omega}_{BS}$ is the bound state energy.

Finally, we have derived the sum rules satisfied by the spectral functions within the two-magnon band for the various spin pair excitations given by the imaginary part of $G(ii, \boldsymbol{K}, \omega)$ for $i = 0, 1, 2$. By evaluating the moments of the spectral functions and comparing them with the prediction of the sum rules we can (*i*) determine whether a bound state exists outside the band, and (*ii*) if a bound state does exist, determine its energy. We found excellent agreement with all the results presented above.

5. *Conclusions*

In this paper we have derived explicit analytic expressions for the two-magnon propagators in the nnn SC Heisenberg ferromagnet with total pair wavevector at the zone corner. Only bound states corresponding to 1st neighbour spin excitations were found consistent with a simple Ising analysis [10, 16]. The bound state conditions were obtained and compared with the Ising results [10, 16] and with those of Krompiewski [13]. Finally, the bound state energy as a function of η and spin S is given.

The Green's function approach used in this paper enables us to find the spectral function within the two-magnon band in addition to bound states outside it. For the SC problem with nnn interactions, so far it has only been possible to obtain compact analytic solutions for $\boldsymbol{K}$ at the zone corner. In a recent study [17] of two-magnon resonances within the band, equation (7) was expressed in matrix form for $\boldsymbol{K}$ along the (111)-direction and its solutions obtained by numerical methods.

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