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Two-Magnon Bound State in fcc Ferromagnets

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We show that an optically active two-magnon bound state exists for fcc Heisenberg ferromagnets. There is a repulsive interaction between magnons with equal and opposite wave vectors near the zone edge which in most lattices does not lead to any dramatic effects. However, in the fcc lattice there is a logarithmic divergence in the density of states at the zone boundary, which causes a bound state to split off from above the band. We discuss the possibility of observing such a state experimentally by Raman scattering, possibly in EuO.

I. INTRODUCTION

In 1963, Wortis[1] calculated the two-magnon states for the linear chain, quadratic layer, and simple-cubic lattices. He found that as well as a continuum of states, bound states could be formed below the continuum. These bound states exist even for small wave vectors and low energies in one and two dimensions, and indeed one- and two-dimensional Heisenberg ferromagnets do not order in the usual sense.[2] However, in the simple-cubic structure, these bound states only exist near the zone boundary, where they have a minimal effect on the low-temperature thermodynamics, and would be difficult to observe experimentally.

We have repeated the calculation of Wortis but for the fcc structure. The fcc ferromagnet has a logarithmic divergence in the density of one-magnon states at the highest magnon energy (this is in contrast to the sc and bcc ferromagnets which have a finite density of states everywhere), and this divergence means that the weak repulsive force between magnons can split off a bound state *above* the two-magnon band that will be optically active. This $\Delta M=2$ process should be observable in a Raman-scattering experiment.

We describe in Sec. II how the coupling to the two-magnon states might take place in the rare-earth compounds with particular reference to Eu^{2+}. In Sec. III we develop the theory, and apply it to the sc lattice in Sec. IV, and the fcc lattice in Sec. V, where it is shown that the bound state would be difficult to observe in EuO because of the large spin value of the Eu^{2+} ion ($S=\frac{7}{2}$). Nevertheless, the Raman cross section shows some interesting structure within the band that should be observable experimentally.

II. LIGHT SCATTERING IN FERROMAGNETS

In the past few years, light scattering has been shown to be a useful probe of the properties of antiferromagnets, especially the iron-group fluorides.[3] The single-magnon excitation ($\Delta M=1$) is seen through the spin-orbit coupling, whereas the two-magnon band ($\Delta M=0$), which consists of one magnon from either branch, is seen through the exchange coupling of pairs of neighboring magnetic ions in excited states. This exchange is of the Heisenberg form in many of the transition-metal compounds which have quenched angular momentum in the ground state, and so conserves the total value of the spin and only permits $\Delta M=0$ processes.

The two-magnon excitations in ferromagnets are $\Delta M=2$ excitations. In a recent paper, Moriya[4] has suggested that these processes could be seen in the rare-earth metals either by use of the spin-orbit coupling or through the s-f exchange interactions. This spin-orbit coupling mechanism (which was first proposed by Elliott and Loudon[5] for insulators) will clearly have comparable magnitude in the rare-earth compounds, as it is a single-site mechanism that depends primarily on the $4f$ electrons.

To illustrate this mechanism, we look at EuO. The ground state of the Eu^{2+} ion is an ^8S state, and we can assume that the effects of the crystal field are small enough so that the Eu^{2+} ion can be considered to be spherically symmetric. The electric dipole operator of the incident radiation can promote a $4f$ electron to either a $5d$ or $5g$ state. The spin-orbit coupling then acts twice in the excited configuration, and the electron returns to the $4f$ shell via the electric dipole operator of the scattered radiation. In fourth-order perturbation theory, we can get an effective interaction in the ground state that contains two spin operators (the orbital matrix elements having been taken). The size of the interaction will depend on the details of the state involved, but the form is determined by symmetry. The effective Hamiltonian for Raman scattering will contain all the invariant combinations of the incident electric field $\vec{E}_1$, the scattered electric field $\vec{E}_2$, and the spin operator $\vec{S}$ twice:

$$(\vec{E}_1\cdot\vec{E}_2)(\vec{S}\cdot\vec{S})=(\vec{E}_1\cdot\vec{E}_2)S(S+1)\ ,$$

$$(\vec{E}_1\times\vec{E}_2)\cdot(\vec{S}\times\vec{S})=i(\vec{E}_1\times\vec{E}_2)\cdot\vec{S}\ ,$$

$$(\vec{E}_1\cdot\vec{S})(\vec{E}_2\cdot\vec{S})\ .$$

In particular, the third combination will give rise to $\Delta M = 2$ processes, i.e., two-magnon excitations, and we may write an effective Hamiltonian that describes the excitation of these magnons within the ground state of the system:

$$H_{\text{eff}} = A\sum_i \left(E_1^+E_2^+S_i^-S_i^- + E_1^-E_2^-S_i^+S_i^+\right) . \tag{2.1}$$

III. THEORY

We consider a ferromagnet described by a Hamiltonian

$$H = -\frac{1}{2}\sum_{i,\delta} J(\delta)\vec{S}_i \cdot \vec{S}_{i+\delta} , \tag{3.1}$$

where the exchange $J(\delta)$ only acts between the z nearest neighbors whose separation is described by a vector $\vec{\delta}$. The components of the spin obey the usual commutation relations

$$[S^+, S^-] = 2S^z , \qquad [S^z, S^\pm] = \pm S^\pm . \tag{3.2}$$

We define a Green's function at zero temperature,

$$G_{\delta\delta'} = \langle\langle A_\delta^\dagger ; A_{\delta'} \rangle\rangle$$
$$= \sum_i \left(\frac{\langle 0|A_\delta^\dagger|i\rangle\langle i|A_{\delta'}|0\rangle}{\omega - E_i + E_0} - \frac{\langle 0|A_{\delta'}|i\rangle\langle i|A_\delta^\dagger|0\rangle}{\omega + E_i - E_0} \right) , \tag{3.3}$$

where $A_\delta = \sum_i S_i^- S_{i+\delta}^-$. The second term in (3.3) is actually zero, because $|0\rangle$ is the fully aligned state in which all the spins point in the z direction; however, we shall retain it for convenience. We can see from (2.1) and using Fermi's golden rule that the cross section for Raman scattering will be proportional to

$$A^2[(E_1^x)^2 + (E_1^y)^2][(E_2^x)^2 + (E_2^y)^2]\,\text{Im}\,G_{00} . \tag{3.4}$$

The Green's function obeys the equation of motion

$$\omega G_{\delta\delta'} = \langle 0|[A_\delta^\dagger, A_{\delta'}]|0\rangle + \langle\langle [A_\delta^\dagger, H]; A_{\delta'}\rangle\rangle . \tag{3.5}$$

If we allow the S_i^z operators that result from evaluating $[A_\delta^\dagger, H]$ to operate on the ground state to the left, so that $\langle 0|S_i^z = \langle 0|S$, we obtain a closed set of equations for $G_{\delta\delta'}$

$$\omega G_{\delta\delta'} = N(2S)^2(\delta_{\delta,\delta'} + \delta_{\delta,-\delta'})[1 - (\delta_{\delta,0}/2S)]$$
$$+ 2S\sum_x J(x)G_{\delta\delta'} - 2S\sum_x J(x-\delta)G_{x\delta'}$$
$$- J(\delta)G_{\delta\delta'} + \sum_x J(x)G_{x\delta'}\delta_{\delta,0} . \tag{3.6}$$

The last two terms on the right-hand side represent interactions between the spin waves as does the $\delta_{\delta,0}/2S$ term that comes from the commutator in the inhomogeneous term in (3.5). Equation (3.6) can be solved by standard techniques; first the interaction terms are dropped, and the resulting equation is solved by a Fourier transform. When the interaction terms are included, the equation is of the Dyson form. This can be solved for the various $G_{\delta\delta'}$ because the interactions are of short range in real space. In particular, we are interested in G_{00} which is given by

$$\frac{G_{00}}{4NS^2} = \left(1 - \frac{1}{2S}\right)\left(\frac{g_{00} + (1/2S)g_{\delta 0}}{1 + (\omega/2S)g_{\delta 0}}\right) , \tag{3.7}$$

where

$$g_{00} = \frac{1}{N}\sum_{\vec{k}} \frac{1}{\omega - 2\omega_{\vec{k}}} ,$$
$$g_{\delta 0} = g_{0\delta} = \frac{1}{N}\sum_{\vec{k}} \frac{e^{i\vec{k}\cdot\vec{\delta}}}{\omega - 2\omega_{\vec{k}}} . \tag{3.8}$$

The spin-wave energies $\omega_{\vec{k}}$ are given by

$$\omega_{\vec{k}} = S[J(0) - J(\vec{k})] , \tag{3.9}$$

where $J(\vec{k}) = \sum_\delta J(\delta)e^{i\vec{k}\cdot\vec{\delta}}$. The crystal Green's functions g_{00} and $g_{\delta 0}$ are not independent but connected by the relationship

$$1 = \frac{1}{N}\sum_{\vec{k}} \frac{\omega - 2\omega_{\vec{k}}}{\omega - 2\omega_{\vec{k}}} = (\omega - 2JSz)g_{00} + 2JSzg_{\delta 0} . \tag{3.10}$$

The energy denominator in (3.7) is the same as that given by Wortis[1] and by Silberglitt and Harris[6] for the two spin-wave states with center-of-mass wave vector zero (i.e., the two spin waves have wave vectors $+\vec{k}$ and $-\vec{k}$, respectively). The numerator in (3.7) depends upon the particular way in which the coupling to the two-magnon states takes place, and is determined by (2.1) in this case. For large spin $G_{00} \propto g_{00}$, and the poles of G_{00} coincide with the poles of g_{00}, i.e., the spin-wave energies. For finite spin, new poles may occur because of the denominator in (3.7) vanishing, and we shall see that this happens in the fcc lattice.

The cross section is proportional to the imaginary part of G_{00}. By expanding the right-hand side of (3.7) in powers of $1/\omega$, it is easy to show that

$$\frac{G_{00}}{4NS^2} = \left(1 - \frac{1}{2S}\right)\frac{1}{\omega} + \left(1 - \frac{1}{2S}\right)\frac{2JSz}{\omega^2} + \cdots ,$$

and therefore the total integrated cross section is

$$\int_0^\infty \text{Im}\left(\frac{G_{00}}{4NS^2}\right) d\omega = \pi\left(1 - \frac{1}{2S}\right) , \tag{3.11}$$

and the first moment,

$$\int_0^\infty \text{Im}\left(\frac{G_{00}}{4NS^2}\right) \omega\, d\omega \Big/ \int_0^\infty \text{Im}\left(\frac{G_{00}}{4NS^2}\right) d\omega = 2JSz . \tag{3.12}$$

Thus, whereas the first moment is unaffected by the interaction, the total intensity is reduced by a factor $(1 - 1/2S)$.

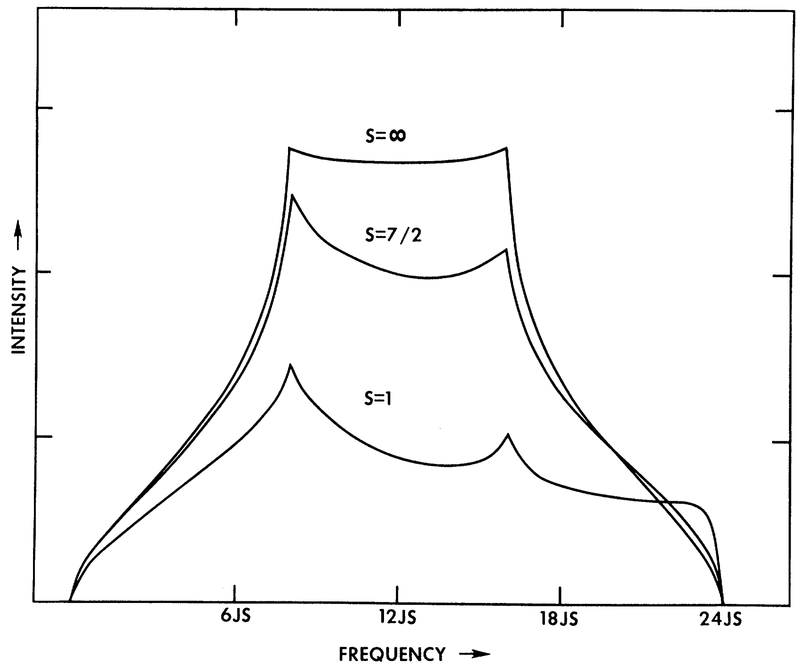

FIG. 1. Raman intensity for a sc ferromagnet for $S=\infty$ (no interactions), $S=\frac{7}{2}$, and $S=1$.

Another feature of (3.7) that is useful in checking numerical calculations is that, when $\omega = 2JSz$,

$$\mathrm{Im}\left(\frac{G_{00}}{4NS^2}\right)=\left[\left(1-\frac{1}{2S}\right)\Big/\left(1+\frac{1}{2S}\right)\right]\mathrm{Im}g_{00}\ . \qquad (3.13)$$

IV. SIMPLE-CUBIC LATTICE

Wortis[1] showed that no bound states exist for zero wave vector in the sc lattice, and we calculate the spectral weight function merely for comparison with the fcc lattice. If the separation between nearest neighbors is a, the spin-wave energies (3.9) become

$$\omega_{\vec{k}} = 6JS[1-\tfrac{1}{3}(\cos k_x a+\cos k_y a+\cos k_z a)]\ ,$$

and the crystal Green's function g_{00} in (3.8) may be written

$$g_{00}(\omega)=(-1/4JS)\,I_{sc}[3(1-\omega/12JS)]\ ,$$

where the integrals

$$I_{sc}(\epsilon)=\left(\frac{1}{2\pi}\right)^3\int\int\int_{-\pi}^{\pi}\frac{dx\,dy\,dz}{\epsilon-\cos x+\cos y+\cos z}$$

have been evaluated numerically by Wolfram and Callaway.[7]

The spin-wave band stretches from $0\to 12JS$, and so the two spin-wave band goes from $0\to 24JS$. In Fig. 1, we plot the Raman intensity $\mathrm{Im}(G_{00}/4NS^2)$ against ω. The $S=\infty$ case corresponds to no interaction, when we just get the single spin-wave density of states stretched over twice the frequency scale. The interactions introduce an asymmetry into the spectrum, but no bound states are produced, as the sum rule (3.11) is satisfied by the band modes. Note that the interaction has a very small effect at low energies, as can be seen in both Figs. 1 and 2. This is a consequence of the result first obtained by Dyson[8] that long-wavelength magnons only interact weakly.

V. FACE-CENTERED-CUBIC LATTICE

The spin-wave dispersion (3.9) in an fcc lattice becomes

$$\omega_{\vec{k}} = 12JS[1-\tfrac{1}{3}(\cos k_x a\cos k_y a+\cos k_y a\cos k_z a+\cos k_z a\cos k_x a)]\ ,$$

where $2a$ is the size of the unit cube so that the distance between nearest neighbors is $\sqrt{2}a$. The crystal Green's function G_{00} in (3.8) may be written

$$g_{00}(\omega)=(-1/8JS)\,I_{fcc}[3(1-\omega/24JS)]\ ,$$

where the integrals

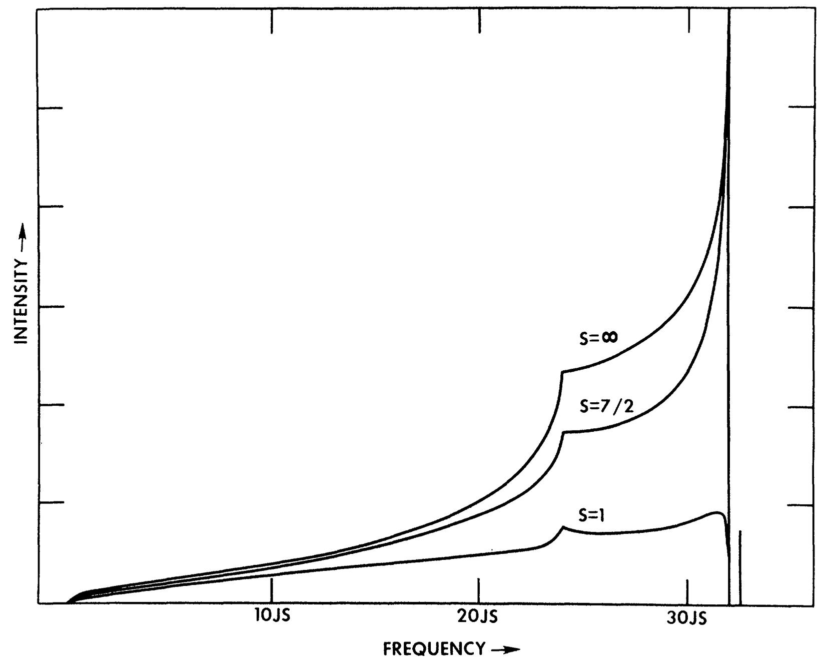

FIG. 2. Raman intensity for an fcc ferromagnet for $S=\infty$ (no interactions), $S=\frac{7}{2}$, and $S=1$. Position of the bound state is shown for $S=1$ but is too near the band edge to be shown for $S=\frac{7}{2}$.

$$I_{fcc}(\epsilon)=\left(\frac{1}{2\pi}\right)^3 \iiint_{-\pi}^{\pi} \frac{dx\,dy\,dz}{\epsilon-(\cos x\cos y+\cos y\cos z+\cos z\cos y)} \tag{5.1}$$

have been tabulated by Frikkee.[9]

The single spin-wave energies go from $0\rightarrow 16JS$, and so the two spin-wave band goes from $0\rightarrow 32JS$. The integral $I_{fcc}(\epsilon)$ is defined over a cubic zone (which has a volume of twice the Brillouin zone). This is convenient and introduces no error, as everything is counted exactly twice in the integral and a factor of 2 introduced to correct for the overcounting. When $\omega=32JS$ (i.e., the top of the band), $\epsilon=-1$, and the integrand in (5.1) is degenerate along lines such as $(x, 0, \pi)$ in reciprocal space. This leads to a logarithmic divergence in the density of states [which is proportional to $\mathrm{Im}I_{fcc}(\epsilon)$] at $\epsilon=-1$. The precise form of this divergence may be calculated by constructing a surface around, and close to, the degenerate lines in reciprocal space, calculating the volume enclosed by this surface, and hence, by differentiation, obtaining the density of states. We find that

$$\mathrm{Im}I_{fcc}(\epsilon)\sim\frac{1}{\pi}\left[\frac{12}{\pi}-\frac{3}{2}-\frac{3}{2}\ln\left(\frac{1+\epsilon}{4}\right)\right]+O(1+\epsilon)\ . \tag{5.2}$$

We have used this asymptotic form to calculate $\mathrm{Im}I_{fcc}(\epsilon)$ close to the zone boundary, and the numerical results are given in the Appendix. These results join smoothly onto the computer calculations of Frikkee who does not attempt to get very close to the zone edge. The real part of $I_{fcc}(\epsilon)$ was evaluated using the Kramers-Kronig relation

$$\mathrm{Re}I_{fcc}(\epsilon)=\frac{1}{\pi}P\int_{-1}^{3}\frac{d\epsilon'\,\mathrm{Im}I_{fcc}(\epsilon')}{\epsilon-\epsilon'}\ , \tag{5.3}$$

and integrating numerically. The real part diverges both above and below the zone boundary, this divergence being caused by the logarithmic divergence in the density of states, so that great care had to be taken with the integral (5.3). Frikkee's values for $\mathrm{Im}I_{fcc}(\epsilon)$ were used up to $\epsilon=-0.96$, and the asymptotic form (5.2) was used between $\epsilon=-0.96$ and $\epsilon=-1$. The results for the real part near the zone boundary are also given in the Appendix.

In Fig. 2, we plot the Raman intensity $\mathrm{Im}(G_{00}/4NS^2)$ against ω. The $S=\infty$ case (no interaction) shows the divergent density of states at $\omega=32JS$. When the interaction is included, the intensity drops to zero at the zone boundary. However, in this case, the sum rule (3.11) is *not* satisfied by the band modes, which means that a bound state must be split off above the band. We find that this is indeed the case as the energy denominator in (3.7) vanishes for a certain $\omega>32JS$. The position of the bound state is shown in Fig. 2 for $S=1$, but is too close to the band edge to be shown for $S=\frac{7}{2}$. Figure 3 shows the position of the bound state for $S=1$ up to $S=\frac{7}{2}$ and also the weight in the bound

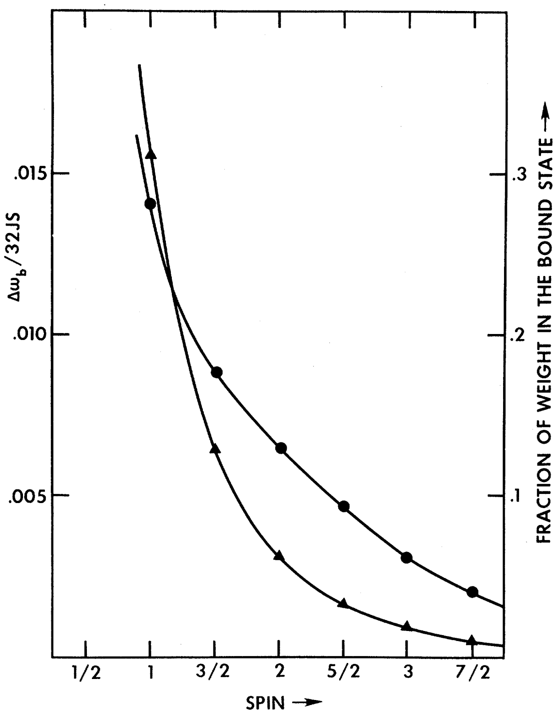

FIG. 3. Left-hand scale refers to the triangles which give the ratio of the splitting of the bound state from the top of the band ($\Delta\omega_b$) to the bandwidth ($32JS$). Right-hand scale refers to the circles which give the ratio of the weight in the bound state to the weight in the band.

state. We have verified numerically that the total weight satisfies the sum rule (3.11). We see from Fig. 3 that the spin-1 bound state is well split off from the band and contains considerable weight, whereas for spin $\frac{7}{2}$ it is only just split off and contains very little weight. In this case, however, the band intensity is correspondingly more peaked near the zone boundary. We note that for $S=\frac{1}{2}$, $\Delta\omega_b/32JS=0.052$, where $\Delta\omega_b$ is the distance of the bound state from the top of the band. However, the intensity is zero everywhere, as two spin deviations cannot exist on the same site for $S=\frac{1}{2}$.

The rare-earth ferromagnet EuO has the fcc structure and a Curie temperature of 69.3 °K.[10] If we assume that the exchange is entirely nearest neighbor, we can use the formula of Rushbrooke and Wood[11] (with $z=12$ and $S=\frac{7}{2}$) for the transition temperature to determine J. Hence, the top of the two spin-wave band ($32JS$ for EuO) would be at 109 cm^{-1}. Unfortunately, the bound state is only split off from the band by 0.05 cm^{-1}, which would make it almost impossible to resolve with current Raman apparatus. This small splitting occurs because the interaction is a $1/S$ effect and S is large in this case. We see from (3.4) that the scattered intensity will depend upon the angles that $\vec{E}_1$ and $\vec{E}_2$ make with the axis of spin alignment (the z axis). However, in a multidomain sample, the scattered intensity will be independent of the directions of $\vec{E}_1$ and $\vec{E}_2$.

It is, of course, very possible that exchange constants other than nearest neighbor are important in EuO.[12,13] It is difficult to get an unambiguous determination of the exchange parameters at the present time, as the spin-wave spectrum has not been measured by inelastic neutron scattering. Exchange interactions other than nearest neighbor will remove the degeneracy in reciprocal space that leads to the divergence in the density of states and, hence, may have a severe effect on the bound state.

The existence of a bound state with zero center-of-mass wave vector is perfectly consistent with Dyson's calculation[8] of the low-temperature thermodynamics of a ferromagnet as it occurs at a rather high energy, and it will not be populated at low temperatures. The low-temperature behavior of an fcc ferromagnet is no different from that of any other three-dimensional ferromagnet.

VI. CONCLUSIONS

We have shown that a bound state exists with center-of-mass momentum zero in fcc ferromagnets.

TABLE I. Real and imaginary parts of the Green's function (5.1) for the fcc lattice for energies ϵ near the zone edge.

ϵ	$\mathrm{Im}I_{fcc}(\epsilon)$	$\mathrm{Re}I_{fcc}(\epsilon)$
−0.90	2.500	−1.209
−0.91	2.550	−1.294
−0.92	2.606	−1.391
−0.93	2.670	−1.503
−0.94	2.747	−1.636
−0.95	2.831	−1.817
−0.96	2.937	−2.002
−0.97	3.074	−2.278
−0.98	3.268	−2.687
−0.99	3.599	−3.444
−1.001		−7.235
−1.002		−6.235
−1.003		−5.684
−1.004		−5.308
−1.005		−5.025
−1.006		−4.792
−1.007		−4.613
−1.008		−4.454
−1.009		−4.316
−1.01		−4.194
−1.02		−3.437
−1.03		−3.028
−1.04		−2.752
−1.05		−2.567
−1.06		−2.386
−1.07		−2.252
−1.08		−2.141
−1.09		−2.044
−1.10		−1.960

This is in sharp contrast to the sc and bcc ferromagnets where no such state exists. It would be interesting to investigate the behavior of this bound state for an arbitrary wave vector.

It is interesting to compare the present calculation of the two-magnon optical spectrum with a similar calculation in a Heisenberg antiferromagnet.[14] In that case, the *attractive* interactions caused a resonant peak to develop just below the top of the band. The position of this peak was rather insensitive to the crystal structure and determined by a square-root divergence in the density of states at the zone boundary. This divergence occured as a result of the form of the antiferromagnetic spin waves rather than the structure of the lattice. By contrast, in the present case, we find that the *repulsive* force may lead to a bound state, but the geometry of the lattice is a very important aspect.

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APPENDIX

The Green's function $I_{fcc}(\epsilon)$ is calculated near to the zone boundary. The imaginary part is obtained from (5.2) and the real part from the Kramers-Kronig relation (5.3) using the computations of Frikkee[9] for $\mathrm{Im}I_{fcc}(\epsilon)$ for $-0.96<\epsilon<3$ and the asymptotic form (5.2) for $-1<\epsilon<-0.96$. We estimate that, due to the difficulties of the numerical integration, the real part is correct to about 5%. See Table I.

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Spin-$\frac{1}{2}$ Heisenberg Ferromagnet on Cubic Lattices: Analysis of Critical Properties by a Transformation Method*

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The high-temperature series expansions for the spin-$\frac{1}{2}$ Heisenberg ferromagnetic model on cubic lattices are analyzed by a transformation method. Evidence is presented suggesting that the susceptibility critical exponent (γ) and the gap parameter (2Δ) are both smaller than the original estimates obtained by Padé approximant techniques. Specifically, we find that $\gamma=1.36\pm0.04$ and $2\Delta=3.50\pm0.20$. The error limits are to be taken as a reasonable confidence level rather than as a strict bound.

I. INTRODUCTION

Critical properties of all realistic three-dimensional models of magnetism are determined by the method of exact series expansions. It is generally accepted that critical values of the Ising model are, on the whole, reliably established.[1] Critical values of other models, such as the spin-$\frac{1}{2}$ *XY* model[2] and the spin-$\frac{1}{2}$ Heisenberg model,[3] have been determined only recently and with an uncertainty generally greater than in the Ising counterparts. In these extreme quantum models, the noncommutativity of spin operators complicates the evaluation of expansion coefficients enormously; moreover, there is an irregularity in the resulting series, apparently related to the noncommutativity in some way not yet