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Effect of Single-Ion Anisotropy on Two-Spin-Wave Bound State in a Heisenberg Ferromagnet*

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We consider the scattering of two spin waves in a uniaxial (easy axis) Heisenberg ferromagnet with single-ion anisotropy. The two-spin-deviation problem is solved exactly at zero temperature. We find (for $S > \frac{1}{2}$), in addition to the usual two-spin-wave bound states, a new "single-ion bound state," in which at the zone corner the two spin deviations are on the same site. When the magnitude of the anisotropy is comparable to the exchange interaction, the single-ion bound state becomes the dominant feature of the bound-state spectrum. For arbitrary spin there is a critical anisotropy strength above which the single-ion bound state exists throughout the Brillouin zone. We conclude that the presence of single-ion anisotropy enhances the possibility of experimental observation of the bound states.

I. INTRODUCTION

The Heisenberg model of ferromagnetism has been extensively studied.[1] The elementary excitations of this model are the spin waves, which consist of single spin deviations propagating through the lattice.[2] Considering only the Ising part

$$-J \sum_{\langle i,j \rangle} S_i^z S_j^z$$

of the Heisenberg Hamiltonian, one finds that the excitation energy of two *adjacent* spin deviations is lower by J than that of two nonadjacent ones, giving rise to an effective attractive interaction between spin waves. Although the transverse terms in the Heisenberg Hamiltonian tend to weaken this attraction, it has been shown by Wortis[3] and Hanus[4] that the attractive interaction results in the formation of bound states of two spin waves for a sufficiently large total wave vector $\vec{q}$. Physically, these "exchange bound states" correspond to two spin deviations close together in space and propagating through the lattice in a correlated fashion with total wave vector $\vec{q}$. At the zone corner $\vec{q}a=(\pi,\pi,\pi)$ the two spin deviations are in fact on adjacent sites, as would be the case for the Ising model.

We consider a uniaxial Heisenberg ferromagnet with single-ion anisotropy $-D\sum_i (S_i^z)^2$, with $D>0$. This term causes the excitation energy of two spin deviations on the same site to be lower by $2D$ than that of deviations on two different sites. Thus, the single-ion anisotropy also gives rise to an effective attractive interaction between spin waves. In this paper, we show that for $S>\frac{1}{2}$ an additional bound state of two spin waves results, in which at $\vec{q}a=(\pi,\pi,\pi)$ the two spin deviations are on the same site. We find that for $D\gtrsim J$ this "single-ion bound state" is the dominant feature of the bound-state spectrum. As the ratio of D to J is increased, the single-ion bound state exists over an increasing volume of $\vec{q}$ space. For sufficiently large D/J, the single-ion bound state is found to exist throughout the Brillouin zone.

II. PROBLEM FORMULATION

We consider a uniaxial Heisenberg ferromagnet described by the Hamiltonian

$$H=-J\sum_{\langle i,j\rangle}\vec{S}_i\cdot\vec{S}_j-D\sum_i (S_i^z)^2+h\sum_i S_i^z\ , \tag{1}$$

where $\langle i,j\rangle$ denotes a sum over nearest-neighbor pairs. Both the exchange interaction J and the single-ion anisotropy strength D are assumed to be positive. The magnetic field h aligns the spins along the negative-z direction in the ground state. Since $\sum_i S_i^z$ commutes with H, the states of the system can be classified according to the number of spin deviations n. Here $n=0$ corresponds to the ground state, and the $n=1$ space is diagonalized by the simple spin waves. We shall enumerate the $n=2$ states of the Hamiltonian [Eq. (1)].

We employ the Dyson-Maléev boson representation for the spin operators[5] and solve the two-spin-deviation problem exactly by calculating the zero-temperature two-boson t matrix. The singularities of this t matrix occur at the true two-spin-wave energies.[6] The same method has been used by Peletminskii and Baryakhtar,[7] who treated the present case with $\vec{q}=0$, and by Silberglitt and Harris,[8] who calculated the t matrix for all $\vec{q}$, but with $D=0$. The Dyson-Maléev representation is defined by

$$S_i^+=(2S)^{1/2}a_i^\dagger(1-a_i^\dagger a_i),\ S_i^-=(2S)^{1/2}a_i\ , \quad S_i^z=-S+a_i^\dagger a_i\ , \tag{2}$$

where a_i and $a_i^\dagger$ destroy and create, respectively, bosons at lattice site i. Using the Fourier transformation $a_k=(N)^{1/2}\sum_i e^{i\vec{k}\cdot\vec{x}_i}a_i$ we write the Hamiltonian [Eq. (1)] in terms of bosons and obtain[7,8]

$$H_{\rm DM}=E_0+\sum_k\epsilon_k a_k^\dagger a_k+(2N)^{-1}\sum_{k_1k_2q}V(k_1k_2q) \times a^\dagger_{q/2+k_2}a^\dagger_{q/2-k_2}a_{q/2+k_1}a_{q/2-k_1}\ , \tag{3}$$

where the ground-state energy $E_0=-\frac{1}{2}JNzS^2-DNS^2-hNS$, ϵ_k is the simple spin-wave energy, given by

$$\epsilon_k=JzS(1-\gamma_k)+2D(S-\tfrac{1}{2})+h\ , \tag{4}$$

and $V(k_1k_2q)$ is the full spin-wave interaction, given for a lattice with inversion symmetry by

$$V(k_1k_2q)=-2J\sum_{\delta>0}\cos(\vec{k}_1\cdot\vec{\delta})\times[\cos(\vec{k}_2\cdot\vec{\delta})-\cos(\tfrac{1}{2}\vec{q}\cdot\vec{\delta})]-2D\ . \tag{5}$$

In the above, z is the number of nearest neighbors of a given spin, $\vec{\delta}$ is a vector from a spin to one of its nearest neighbors, $\vec{k}_1$ and $\vec{k}_2$ represent, respectively, the incoming and outgoing relative wave vectors of an interacting spin-wave pair, and $\vec{q}$ is the total wave vector of the pair. (For simplicity, vector notation is suppressed in subscripts and arguments.) The function γ_k is defined in the usual way:

$$\gamma_k=z^{-1}\sum_\delta e^{i\vec{k}\cdot\vec{\delta}}\ . \tag{6}$$

We note from Eq. (5) that at $\vec{q}\cdot\vec{\delta}=\pi$ for all δ, the potential V is just that which would be obtained from the Ising model. Thus, the Ising wave functions and energies are the exact solutions at the Brillouin zone corner, as was anticipated earlier. This result is quite useful in that it gives a simple way of estimating the bound-state energies and their dependence on S and D/J.

At zero temperature, the t matrix obeys the integral equation[7,8]

$$t(k_1k_2q\omega)=V(k_1k_2q)+\frac{1}{N}\sum_{k_3}\frac{V(k_1k_3q)\,t(k_3k_2q\omega)}{\omega-\epsilon_{q/2+k_3}-\epsilon_{q/2-k_3}+i\eta}\ , \tag{7}$$

where $\eta\to0^+$. Equation (7) may be solved by the substitution (all sums are over positive δ only)

$$t(k_1k_2q\omega)=-2Dt_0(k_2q\omega)-2J\sum_\delta\cos(\vec{k}_1\cdot\vec{\delta})\,t_\delta(k_2q\omega) \tag{8}$$

yielding

$$\sum_{\beta=0,\delta}M_{\alpha\beta}(q,\omega)\,t_\beta(k_2q\omega)=K_\alpha(k_2q)\ . \tag{9}$$

Here
$$K_\alpha(k_2q)=1 \qquad \alpha=0$$
$$=\cos(\vec{k}_2\cdot\vec{\delta}_i)-\cos(\tfrac{1}{2}\vec{q}\cdot\vec{\delta}_i),\quad \alpha=\delta_i \tag{10}$$

and the matrix $\underline{M}$ may be written in terms of the lattice sums

$$I_o(q,\omega)=N^{-1}\sum_k\left[\tfrac{1}{2}z(\tilde{\omega}-1)+\sum_\delta\cos(\vec{k}\cdot\vec{\delta})\cos(\tfrac{1}{2}\vec{q}\cdot\vec{\delta})\right]^{-1}, \quad (11a)$$

$$I_j(q,\omega)=N^{-1}\sum_k\cos(\vec{k}\cdot\vec{\delta}_j)\left[\tfrac{1}{2}z(\tilde{\omega}-1)+\sum_\delta\cos(\vec{k}\cdot\vec{\delta})\cos(\tfrac{1}{2}\vec{q}\cdot\vec{\delta})\right]^{-1}, \quad (11b)$$

$$I_{mj}(q,\omega)=N^{-1}\sum_k\cos(\vec{k}\cdot\vec{\delta}_m)\cos(\vec{k}\cdot\vec{\delta}_j)\times\left[\tfrac{1}{2}z(\tilde{\omega}-1)+\sum_\delta\cos(\vec{k}\cdot\vec{\delta})\cos(\tfrac{1}{2}\vec{q}\cdot\vec{\delta})\right]^{-1}, \quad (11c)$$

where $\tilde{\omega}\equiv[\omega-4D(S-\frac{1}{2})-2h]/(2JzS)$.

We obtain for the matrix $\underline{M}$

$$\underline{M}(q,\omega)=\begin{bmatrix}1+(D/2JS)I_o(q,\omega) & (2S)^{-1}[I_i(q,\omega)] \\ (D/2JS)[I_i(q,\omega)-\cos(\frac{1}{2}\vec{q}\cdot\vec{\delta}_i)I_o(q,\omega)] & \delta_{ij}+(2S)^{-1}[I_{ij}(q,\omega)-\cos(\frac{1}{2}\vec{q}\cdot\vec{\delta}_i)I_j(q,\omega)]\end{bmatrix}, \quad (12)$$

where δ_{ij} is the Kronecker δ. The solution of Eq. (7) will have singularities within the usual two-spin-wave continuum[3,4,8,9] and at the solutions of $|\underline{M}(q,\omega)|=0$. The latter solutions are the bound states. From Eq. (12), we see that for $D=0$ this condition gives

$$\left|\delta_{ij}+(2S)^{-1}[I_{ij}(q,\omega)-\cos(\tfrac{1}{2}\vec{q}\cdot\vec{\delta}_i)I_j(q,\omega)]\right|=0, \quad (13)$$

in agreement with all of the previously referenced calculations.

A point at which a trivial evaluation of Eq. (12) may be made is at the zone corner $\vec{q}\cdot\vec{\delta}=\pi$ (all $\vec{\delta}$). There $I_i(q,\omega)=0$, and we have one solution of $|\underline{M}(q,\omega)|=0$ at[10]

$$\omega=4D(S-\tfrac{1}{2})+2h+2JzS-2D\ , \quad (14a)$$

and d solutions at

$$\omega=4D(S-\tfrac{1}{2})+2h+2JzS-J\ , \quad (14b)$$

where d is the dimensionality of the lattice. Equation (14b) represents the usual (exchange) bound states, and Eq. (14a) the single-ion bound state. Since the two-spin-wave continuum ($\omega=\epsilon_{q/2+k}+\epsilon_{q/2-k}$) narrows to a single energy $\omega=4D(S-\frac{1}{2})+2h+2JzS$ at the zone corner, these are just the (Ising) bound states alluded to above. In the following, we will evaluate the bound-state condition $|\underline{M}(q,\omega)|=0$ for various values of S and D/J in one and three dimensions. The two-dimensional case is omitted for simplicity only, and in the three-dimensional case we will treat a simple cubic lattice for the purposes of numerical evaluation. We present our results for one dimension in Sec. III and those for three dimensions in Sec. IV. Section V is devoted to conclusions and the relation of the present work to experiment.

III. BOUND STATES IN ONE DIMENSION

In the one-dimensional case the lattice sums in Eq. (11) may be found from an elementary integral.[3] We find with $z=2$ and the definitions $\alpha=\cos(\frac{1}{2}qa)$ and $\gamma=(\tilde{\omega}-1)/\alpha$ [see the definition of $\tilde{\omega}$ after Eq. (11c)] and for $|\gamma|>1$ (outside the band)

$$I_o(q,\omega)=\pm\alpha^{-1}(\gamma^2-1)^{-1/2}\ , \quad (15a)$$

$$I_i(q,\omega)=\alpha^{-1}-\gamma I_o(q,\omega)\ , \quad (15b)$$

$$I_{ii}(q,\omega)=-\gamma I_i(q,\omega)\ , \quad (15c)$$

where the plus sign is used above the band ($\gamma>+1$) and the minus sign is used below ($\gamma<-1$). Using Eqs. (12) and (15), one finds that the condition $|\underline{M}(q,\omega)|=0$ yields

$$a(\gamma)\alpha^2+b(\gamma)\alpha+c(\gamma)=0\ , \quad (16)$$

where

$$a(\gamma)=1-(2S)^{-1}[1-\gamma D_0(\gamma)]\ , \quad (17a)$$

$$b(\gamma)=(D/2JS)D_0(\gamma)-(\gamma/2S)[1-\gamma D_0(\gamma)]\ , \quad (17b)$$

$$c(\gamma)=-(D/4JS^2)[1-\gamma D_0(\gamma)]\ , \quad (17c)$$

and

$$D_0(\gamma)\equiv\pm(\gamma^2-1)^{1/2}\ . \quad (18)$$

Equation (16) has two solutions with positive α for any $\gamma<-1$, one corresponding to the exchange bound state,[3] the other to the single-ion bound state; the lower bound state always exists at $q=0$ ($\alpha=1$).[11] For large negative γ both solutions approach $\alpha=0$ or $q=\pi$, with binding energies J and $2D$, respectively, as predicted by Eq. (14) and the Ising model. A numerical evaluation of Eq. (16) for $S=1$, $D=1$, and $h=0$ is shown in Fig. 1. The single-ion bound state is seen to be present at $q=0$, while the exchange bound state does not appear until about $q=\frac{5}{6}q_{max}$.

IV. BOUND STATES IN THREE DIMENSIONS

In the three-dimensional case $\underline{M}(q,\omega)$ is a 4×4 matrix. However, for $\vec{q}$ along the [111] direction of a simple cubic lattice there are only five distinct elements, M_{11}, M_{22}, M_{12}, M_{21}, and M_{23}. Moreover, from Eq. (12) we see that, for $D=0$, $M_{11}=1$

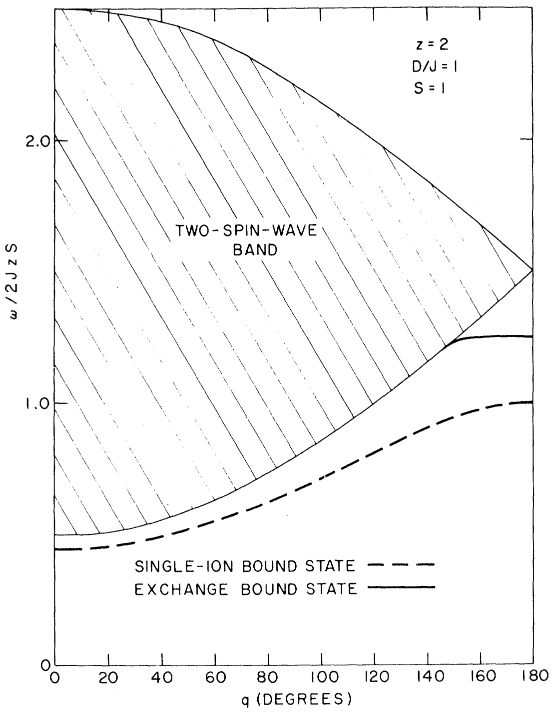

FIG. 1. Manifold of two-spin-wave states in one dimension with $S=1$ and $D=J$.

and $M_{21}=0$, so that in this limit the bound state condition $|\underline{M}|=0$ yields

$$(M_{22}-M_{23})^2(M_{22}+2M_{23})=0, \quad D=0 \ . \tag{19}$$

These are the bound states found by previous authors,[3,4] and are plotted for the purpose of later comparisons in Fig. 2 for $S=1$. The lower curve represents the singly degenerate (s-wave) state, while the upper curve represents the doubly degenerate (d-wave) state. The dashed line represents a resonant d-wave state [solution of the real part of Eq. (19)] within the two-spin-wave band. The presence of the resonant state, whose damping increases as it penetrates into the band, was first demonstrated in Ref. 9 and was shown in Ref. 8 to lead to an anomalous single spin-wave self-energy. This resonant d-wave state is not included in the rest of the figures.

For arbitrary D the bound-state condition $|\underline{M}|=0$ gives

$$(M_{22}-M_{23})^2\,[(M_{22}+2M_{23})M_{11}-3M_{12}M_{21}]=0 \ . \tag{20}$$

From Eq. (20) we see that the d-wave bound state, whose energy is determined by $M_{22}=M_{23}$, is unaffected by the single-ion anisotropy. The other bound-state condition ([ ] = 0) obtained from Eq. (20), which displays a mixing between the s-wave and single-ion bound states, may be written as

$$a(\lambda)\,\alpha^2+b(\lambda)\,\alpha+c(\lambda)=0 \ , \tag{21}$$

where $a(\lambda)=1-(3/2S)D_1(\lambda)$, (22a)

$$b(\lambda)=(D/2JS)D_0(\lambda)-(\lambda/2S)D_1(\lambda), \tag{22b}$$

$$c(\lambda)=-(D/4JS^2)D_1(\lambda)\,[\lambda D_0(\lambda)+3D_1(\lambda)]\,, \tag{22c}$$

with $\alpha=\cos(\tfrac{1}{2}q_i a)$, (23a)

$$\lambda=3(\bar{\omega}-1)/\alpha \ , \tag{23b}$$

$$D_0(\lambda)=\alpha\, I_0(q,\omega) \ , \tag{23c}$$

$$D_1(\lambda)=\alpha\, I_i(q,\omega) \ . \tag{23d}$$

The functions $I_0(q,\omega)$ and $I_i(q,\omega)$ have been computed numerically using summation formulas outside the band ($|\lambda|>3$) and Bessel-function representations inside the band ($|\lambda|<3$).[8] We have used these results to evaluate Eq. (21) for several values of S and D/J, in order to explicitly display the behavior of the bound states. For simplicity, we have taken the external magnetic field h to be zero.

In Fig. 3, we show the two-spin-wave states for $D=\frac{1}{4}J$ and $S=1$. Figure 3 (as well as Fig. 4 to follow) represents the blown-up region of the Brillouin zone which is enclosed by the dashed lines in Fig. 2. This is done to emphasize the bound-state

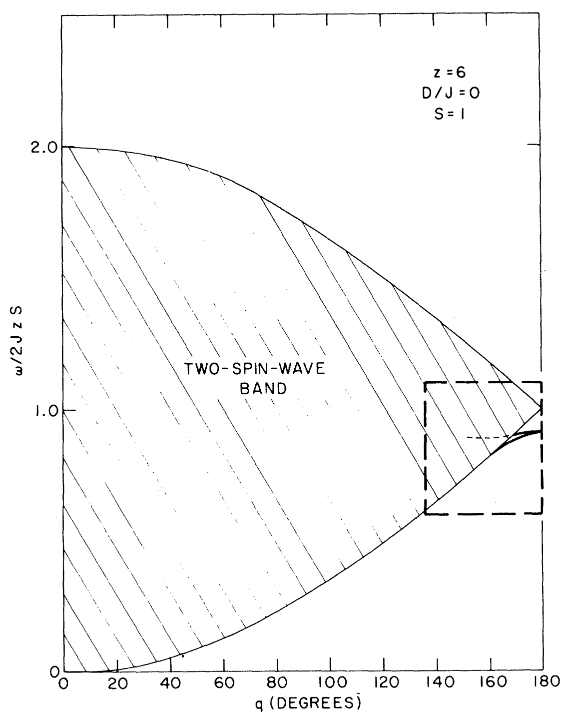

FIG. 2. Manifold of two-spin-wave states for a simple cubic lattice with $S=1$ and $D=0$. Bound states are found only in the dashed box near the zone corner. Here "$q=d$ degrees" means $q_x a=q_y a=q_z a=d\pi/180$.

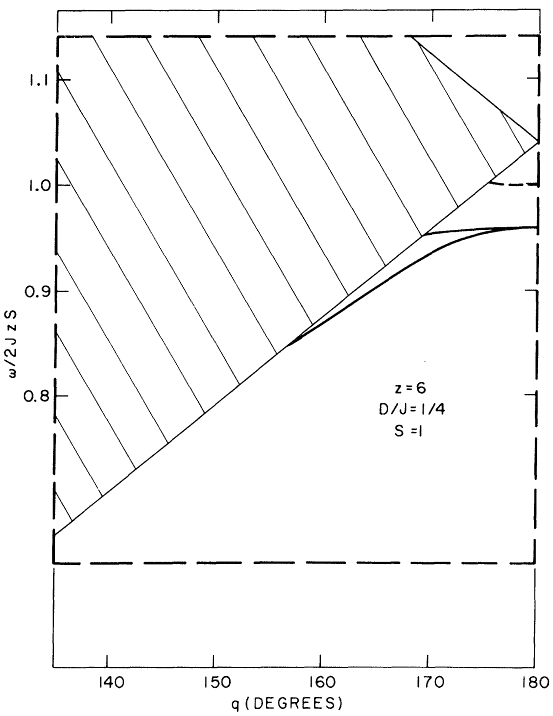

FIG. 3. Two-spin-wave states for $S=1$ and $D/J=\frac{1}{4}$. Note that this is an enlargement of the dashed box in Fig. 2. The single-ion bound state is shown as a heavy dashed line, while the exchange bound states are the heavy solid lines.

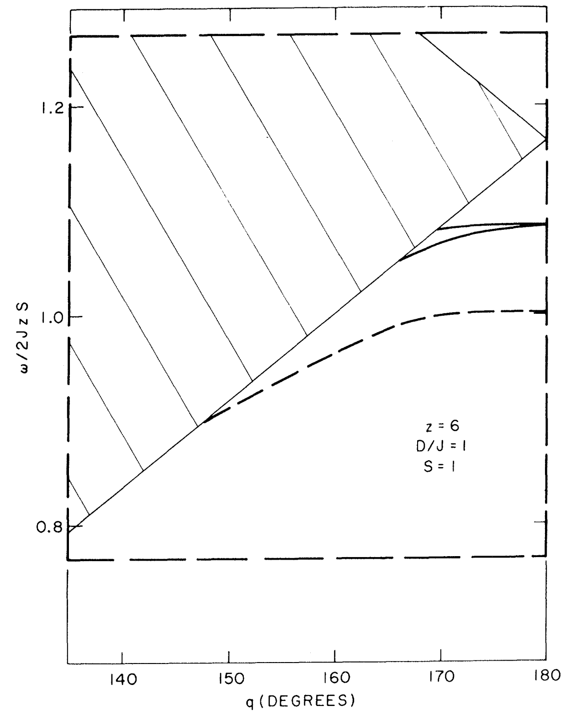

FIG. 4. Two-spin-wave states for $S=1$ and $D=J$, showing the same region of the Brillouin zone as in Fig. 3.

structure which, for the parameters used in these figures, occurs relatively near the zone corner. For $D=\frac{1}{4}J$ the single-ion bound state, which is denoted in all the figures by a heavy dashed line, is higher in energy than the exchange bound states and has little effect on the s-wave state (recall that there is no coupling to the d-wave state). However, for $D=J$ and $S=1$ (Fig. 4), the single-ion bound state has already become the lowest bound state and has a threshold somewhat smaller than that of the s-wave state for $D=0$ (Fig. 2).

As the ratio of D to J is increased further the single-ion bound state becomes more tightly bound and eventually appears at $q=0$. This behavior is illustrated for the $S=1$ case by Figs. 5 and 6, where it is no longer necessary to enlarge the scale as was done in Figs. 3 and 4 in order to clearly see the single-ion bound state. In Fig. 5 the lighter dashed line within the band represents a resonant single-ion state similar to the resonant d-wave state mentioned earlier. It represents a solution of the real part of Eq. (20) within the two-spin-wave band, whose damping increases as it penetrates deeper into the band and as the imaginary parts of $I_0(q,\omega)$ and $I_i(q,\omega)$ build up. We note here that for $D=0$ the s-wave exchange bound state does

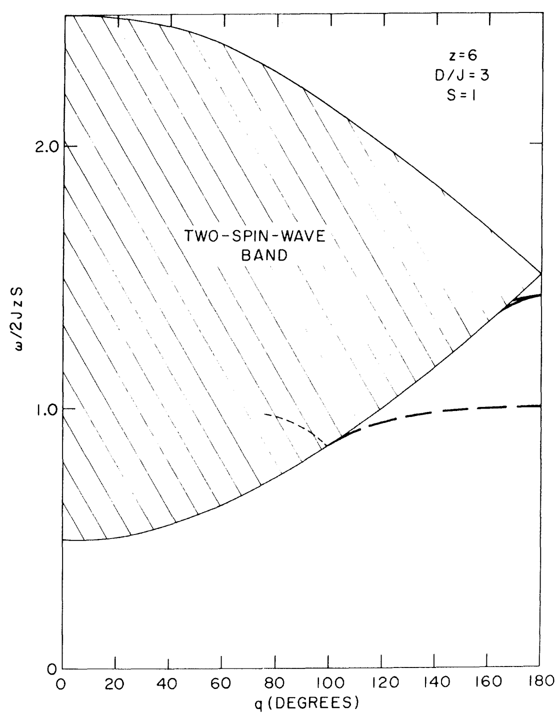

FIG. 5. Two-spin-wave states for $S=1$ and $D/J=3$, with notation as in Fig. 3, but showing all q. The lighter dashed line within the band represents a resonant single-ion state.

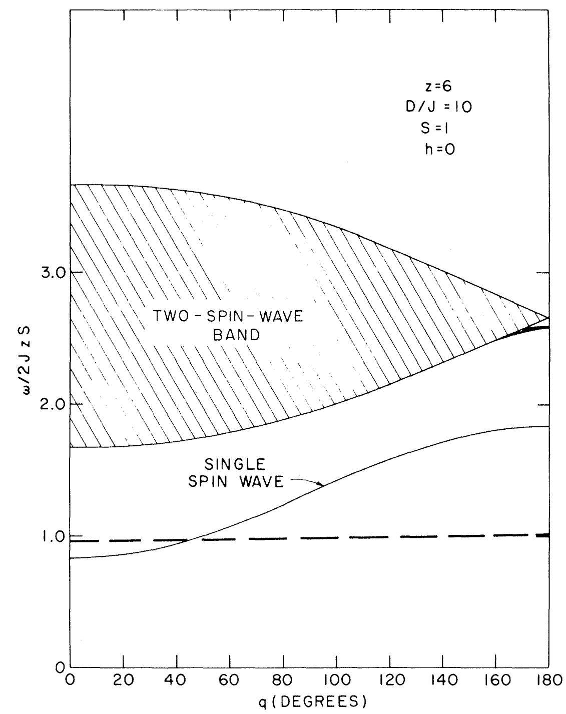

FIG. 6. Two-spin-wave states for $S=1$, $D/J=10$, and $h=0$, with single spin wave superimposed. The notation is as in Fig. 3.

not extend to smaller q within the band as a resonant state, and that for small D/J, neither does the single-ion solution of Eq. (20). However, for D/J large enough to separate the single-ion solution from the s-wave solution, but too small to cause it to exist at the center of the zone ($q=0$), a resonant state does exist at wave vectors smaller than the threshold, as shown in Fig. 5.

The large-D/J case is displayed in Fig. 6, where $S=1$ and $D=10J$. Here the single-ion bound state is split considerably below the two-spin-wave band, even at $q=0$, and appears to be almost dispersionless. The single-spin-wave state for zero magnetic field is shown in the same figure, and we note that it is higher in energy than the single-ion bound state over most of the Brillouin zone. Note from Fig. 6 that in this large-D/J region there is a crossing of the single-spin-wave and single-ion bound-state energies. The use of an external magnetic field would make experimental observation of this crossover considerably simpler, since the single-ion bound state shifts by $2h$, while the single-spin-wave state shifts by only h, so that one may "tune" the levels. If D/J were somewhat larger, the single-ion bound state would be the lowest excitation for all q. In this limit, we have effectively a two-level system, which is often described by a pseudospin $S'=\frac{1}{2}$.

In the final figure, Fig. 7, we have chosen $D=3J$ and $S=\frac{5}{2}$. Comparison with Fig. 5 shows that the interactions between spin waves become weaker for larger spin, as expected. However, the single-ion bound state is still significantly more bound than the exchange bound states, and an increase in D/J would yield the same type of behavior as that shown above for the $S=1$ case. That is, even for large spin, there is a single-ion bound state within a significant volume of the Brillouin zone for sufficiently large D/J.

V. CONCLUSIONS

We have shown above that single-ion anisotropy gives rise to an additional two-magnon bound state. The energy of this state has been calculated for various values of S and D/J in one and three dimensions. In all cases, the effect of the single-ion anisotropy is to *lower* the energy of the lowest two-spin-wave bound state. For sufficiently large D/J the single-ion bound state is found to exist throughout the Brillouin zone.

More generally, we would expect to find single-ion bound states not only in ferromagnets, but also in some anti- and ferrimagnetic systems, for example, those with $D \gg J$, where $-D(S^z)^2$ is the dom-

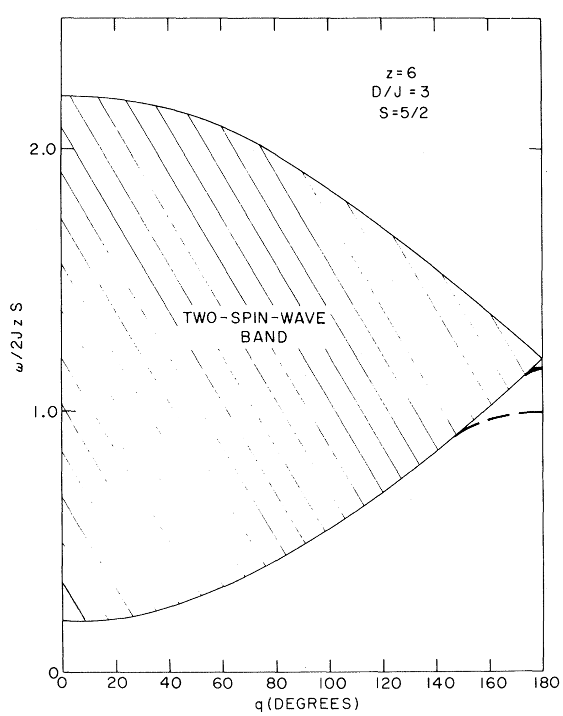

FIG. 7. Two-spin-wave states for $S=\frac{5}{2}$ and $D/J=3$. The notation is as in Fig. 3.

inant term in the Hamiltonian. In addition, for $S >> 1$, single-ion bound states containing more than two spin deviations may exist.[12] For example, the state with $2S$ spin deviations on the same site certainly exists for $D >> J$, and in fact becomes the lowest-lying excitation. This is simply the usual single-ion limit often described by a pseudospin $S' = \frac{1}{2}$. In the case $S = 1$, there is only the two-magnon single-ion bound state. We have calculated its energy for arbitrary D/J, i.e., from $D = 0$ continuously to the pseudospin limit, where it becomes lower in energy than the single-spin-wave state.

As a final point, we discuss the possible experimental consequences of the above work. The most significant result in this respect is that in a uniaxial ferromagnet with single-ion anisotropy, two-magnon bound states exist over a larger volume of the Brillouin zone than in the isotropic case, thus improving the possibility of their observation. This behavior was demonstrated by Wortis[3] for the case of exchange anisotropy, and exchange bound states have in fact been observed[13] at $k = 0$ in the linear chains of $CoCl_2 \cdot 2H_2O$, which has strong exchange anisotropy. In systems with strong single-ion anisotropy, we predict the presence of an additional (single-ion) bound state which should also be observable at $k = 0$ in an appropriate material. These effects would be most dramatic in a $S = 1$ linear chain system with $D >> J$. One possible mechanism for direct observation of the single-ion bound state is photon absorption proceeding via transverse anisotropy which breaks the $\Delta m = 1$ selection rule.[13,14]

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