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SPIN WAVES IN SYSTEMS WITH WEAK ANTIFERROMAGNETIC EXCHANGE FIELDS*

By M. Cieplak**

Physics and Astronomy Department of the University of Pittsburgh
and Institute of Theoretical Physics, Warsaw University

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Four-boson spin-waves are constructed to represent the low-temperature collective states of spin-1 systems with strong single-ion uniaxial and orthorhombic anisotropy fields and with weak antiferromagnetic exchange coupling. Necessary conditions for the stability of such systems and the phase boundaries are derived. Effects of an r.f. field are investigated. These effects are different from those characteristic for systems with weak ferromagnetic coupling. In particular some of the parallel pumping processes are absent and new small-energy incoherent resonances appear.

1. *Introduction*

Recently Cieplak and Keffer [1] have presented a multi-boson spin-wave theory to describe the low-temperature behaviour of paramagnets, ferromagnets, and antiferromagnets whose spins are located in strong single-ion anisotropy fields and are coupled by weak exchange interactions. The different sets of bosons are particles excitable to the different eigenstates of the single-ion part of the hamiltonian, and boson representations of the spin operators are constructed by a matrix-elements-matching method. The method has been applied to the spin-1 systems with uniaxial (either easy or hard axis) and orthorhombic anisotropies, and to the spin-2 systems with cubic anisotropy. The exchange interaction has been assumed ferromagnetic-like. Subsequently, effects of oscillatory magnetic fields have been investigated. It turns out that such a field can produce three phenomena: a parallel pumping of magnon-pairs, a coherent resonance with $k = 0$ magnons, and an incoherent resonance absorption between the excited states. In one domain of parameters (so-called region S) magnon relaxation times have also been estimated.

Most of the known spin-1 systems with dominant uniaxial and orthorhombic anisotropies, of strength D and E respectively, turn out, however, to be weakly coupled by an

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** Present address: Instytut Fizyki Teoretycznej, Uniwersytet Warszawski, Hoża 69, 00-681 Warszawa, Poland.

antiferromagnetic-like exchange. These are the Ni^{++} compounds. The typical example is the salt $NiSnCl_6 \cdot 6H_2O$, studied by How and Svare [2] and by Friedberg and co--workers [3]. This salt has, at 4.2 K, $D/k_B = +0.65$ K, $|E|/k_B \lesssim 0.07$ K and the antiferromagnetic exchange constant $J/k_B \approx 0.02$ K. Other examples are $NiCl_2 \cdot 4H_2O$, investigated by McElearney et al. [4], with $D/k_B = -11.5$ K, $E/k_B = 0.1$ K, and $2J/k_B \approx 5.25$ K; $Ni(NO_3)_2 \cdot 6H_2O$ [5] with $D/k_B = +6.43$ K, $E/k_B = +1.63$ K, and $2J/k_B \approx 0.6$ K; $Ni(CH_3COO)_2 \cdot 4H_2O$ [6] with $D/k_B = 5.79$ K, $E/k_B = 2.27$ K, and $2J/k_B = 0.08$ K (below 4.2 K).

It seems therefore desirable to describe the collective states of the spin-1 systems with a weak antiferromagnetic coupling. We shall do this in the present paper using the method of reference [1]. We shall see that the r.f.-field-effects pattern is in several respects unlike the one for systems with a weak ferromagnetic coupling. The major difference is the occurence of a new incoherent resonance absorption at a small, proportional to J, energy which corresponds to transitions that essentially flip sublattices. The other difference is that, when the r.f. field is applied parallel to the uniaxis, some of the pumping processes disappear. The absent processes are those in which magnons of the same kind are excited.

In this paper we shall discuss the case of the static field applied parallel to the uniaxis, so the hamiltonian under study is

$$\mathscr{H} = \sum_i \{D(S_i^z)^2 + E[(S_i^x)^2 - (S_i^y)^2] - HS_i^z\} + 2J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j. \tag{1.1}$$

H denotes the magnetic field multiplied by the Bohr magneton and by the Landé factor. The anisotropy constants can have either sign. The J-constant is positive and much smaller than either $|D|$, or $|E|$, or H. The exchange coupling extends between the z nearest neighbors.

If $D > 0$, $|E| < D$, and $H \leqslant H_{c1} = (D^2 - E^2)^{1/2} - O(J)$ the ground state of the system is a singlet with zero energy. The two sublattice moments are therefore formed by local excitations, from the singlet state, in opposite directions. Following reference [1] we shall call this region S (small magnetic fields). On the other hand if $H \geqslant H_{c2} = (D^2 - E^2)^{1/2} + O(J)$ with $D > 0$, $|E| < D$ the ground state is a product of the states $|-\rangle_i$:

$$|-\rangle_i = (1/\sqrt{2})\mathscr{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}|+1\rangle_i - \eta[(H^2+E^2)^{1/2}-H]^{1/2}|-1\rangle_i\}, \tag{1.2}$$

which are the eigenstates of the single-ion part of the hamiltonian, corresponding to the eigenvalue $D-(H^2+E^2)^{1/2}$, and where

$$\mathscr{N} = (H^2+E^2)^{-1/4},$$

$$\eta = \begin{cases} +1 & \text{for } E \geqslant 0, \\ -1 & \text{for } E < 0. \end{cases}$$

In this case we speak of the region L (large magnetic fields). In the region L the sublattices have nonvanishing moments along the z-axis. The two regions are separated by a small intermediate region of width proportional to J, where a canted configuration [7, 8] should be present. In this region the two lowest energy levels are heavily mixed by exchange and our theory does not apply there. If $D < 0$ or if $|E| \geqslant |D|$ then the system is in the region L at any value of the magnetic field.

In Sec. 2 we shall construct the spin-wave theory for the region S and the stability criteria will be derived. Subsequently we shall discuss the possible effects of the r.f. field.
In Sec. 3 the analysis for the region L is presented.

2. *Region S*

Following reference [1] we introduce two sets of Bose-operators, a_i and b_i, for the $\frac{1}{2}N$ sites of the first sublattice, and two sets of operators, A_j and B_j, for the second sublattice. The a_i^+ and A_j^+ operators excite the $|-\rangle_{i(j)}$ states out of the ground state, $|0\rangle_{i(j)}$. On the other hand b_i^+ and B_j^+ excite the third and the highest, state of energy $D+(H^2+E^2)^{1/2}$ out of $|0\rangle_{i(j)}$. The spin-operators representation can be obtained by the operator-matching method and it reads

$$S_i^z = \mathcal{N}^2[H(a_i^+a_i - b_i^+b_i) + E(a_i^+b_i + b_i^+a_i)], \tag{2.1a}$$

$$S_i^+ = \mathcal{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}(a_i^+ + b_i) + \eta[(H^2+E^2)^{1/2}-H]^{1/2}(b_i^+ - a_i)\} + \dots, \tag{2.1b}$$

$$S_i^- = (S_i^+)^\dagger, \tag{2.1c}$$

and

$$S_j^z = -\mathcal{N}^2[H(A_j^+A_j - B_j^+B_j) + E(A_j^+B_j + B_j^+A_j)], \tag{2.2a}$$

$$S_j^+ = \mathcal{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}(B_j^+ + A_j) + \eta[(H^2+E^2)^{1/2}-H]^{1/2}(B_j - A_j^+)\} + \dots, \tag{2.2b}$$

$$S_j^- = (S_j^+)^\dagger. \tag{2.2c}$$

The cubic and higher order terms in (2.1b) and (2.2b) have been omitted.
In the harmonic approximation the spin-wave hamiltonian becomes

$$\begin{aligned}\mathcal{H} = \sum_k \{&[D-(H^2+E^2)^{1/2}]\,(a_k^+a_k + A_k^+A_k) \\ &+[D+(H^2+E^2)^{1/2}]\,(b_k^+b_k + B_k^+B_k) \\ &+2J\gamma(k)\,(a_k^+A_{-k}^+ + a_kA_{-k} + b_k^+B_{-k}^+ + b_kB_{-k}) \\ &+2JE\mathcal{N}^2\gamma(k)\,(b_k^+B_k + B_k^+b_k - a_k^+A_k - A_k^+a_k) \\ &+2JH\mathcal{N}^2\gamma(k)\,(a_k^+B_k + B_k^+a_k + b_k^+A_k + b_kA_k^+)\} + \dots,\end{aligned} \tag{2.3}$$

where

$$a_k = (\tfrac{1}{2}N)^{1/2}\sum_i e^{-ikR_i}a_i,$$

and

$$\gamma(k) = \sum_\delta e^{ik\delta}$$

with δ denoting the positions of the nearest neighbors. The form of the quartic and higher order single-ion terms, if needed, can be obtained by employing the correspondence between our spin-representation and the rigorous one due to Homma et al. [9], which has also been used by Ishikawa and Oguchi [10].

In order to find the spin-wave energies we found it convenient to perform first a partial diagonalization of (2.3) without the terms proportional to $2JH\mathcal{N}^2\gamma(k)$. As a final result we obtain the following four branches of energies

$$\varepsilon_{1,2}(k) = \{D^2+H^2+E^2-4JE\gamma(k)-8J^2H^2\mathcal{N}^4\gamma^2(k) \pm 2D[H^2+E^2-4JE\gamma(k)+4J^2\gamma^2(k)]^{1/2}\}^{1/2}, \tag{2.4a}$$

and

$$\varepsilon_{3,4}(k) = \{D^2+H^2+E^2+4JE\gamma(k)-8J^2H^2\mathcal{N}^4\gamma^2(k) \pm 2D[H^2+E^2+4JE\gamma(k)+4J^2\gamma^2(k)]^{1/2}\}^{1/2}. \tag{2.4b}$$

These four modes merge into two in the absence of the orthorhombic anisotropy. Consider now the case of the vanishing magnetic field. For positive E the energy ε_4 is minimal. On the other hand for negative $E-\varepsilon_2$ is minimal. The corresponding $k = 0$ magnons go soft when

$$4Jz = D-|E|. \tag{2.5}$$

It means that the theory applies whenever $4Jz$ is smaller than $D-|E|$. For slightly bigger J the system should be in the intermediate phase and for even bigger exchange the system becomes antiferromagnetic. Note that the condition (2.5) coincides exactly with Moriya's [11] criterion for a long range order given in his study of the antiferromagnetism of NiF_2.

An inspection of the energies (2.4) leads to the conclusion that the system becomes unstable when $H = H_{c1}$:

$$H_{c1} = (D^2-E^2)^{1/2}-2[(D+|E|)/(D-|E|]^{1/2}Jz+O(J^2). \tag{2.6}$$

This marks the upper boundary for the region S. The equation (2.6) coincides, in the first order with respect to J, with the critical field H_{c1} for systems with a weak ferromagnetic coupling.

If we neglect terms proportional to J^2, the hamiltonian (2.3) can be brought into its diagonal form:

$$\mathcal{H} = \sum_k [\varepsilon^c(k)c_k^+c_k+\varepsilon^C(k)C_k^+C_k+\varepsilon^d(k)d_k^+d_k+\varepsilon^D(k)D_k^+D_k]+O(J^2)+\ \dots, \tag{2.7}$$

with

$$\varepsilon^{c,C}(k) = D-(H^2+E^2)^{1/2}\pm 2JE\mathcal{N}^2\gamma(k) = \varepsilon_{2,4}(k)+O(J^2), \tag{2.8a}$$

and

$$\varepsilon^{d,D}(k) = D+(H^2+E^2)^{1/2}\mp 2JE\mathcal{N}^2\gamma(k) = \varepsilon_{1,3}(k)+O(J^2), \tag{2.8b}$$

by means of the following transformation

$$a_k = \frac{1}{\sqrt{2}}\,[c_k + C_k + p_-(c^+_{-k} - C^+_{-k}) + q(d_k - D_k)], \tag{2.9a}$$

$$A_k = \frac{1}{\sqrt{2}}\,[C_k - c_k - p_-(c^+_{-k} + C^+_{-k}) - q(d_k + D_k)], \tag{2.9b}$$

$$b_k = \frac{1}{\sqrt{2}}\,[d_k + D_k + p_+(d^+_{-k} - D^+_{-k}) + q(c_k - C_k)], \tag{2.9c}$$

$$B_k = \frac{1}{\sqrt{2}}\,[D_k - d_k - p_+(d^+_{-k} + D^+_{-k}) - q(c_k + C_k)], \tag{2.9d}$$

where

$$p_\pm = J[D \pm (H^2 + E^2)^{1/2}]^{-1}\gamma(k), \tag{2.10a}$$

and

$$q = JHD^{-1}\mathcal{N}^2\gamma(k). \tag{2.10b}$$

Consider an oscillatory field of frequency ω applied along the z-axis. The field will couple to the two sublattices simultaneously, i.e. it will couple to

$$\begin{aligned}
&\sum_i S^z_i + \sum_j S^z_j \\
&= \sum_k \{\mathcal{N}^2 H(c^+_k C_k + C^+_k c_k - d^+_k D_k - D^+_k d_k) \\
&+ \mathcal{N}^2[E + 2JH^2 D^{-1}\mathcal{N}^2\gamma(k)]\,(C^+_k d_k + d^+_k C_k) \\
&+ \mathcal{N}^2[E - 2JH^2 D^{-1}\mathcal{N}^2\gamma(k)]\,(c^+_k D_k + D^+_k c_k) \\
&+ 2JE(D^2 - H^2 - E^2)^{-1}\gamma(k)\,(c^+_k D^+_{-k} + D_{-k}c_k - d^+_k C^+_{-k} - d_k C_{-k}) + O(J^2).
\end{aligned} \tag{2.11}$$

The field will therefore trigger the following processes:

(1) The incoherent resonance [1] at a very small frequency $\omega = \varepsilon^c(k) - \varepsilon^C(k) = \varepsilon^D(k) - \varepsilon^d(k) = 4JE\mathcal{N}^2\gamma(k)$. This is the process in which a c_k-magnon is replaced by a C_k-one and similarly d_k by D_k (with the inverse processes also present). This resonance disappears at $H = 0$. It is also absent when the coupling is ferromagnetic-like, since there is no distinction between the sublattices then. A study of such a resonance may supply additional information about the parameters in the hamiltonian. However the net absorption will be small since it is proportional to the difference in population between the c and C-levels and none of them is the ground state (in addition they differ only slightly in energy).

(2) The incoherent resonance at $\omega = \varepsilon^d(k) - \varepsilon^C(k) = \varepsilon^D(k) - \varepsilon^c(k) = 2(H^2 + E^2)^{1/2}$. In this process, which is very similar to the one found in the system with weak ferromagnetic coupling, the whole spectrum of the C-magnons is replaced by the d-ones, and similarly c- by D-magnons.

(3) Parallel pumping of the c_k, D_{-k} particles at $\omega = \varepsilon^c(k) + \varepsilon^D(k) = 2D + 4JE\mathcal{N}^2\gamma(k)$ and parallel pumping of the C_{-k}, d_k particles at $\omega = 2D - 4JE\mathcal{N}^2\gamma(k)$. These processes

are similar to the pumping of unlike magnons in systems with weak ferromagnetic exchange. On the basis of an analysis presented in [1] we expect that at $H = 0$ and at very low temperatures the threshold amplitude of the r.f. field triggering the significant power absorption is at least equal to D. At temperatures comparable to $D-|E|$ the process requires much more intense fields, namely proportional to $D|E|J^{-1}$.

Note that, unlike the ferromagnetic coupling case, no pumping of the types $c_{-k}^{+}c_{k}^{+}$ or $C_{-k}^{+}C_{k}^{+}$, or $C_{-k}^{+}c_{k}^{+}$ appear. Only pairs of excitations to essentially different levels can be pumped.

If the r.f. field is applied along the x-axis it can produce a coherent resonance with the C_0 and D_0-magnons. On the other hand if it is applied along the y-axis a coherent resonance with the c_0 and d_0-magnons will occur.

3. *Region L*

In this region we again introduce four sets of Bose operators. The operators a_i^+ and A_j^+ excite the $|0\rangle_{i(j)}$ state out of the $|-\rangle_{i(j)}$ state and the operators b_i^+ and B_j^+ excite the third level. (The relative positions of the two excited states may switch under the influence of the field.) The operator-matching method yields

$$S_i^z = \mathcal{N}^2[H+E(b_i^+ + b_i) - Ha_i^+ a_i - 2Hb_i^+ b_i] + \dots, \tag{3.1a}$$

$$S_i^+ = \mathcal{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}(a_i + a_i^+ b_i) - \eta[(H^2+E^2)^{1/2}-H]^{1/2}(a_i^+ - b_i^+ a_i) + \dots, \tag{3.1b}$$

$$S_i^- = (S_i^+)^\dagger \tag{3.1c}$$

and

$$S_j^z = -\mathcal{N}^2[H+E(B_j^+ + B_j) - HA_j^+ A_j - 2HB_j^+ B_j] + \dots, \tag{3.2a}$$

$$S_j^+ = \mathcal{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}(A_j^+ + B_j^+ A_j) - \eta[(H^2+E^2)^{1/2}-H]^{1/2}(A_j - A_j^+ B_j) + \dots, \tag{3.2b}$$

$$S_j^- = (S_j^+)^\dagger. \tag{3.2c}$$

The spin-wave hamiltonian becomes

$$\begin{aligned}\mathcal{H} = {} & N[D-(H^2+E^2)^{1/2} - J\mathcal{N}^4 H^2 z] \\ & -2J(\tfrac{1}{2}N)^{1/2}\mathcal{N}^4 HEz(b_0^+ + b_0 + B_0^+ + B_0) \\ & +[(H^2+E^2)^{1/2} - D + 2J\mathcal{N}^4 H^2 z]\sum_k (a_k^+ a_k + A_k^+ A_k) \\ & +2J\sum_k \gamma(k)\,[a_k^+ A_{-k}^+ + a_k A_{-k} - E\mathcal{N}^2(a_k^+ A_k + A_k^+ a_k)] \\ & +[2(H^2+E^2)^{1/2} + 4Jz\mathcal{N}^4 H^2]\sum_k (b_k^+ b_k + B_k^+ B_k) \\ & -2J\mathcal{N}^4 E^2 \sum_k \gamma(k)\,(b_k^+ B_{-k}^+ + B_k b_{-k} + B_k^+ b_k + b_k^+ B_k) + \dots .\end{aligned} \tag{3.3}$$

The linear terms can be eliminated by the transformation

$$b_k = \tilde{b}_k + (\tfrac{1}{2} N)^{1/2} JHEz\mathcal{N}^6 + O(J^2), \tag{3.4a}$$

$$B_k = \tilde{B}_k + (\tfrac{1}{2} N)^{1/2} JHEz\mathcal{N}^6 + O(J^2). \tag{3.4b}$$

Due to the presence of the triple interactions, which should be written down in (3.3), the transformation (3.4) introduces new harmonic terms to the hamiltonian. These are however proportional to J^2 and will be neglected. The hamiltonian rewritten in terms of the operators $\tilde{b}_k$ and $\tilde{B}_k$ looks like (3.3) with the linear terms missing.

The four branches of the spin-waves excitations have the energies

$$\varepsilon_{1,2}(k) = \{[(H^2+E^2)^{1/2} - D + 2Jz\mathcal{N}^4 H^2 \pm 2JE\mathcal{N}^2\gamma(k)]^2 - 4J^2\gamma^2(k)\}^{1/2}, \tag{3.5a}$$

and

$$\varepsilon_{3,4}(k) = 2\{[(H^2+E^2)^{1/2} + 2Jz\mathcal{N}^4 H^2 \pm J\mathcal{N}^4 E^2\gamma(k)]^2 - J^2\mathcal{N}^8 E^4\gamma^2(k)\}^{1/2}. \tag{3.5b}$$

Again the four branches degenerate into two if $E = 0$.

The inspection of energies (3.5) leads to the following conclusions. If $D > 0$ with $|E| < D$, then the configuration of region L becomes unstable at $H = H_{c2}$:

$$H_{c2} = (D^2 - E^2)^{1/2} + 2Jz|E|D^{-1}[(D+|E|)/(D-|E|)] + O(J^2). \tag{3.6}$$

This is the lower boundary for the region L. If $|E| > |D|$ then at $H = 0$ the system remains paramagnetic if $4Jz < |E| - D$. For slightly bigger J our theory does not apply at $H = 0$ since the two lowest states are mixed and the system is in its intermediate phase. All of the three levels are mixed if $2Jz > |E|$. The system is then an antiferromagnet. The theory works then for sufficiently strong magnetic fields that unscramble the levels. Finally if $D < 0$ and $|E| < |D|$ the crystal is antiferromagnetic for $2Jz > |E|$. In particular a system with $E = 0$ is an easy axis antiferromagnet (with two modes of oscillations) for any J. If we switch off the E anisotropy first then the theory yields frequencies which never become soft, even for vanishing H. The theory is then meaningful as long as $D \gtrsim 4Jz$.

Now, if the J^2 terms in the energies are neglected, the harmonic hamiltonian can be brought into the diagonal form like (2.7), with $\varepsilon^{c,C}(k) = \varepsilon_{1,2}(k) + O(J^2)$ and $\varepsilon^{d,D}(k) = \varepsilon_{3,4}(k) + O(J^2)$, under the substitution

$$a_k = \frac{1}{\sqrt{2}}[c_k + C_k - p_-(c^+_{-k} - C^+_{-k})] + O(J^2), \tag{3.7a}$$

$$A_k = \frac{1}{\sqrt{2}}[C_k - c_k - p_+(c^+_{-k} + C^+_{-k})] + O(J^2), \tag{3.7b}$$

$$\tilde{b}_k = \frac{1}{\sqrt{2}}[d_k + D_k + \tfrac{1}{2} J\mathcal{N}^6 E^2\gamma(k)(D^+_{-k} - d^+_{-k})] + O(J^2), \tag{3.7c}$$

$$\tilde{B}_k = \frac{1}{\sqrt{2}}[D_k - d_k + \tfrac{1}{2} J\mathcal{N}^6 E^2\gamma(k)(d^+_{-k} + D^+_{-k})] + O(J^2). \tag{3.7d}$$

The r.f. field applied along the uniaxis will hence couple to

$$\sum_i S_i^z + \sum_j S_j^z = -\tfrac{1}{2} N^{1/2} \mathcal{N}^8 E^3 Jz(d_0^+ + d_0)$$
$$-\mathcal{N}^2 H \sum_k (c_k^+ C_k + C_k^+ c_k + 2d_k^+ D_k + 2D_k^+ d_k) + O(J^2). \tag{3.8}$$

In contradistinction to the ferromagnetic exchange case no parallel pumping will be triggered. In the former case the r.f. field could pump pairs of like magnons provided $E \neq 0$. In addition the coherent resonance with the $k = 0$ d-magnons is here much weaker than for ferromagnetically coupled spins, since the matrix element is proportional to J. The third difference is the occurence of the new incoherent resonances $c_k^+ C_k$ and $d_k^+ D_k$ which should happen when $\omega = 4JE\mathcal{N}^2\gamma(k)$. These incoherent resonances disappear at $H = 0$.

On the other hand the pattern of the possible effects induced by the r.f. field applied along the x-axis is very similar to the one characteristic for systems with weak ferromagnetic exchange. The field will then trigger a coherent resonance with the $k = 0$ C-magnons, two incoherent resonances $c_k^+ d_k$ and $C_k^+ D_k$ at $\omega = (H^2 + E^2)^{1/2} - D + 2Jz\mathcal{N}^4 H^2$, and finally the parallel pumpings of unlike particles $c_k^+ D_{-k}^+$ and $C_{-k}^+ d_k^+$ at $\omega = 3(H^2 + E^2)^{1/2} - D + 6Jz\mathcal{N}^4 H^2$.

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