SPIN WAVES IN SYSTEMS WITH WEAK EXCHANGE FIELDS. II*

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Spin-wave theories are constructed for two kinds of systems with a weak ferromagnetic-like exchange coupling: (1) spin-1 systems with uniaxial and orthorhombic anisotropies, with the magnetic field applied perpendicular to the uniaxis; (2) spin-2 systems with cubic anisotropy. These theories are based on the matrix-elements matching method proposed recently by Cieplak and Keffer. Spin-wave modes and stability conditions are found. The possible effects of an r.f. field are described. For the spin-1 systems with a perpendicular static magnetic field parallel pumping processes are predicted even in the absence of the orthorhombic anisotropy field.

1. *Introduction*

Spin-wave descriptions of the low temperature behaviour of magnetic materials with localized spins have usually been set up for systems in which the exchange field dominates the single-ion Zeeman and anisotropy interactions [1]. Spin-wave modes are, however, also present in materials with large single-ion fields but small exchange fields. These systems are either paramagnetic or ordered ones.

Two new features are encountered in such materials. The first one is that the ground state of the system, i.e., in the first approximation, the ground state of the single-ion part of the hamiltonian, does not have to coincide with the state of maximal alignment. Instead the ground state may be a product of singlet states or a product of mixtures of spin eigenstates. The second new feature is that the excited spin-states are, in general, not equidistant. Even at low temperatures they can be occupied simultaneously, in the statistical ensemble.

Therefore multi-boson spin representations are appropriate to analyze spin-wave excitations in systems with weak exchange fields at low temperatures. A matrix-elements

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matching method of the construction of such representations has been presented in the first paper [2] of the series. The method is a two-step procedure. First one introduces sets of bosons which excite the single-ion levels. Then one constructs the spin operators out of these bosons in a way that reproduces the matrix elements of the operators in the subspace of the single-ion levels.

In this article we shall continue to discuss systems of N spins described by a hamiltonian of the form

$$\mathscr{H} = \sum_i \mathscr{H}_i - 2J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j. \tag{1.1}$$

We assume that the spins are coupled by a weak, ferromagnetic-like, exchange interaction, of strength $J > 0$, between the z nearest neighbours. The crystal is in a single-ion dominated regime if J is much smaller than one of the anisotropy constants or a magnetic field in the single-ion hamiltonian $\mathscr{H}_i$.

In Ref. [2] we have analyzed spin-1 systems with the uniaxial D and orthorhombic E anisotropies and with the magnetic field H applied parallel to the uniaxis (the symbol H incorporates the Landé factor and the Bohr magneton). For $D > 0$, $|E| < D$ and $H \leqslant (D^2 - E^2)^{1/2} - O(J)$ the ground state of the system is a product of singlet states. The system is then a paramagnet. This region of parameters we called region S (for "small magnetic fields"). For H slightly exceeding the above value the system is in the intermediate phase. There the two lowest single-ion eigenstates are mixed by exchange, as the anisotropy field becomes compensated by the magnetic field. The exchange field ceases to be a small perturbation there. In fact it produces a long-range order with a transverse magnetization [3, 4]. The matrix-elements matching method does not work in the intermediate region. For $H \geqslant (D^2 - E^2)^{1/2} + O(J)$ the ground state is a product of states which are linear combinations of states $|+1\rangle_i$ and $|-1\rangle_i$. This is called region L. Now the magnetic field induces a magnetization along the z-axis. The system is also in the region L for any H if $|E| > |D|$ or if $D < 0$. In the latter case the system is ferromagnetic when $2Jz > |E|$, in particular when $E = 0$.

The magnon hamiltonians for the two regions S and L turn out to be of different forms. In the region L linear terms in the Bose operators appear. These terms are eliminated by a unitary transformation of adding to the operators a constant proportional to J.

For both regions the spin-wave modes have been found and the stability conditions with respect to the size of H and J have been obtained. Subsequently, the possible effects of an oscillatory field on the systems have been discussed in Ref. [2]. This field will couple to the component of the total spin in the direction of the r.f. field. In the Heisenberg picture the component of the total spin participates in various oscillatory motions. The field can tune into the eigenmodes of these oscillations. It turns out that in this way the r.f. field can trigger the following kinds of processes: a parallel pumping of pairs of magnons with opposite wave-vectors, a coherent resonance with $k = 0$ magnons, and finally an incoherent resonance between the excited states, in which a spectrum of magnons is involved.

In this paper we set up a similar spin-wave theory for two types of systems with weak exchange fields: (1) a spin-1 system with the uniaxial and orthorhombic anisotropies, with the magnetic field perpendicular to the uniaxis; (2) a spin-2 system with a cubic anisotropy (the lowest spin that can distinguish cubic symmetry is 2). An approximate theory for spin-4 systems (Pr^{3+}) in the absence of a magnetic field was worked out by Grover [5].

For both systems of our concern in this article, we find two different ground state configurations. The corresponding magnon-hamiltonians resemble either that for region S or that for region L. However, in the cubic system four, and not two, sets of bosons have to be introduced.

The analysis of magnon modes and of possible r.f. field-effects in the spin-1 system with the perpendicular static magnetic field is performed in Sec. 2. It turns out that, unlike the parallel static field case, the presence of an orthorhombic field is not necessary for parallel pumpings and incoherent resonances to occur. A similar discussion for the spin-2 system is presented in Sec. 3.

For neither of these systems are we able to find the single-ion contribution to the quartic terms of the spin-wave hamiltonian and hence the magnon relaxation times are not calculated. We expect, however, that the conclusions concerning the threshold amplitude of the r.f. field for parallel pumpings are qualitatively analogous to those for the spin-1 system with the parallel static magnetic field, in the region S, as discussed in Ref. [2]. Systems in the region S are exceptional ones because they can be treated by means of a rigorous spin representation due to Homma et al. [6], and therefore a thorough analysis is available.

Systems with an antiferromagnetic-like exchange coupling have to be discussed in terms of the two sublattice picture which is done in the following paper.

2. Spin waves in the spin-1 system with the magnetic field perpendicular to the uniaxial anisotropy field

Following Ref. [2] consider the spin-1 system with the uniaxial anisotropy D, along the z-axis, and the orthorhombic anisotropy E. If the magnetic field is now applied along the x-axis, the single-ion hamiltonian acquires the following form

$$\mathscr{H}_i = D(S_i^z)^2+\tfrac{1}{2}E[(S_i^+)^2+(S_i^-)^2]-\tfrac{1}{2}H(S_i^+ +S_i^-). \tag{2.1}$$

This hamiltonian has three eigenstates:

$$\mathscr{H}_i|1\rangle_i = \tfrac{1}{2}\{D+E-[(D+E)^2+4H^2]^{1/2}\}\,|1\rangle_i, \tag{2.2a}$$

$$\mathscr{H}_i|2\rangle_i = (D-E)\,|2\rangle_i, \tag{2.2b}$$

$$\mathscr{H}_i|3\rangle_i = \tfrac{1}{2}\{D+E+[(D+E)^2+4H^2]^{1/2}\}\,|3\rangle_i, \tag{2.2c}$$

where

$$|1\rangle_i = \frac{1}{\sqrt{2}}\mathscr{N}_\perp[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}|0\rangle_i$$

$$+\frac{1}{\sqrt{2}}[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}(|+1\rangle_i+|-1\rangle_i), \tag{2.3a}$$

$$|2\rangle_i = \frac{1}{\sqrt{2}}(|+1\rangle_i - |-1\rangle_i), \tag{2.3b}$$

$$|3\rangle_i = \frac{1}{\sqrt{2}}\mathcal{N}_\perp\{-[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}\}\,|0\rangle_i$$

$$+\frac{1}{\sqrt{2}}[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}(|+1\rangle_i+|-1\rangle_i), \tag{2.3c}$$

with

$$\mathcal{N}_\perp = [(D+E)^2+4H^2]^{-1/4}.$$

For fields applied along the y-axis E has to be replaced by $-E$.

None of the single-ion eigenstates has a magnetic moment in the z-direction.

Apart from insignificant changes in the relative positions of the two upper states there are two patterns of the single-ion levels:

(1) The pattern in which the state $|2\rangle_i$ constitutes the ground state and the state $|3\rangle_i$ is the highest one. This happens for $H < H_c^{\perp(0)} \equiv [2E(E-D)]^{1/2}$ if (a) $D \geqslant 0$, $E > 0$, $E > D$, or (b) $D \leqslant 0$, $E > 0$. If the state $\prod_{i=1}^{N} |2\rangle_i$ is stable for small oscillations (spin waves) when the exchange interaction is introduced, we speak of the system as being in its region $S^\perp$.

(2) The pattern in which the state $|1\rangle_i$ is lowest. This happens when (a) $E < 0$, or (b) $D > 0$, $E \geqslant 0$, $E < D$, any H, or (c) $D \geqslant 0$, $E > 0$, $E > D$, $H > H_c^{\perp(0)}$, or (d) $D \leqslant 0$, $E > 0$, $H > H_c^{\perp(0)}$. The domain of parameters H, D, E for which $\prod_{i=1}^{N} |1\rangle_i$ remains stable to small oscillations of interacting spin system will be referred to as region $L^\perp$.

Note that in either of the two regions $S^\perp$ and $L^\perp$ there can be both paramagnetic and ferromagnetic systems.

For H directed along the x-axis, the magnetic field induced transitions between the regions $S^\perp$ and $L^\perp$ can occur only for positive E and either if $D > 0$, $E > D$ or if $D < 0$. Similarly, for H directed along the y-axis, transitions can appear only for negative E and if $D > 0$, $|E| > D$ or if $D < 0$. The presence of a transverse field makes the sign of E crucial for the occurrence of phase transitions.

Now we shall set up a spin-wave theory for the system with the magnetic field along the x-axis by means of the two-bosons matrix-elements matching method. Note that the operator S_i^+ should act on the three states $|1\rangle_i$, $|2\rangle_i$, $|3\rangle_i$ as follows

$$S_i^+|1\rangle_i = \mathcal{N}_\perp^2[2H|1\rangle_i+(D+E)\,|3\rangle_i]$$

$$+\frac{1}{\sqrt{2}}\mathcal{N}_\perp\{D+E+[(D+E)^2+4H^2]^{1/2}\}^{1/2}|2\rangle_i, \tag{2.4a}$$

$$S_i^+|2\rangle = \frac{1}{\sqrt{2}}\mathcal{N}_\perp\{-[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}|1\rangle_i$$

$$+[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}|3\rangle_i\}, \tag{2.4b}$$

$$S_i^+|3\rangle_i = \mathcal{N}_\perp^2[(D+E)\,|1\rangle_i - 2H|3\rangle_i]$$

$$-\frac{1}{\sqrt{2}}\mathcal{N}_\perp\{[(D+E)^2+4H^2]^{1/2}-(D+E)\}^{1/2}|2\rangle_i. \tag{2.4c}$$

Similar equations can be written for $S_i^-, S_i^z, (S_i^z)^2, (S_i^+)^2$, and $(S_i^-)^2$. The bosonic representation should reproduce the above matrix elements in any of the two regions $S^\perp$ and $L^\perp$.

A. Region $S^\perp$

Since the $|2\rangle_i$ state is the lowest one in this region let

$$|2\rangle_i = |0,0\rangle_i,$$

$$|1\rangle_i = a_i^+|0,0\rangle_i,$$

$$|3\rangle_i = b_i^+|0,0\rangle_i, \tag{2.5}$$

where $|0,0\rangle_i$ is the joint vacuum state for the two bosons a_i and b_i. The single-ion hamiltonian becomes then

$$\mathcal{H}_i = D-E+\tfrac{1}{2}\{3E-D-[(D+E)^2+4H^2]^{1/2}\}a_i^+a_i$$

$$+\tfrac{1}{2}\{3E-D+[(D+E)^2+4H^2]^{1/2}\}b_i^+b_i+\ \dots . \tag{2.6}$$

Now, as in Ref. [2], we look for combinations of operators a_i and b_i which imitate the behaviour of S_i^z, S_i^+, S_i^-, $(S_i^z)^2, (S_i^+)^2$, and $(S_i^-)^2$ on the three states. Again the form of the quartic terms can not be established. For the spin operators we obtain

$$S_i^z = \frac{1}{\sqrt{2}}\mathcal{N}_\perp\{[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}$$

$$\times(b_i^+ + b_i - a_i^+a_ib_i - b_i^+a_i^+a_i - b_i^+b_i^+b_i - b_i^+b_ib_i)$$

$$+[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}$$

$$\times(a_i^+ + a_i - a_i^+b_i^+b_i - b_i^+a_ib_i - a_i^+a_i^+a_i - a_i^+a_ia_i)\}+\ \dots, \tag{2.7a}$$

$$S_i^+ = \frac{1}{\sqrt{2}}\mathcal{N}_\perp\{[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}$$

$$\times(b_i^+ - b_i + a_i^+a_ib_i - b_i^+a_i^+a_i - b_i^+b_i^+b_i + b_i^+b_ib_i)$$

$$+[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}$$

$$\times(a_i - a_i^+ + a_i^+b_i^+b_i - b_i^+a_ib_i + a_i^+a_i^+a_i - a_i^+a_ia_i)\}$$

$$+\mathcal{N}_\perp^2[(D+E)(a_i^+b_i+b_i^+a_i)+2H(a_i^+a_i-b_i^+b_i)]+\ \dots, \tag{2.7b}$$

$$S_i^- = (S_i^+)^\dagger. \tag{2.7c}$$

The above representation allows us to find the exchange part of the magnon hamiltonian. The third order terms in the single-ion part, $\mathscr{H}_i$, cancel out. In terms of the Fourier-transformed operators the total hamiltonian acquires then the following form

$$\begin{aligned}\mathscr{H} &= N(D-E)\\ &+\sum_k a_k^+ a_k\{\tfrac{1}{2}[3E-D-((D+E)^2+4H^2)^{1/2}]-2J\gamma(k)\}\\ &+\sum_k b_k^+ b_k\{\tfrac{1}{2}[3E-D+((D+E)^2+4H^2)^{1/2}]-2J\gamma(k)\}\\ &+\mathscr{N}_\perp^2 J\sum_k \gamma(k)\,[(D+E)\,(a_k^+ a_{-k}^+ + a_k a_{-k} - b_k^+ b_{-k}^+ - b_k b_{-k})\\ &\quad -4H(b_k^+ a_{-k}^+ + a_k b_{-k})]+\dots \qquad (2.8)\end{aligned}$$

where

$$\gamma(k) = \sum_{\delta}{}_{\text{n.n.}}\, e^{ik\delta}.$$

The quadratic part of this hamiltonian resembles the harmonic hamiltonian for the region S. The energies of the two branches of excitations are

$$\begin{aligned}\varepsilon^{\pm}(k) &= \tfrac{1}{2}\{(D+E)^2+4H^2+(3E-D)\,[3E-D-8J\gamma(k)]\\ &\pm 2[(3E-D)\,((D+E)^2+4H^2)\,(3E-D-8J\gamma(k))]^{1/2}\}^{1/2}\\ &= \tfrac{1}{2}\{3E-D\pm[(D+E)^2+4H^2]^{1/2}\}-2J\gamma(k)+O(J^2). \qquad (2.9)\end{aligned}$$

The $k = 0$ ε^--magnons go soft at the critical field $H_{c1}^\perp$:

$$H_{c1}^\perp = H_c^{\perp(0)}-Jz(3E-D)\,(H_c^{\perp(0)})^{-1}+O(J^2). \qquad (2.10)$$

This marks the upper boundary for the region $S^\perp$. Slightly above this field the systems are in the intermediate regions as the $|1\rangle_i$ and $|2\rangle_i$ states are mixed by exchange interactions. For fields bigger than $H_c^{\perp(0)}$ the configuration of the region $L^\perp$ is reached.

Consider now the possible effects of an oscillatory magnetic field. Note that the harmonic hamiltonian for the region $S^\perp$ becomes diagonalized if we substitute

$$a_k = c_k-2J\gamma(k)\,(D+E)\mathscr{N}_\perp^2 p_- c_{-k}^+ + 4J\gamma(k)H\mathscr{N}_\perp^2(3E-D)^{-1}d_{-k}^+ + O(J^2), \qquad (2.11a)$$

$$b_k = d_k+2J\gamma(k)\,(D+E)\mathscr{N}_\perp^2 p_+ d_{-k}^+ + 4J\gamma(k)H\mathscr{N}_\perp^2(3E-D)^{-1}c_{-k}^+ + O(J^2), \qquad (2.11b)$$

where

$$p_\pm = \{3E-D\pm[(D+E)^2+4H^2]^{1/2}\}^{-1}.$$

Within terms proportional to J^2 we identify ε^+ and ε^- with the energies ε^d and ε^c of the d and c-magnons respectively. The r.f. field applied along the z-axis will couple to

$$\begin{aligned}\sum_i S_i^z &= (\tfrac{1}{2}N)^{1/2}\{v_\mp^\perp[1+2Jz(D+E)\mathscr{N}_\perp^2 p_+]+v_\pm^\perp 4JzH\mathscr{N}_\perp^2(3E-D)^{-1}\}\,(d_0^+ + d_0)\\ &+(\tfrac{1}{2}N)^{1/2}\{v_\pm^\perp[1-2Jz(D+E)\mathscr{N}_\perp^2 p_-]+v_\mp^\perp 4JzH\mathscr{N}_\perp^2(3E-D)^{-1}\}\,(c_0^+ + c_0), \qquad (2.12)\end{aligned}$$

whereas if it is applied along the x-axis it will couple to

$$\sum_i S_i^x = \mathcal{N}_\perp^2 \sum_k \{(D+E)(d_k^+ c_k + c_k^+ d_k) + 2H(c_k^+ c_k - d_k^+ d_k)$$
$$- J\gamma(k)(D+E)[(D+E)(2E^2 - 2ED - H^2)^{-1}(c_k^+ d_{-k}^+ + c_k d_{-k})$$
$$-4H(3E-D)^{-1} p_+ (d_k^+ d_{-k}^+ + d_k d_{-k}) + 4H(3E-D)^{-1} p_- (c_k^+ c_{-k}^+ + c_k c_{-k})]\} \qquad (2.13)$$

where

$$v_\pm^\perp = [1 \pm (D+E)\mathcal{N}_\perp]^{1/2}.$$

The formula for $\sum_i S_i^y$ repeats the one for $\sum_i S_i^z$ in which $(d_0^+ + d_0)$ is replaced by $i(c_0^+ - c_0)$, and $(c_0^+ + c_0)$ by $i(d_0^+ - d_0)$. Thus if the r.f. field is applied along an axis which is perpendicular to the static field (the x-axis), coherent resonances with the c_0 and d_0 magnons appear. For $H = 0$, the r.f. field in the z-direction produces only d_0-magnons, while if it is in the x-direction — only c_0-magnons. The incoherent resonance at frequency $|\varepsilon^d(k) - \varepsilon^c(k)|$

TABLE I

Possible effects of an oscillatory field on the spin-1 system

Region	r.f. field along	Form of the r.f. perturbation	Process	Qualifications
$S\perp$ (static field along x)	z	d_0^+	coherent perpendicular resonance	if $D+E < 0$ then $H \neq 0$, otherwise none,
		c_0^+	coherent perpendicular resonance	if $D+E > 0$ then $H \neq 0$, otherwise none,
	x	$d_k^+ c_k$	incoherent resonance	$D \neq -E$,
		$c_k^+ d_{-k}^+$	parallel pumping of unlike magnons	$J \neq 0$, $D \neq -E$,
		$c_k^+ c_{-k}^+$, $d_k^+ d_{-k}^+$	parallel pumping of like magnons	$J \neq 0$, $H \neq 0$, $D \neq -E$.
$L\perp$ (static field along x)	z	c_0^+	coherent perpendicular resonance	if $D+E > 0$ then $H \neq 0$, otherwise none,
		$d_k^+ c_k$	incoherent resonance	if $D+E < 0$ then $H \neq 0$, otherwise none,
		$d_{-k}^+ c_k^+$	"parallel" pumping of unlike magnons	$J \neq 0$, $D \neq -E$, if $D+E < 0$ then $H \neq 0$,
	x	d_0^+	coherent parallel resonance	$D \neq -E$,
		$c_{-k}^+ c_k^+$, $d_{-k}^+ d_k^+$	parallel pumping of like magnons	$J \neq 0$, $H \neq 0$, $D \neq -E$.

is possible for the r.f. field parallel to the static one. Also then, but at frequencies $\varepsilon^c(k) + \varepsilon^d(-k)$, $\varepsilon^c(k) + \varepsilon^c(-k)$, and $\varepsilon^d(k) + \varepsilon^d(-k)$ respectively, the three pumping processes are seen to emerge. Unlike the situation for the region S these processes do not vanish for purely uniaxial systems, i.e. for $E = 0$. Hence the pumping here is allowed as a result of an interplay of the Zeeman and exchange fields. The possible effects of an oscillatory field are summarized in Table I.

B. Region $L^{\perp}$

In this region we put

$$|1\rangle_i = |0, 0\rangle_i,$$

$$|2\rangle_i = a_i^+|0, 0\rangle_i,$$

$$|3\rangle_i = b_i^+|0, 0\rangle_i, \tag{2.14}$$

so the single-ion hamiltonian becomes

$$\mathscr{H}_i = \tfrac{1}{2}(D+E)-\tfrac{1}{2}[(D+E)^2+4H^2]^{1/2}$$
$$+\tfrac{1}{2}\{D-3E+[(D+E)^2+4H^2]^{1/2}\}a_i^+a_i+[(D+E)^2+4H^2]^{1/2}b_i^+b_i+\ \dots\ . \tag{2.15}$$

Now the spin operators are represented by

$$S_i^z = \frac{1}{\sqrt{2}}\mathscr{N}_\perp\{[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}$$
$$\times(a_i^++a_i-a_i^+b_i^+b_i-b_i^+a_ib_i-a_i^+a_i^+a_i-a_i^+a_ia_i)$$
$$+[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}(a_i^+b_i+b_i^+a_i)+\ \dots, \tag{2.16a}$$

$$S_i^+ = 2H\mathscr{N}_\perp^2(1-2b_i^+b_i-a_i^+a_i)$$
$$+(D+E)\mathscr{N}_\perp^2\{b_i^++b_i-b_i^+a_i^+a_i-a_i^+a_ib_i-b_i^+b_i^+b_i-b_i^+b_ib_i\}$$
$$+\frac{1}{\sqrt{2}}\mathscr{N}_\perp\{[((D+E)^2+4H^2)^{1/2}+D+E]^{1/2}(a_i^+-a_i+a_i^+a_ia_i-a_i^+a_i^+a_i)$$
$$+[((D+E)^2+4H^2)^{1/2}-(D+E)]^{1/2}(b_i^+a_i-a_i^+b_i)\}+\ \dots, \tag{2.16b}$$

$$S_i^- = (S_i^+)^\dagger. \tag{2.16c}$$

The total hamiltonian then acquires the form

$$\mathscr{H} = N\{\tfrac{1}{2}(D+E)-\tfrac{1}{2}[(D+E)^2+4H^2]^{1/2}-4zJH^2\mathscr{N}_\perp^4\}$$
$$-N^{1/2}4Jz(D+E)H\mathscr{N}_\perp^4(b_0^++b_0)$$
$$+\sum_k[\mathscr{A}_\perp(k)a_k^+a_k+J(D+E)\mathscr{N}_\perp^2\gamma(k)(a_k^+a_{-k}^++a_ka_{-k})]$$
$$+\sum_k[\mathscr{B}_\perp(k)b_k^+b_k-J(D+E)^2\mathscr{N}_\perp^4\gamma(k)(b_k^+b_{-k}^++b_kb_{-k})]$$
$$+4JH\mathscr{N}_\perp^2N^{-1/2}\sum_{k_1,k_2}\{-\gamma(k_1)(b_{k_1+k_2}^+a_{k_2}a_{k_1}+a_{k_1}^+a_{k_2}^+b_{k_1+k_2})$$
$$+\mathscr{N}_\perp^2(D+E)[z+\gamma(k_1)](b_{k_1}^+a_{k_2}^+a_{k_1+k_2}+a_{k_1+k_2}^+a_{k_2}b_{k_1})$$
$$+\mathscr{N}_\perp^2(D+E)[z+2\gamma(k_1)](b_{k_1}^+b_{k_2}^+b_{k_1+k_2}+b_{k_1+k_2}^+b_{k_2}b_{k_1})\}+\ \dots, \tag{2.17}$$

which reminds one of the structure of the hamiltonian for the region L [2]. In Eq. (2.17)

$$\mathscr{A}_{\perp}(k) = \tfrac{1}{2}\{D-3E+[(D+E)^2+4H^2]^{1/2}\}+8JzH^2\mathscr{N}_{\perp}^4-2J\gamma(k),$$

$$\mathscr{B}_{\perp}(k) = [(D+E)^2+4H^2]^{1/2}-2J\mathscr{N}_{\perp}^4[(D+E)^2\gamma(k)-8H^2z].$$

The linear term $(b_0^+ + b_0)$ can be eliminated by means of the unitary transformation

$$b_k = \beta_k+\delta_{k,0}N^{1/2}4Jz(D+E)H\mathscr{N}_{\perp}^6+O(J^2). \tag{2.18}$$

This transformation brings in some unknown, proportional to J, cubic terms and also new linear and quadratic terms, which are proportional to J^n, with $n \geqslant 2$. Therefore in the harmonic approximation

$$\begin{aligned}\mathscr{H} &= N\{\tfrac{1}{2}(D+E)-\tfrac{1}{2}[(D+E)^2+4H^2]^{1/2}-4JzH^2\mathscr{N}_{\perp}^4\}\\ &+\sum_k[\mathscr{A}_{\perp}(k)a_k^+a_k+J(D+E)\mathscr{N}_{\perp}^2\gamma(k)(a_k^+a_{-k}^++a_ka_{-k})]\\ &+\sum_k[\mathscr{B}_{\perp}(k)\beta_k^+\beta_k-J(D+E)^2\mathscr{N}_{\perp}^4\gamma(k)(\beta_k^+\beta_{-k}^++\beta_k\beta_{-k})]+O(J^2)+\ldots,\end{aligned} \tag{2.19}$$

and the energies of the spin-wave excitations are

$$\begin{aligned}\varepsilon^a(k) &\simeq [\mathscr{A}_{\perp}^2(k)-4J^2(D+E)^2\mathscr{N}_{\perp}^4\gamma^2(k)]^{1/2}\\ &= \tfrac{1}{2}|D-3E+[(D+E)^2+4H^2]^{1/2}|+2J[4\mathscr{N}_{\perp}^4H^2z-\gamma(k)]+O(J^2),\end{aligned} \tag{2.20}$$

$$\begin{aligned}\varepsilon^{\beta}(k) &\simeq \{(D+E)^2+4H^2-4J\mathscr{N}_{\perp}^2[(D+E)^2\gamma(k)-8H^2z]\\ &\quad+64J^2\mathscr{N}_{\perp}^8H^2z[4H^2z-(D+E)^2\gamma(k)]\}^{1/2}\\ &= [(D+E)^2+4H^2]^{1/2}-2J\mathscr{N}_{\perp}^4[(D+E)^2\gamma(k)-8H^2z]+O(J^2).\end{aligned} \tag{2.21}$$

If $D \geqslant 0$, $E > 0$, $E > D$ or if $D \leqslant 0$, $E > 0$, $\varepsilon^a(0)$ vanishes at

$$H_{c2}^{\perp} = H_c^{\perp(0)}+Jz(3E-D)(D+E)^2[H_c^{\perp(0)}(D^2+9E^2-6ED)]^{-1}+O(J^2). \tag{2.22}$$

Thus the width of the intermediate region, $H_{c2}^{\perp}-H_{c1}^{\perp}$, is again proportional to J.

Note that for very large magnetic fields one should obtain a more straightforward description of the system in the region $L^{\perp}$ by choosing the quantization axis parallel to the applied field.

Finally, for all possible combinations of E and D values, the energies (2.9), (2.20), and (2.21) coincide, in the limit of $H = 0$, with the appropriate energies calculated for the regions S and L [2], except that the term proportional to J^2 in one of the energies (ε^a), characteristic for the region L, is not reproduced.

Now, the harmonic hamiltonian for the region $L^{\perp}$ becomes diagonal under the transformation

$$\begin{aligned}a_k &= c_k-2J\gamma(k)\mathscr{N}_{\perp}^2(D+E)\{D-3E+[(D+E)^2+4H^2]^{1/2}\}^{-1}c_{-k}^++O(J^2),\\ \beta_k &= d_k+J\gamma(k)(D+E)^2\mathscr{N}_{\perp}^6d_{-k}^++O(J^2).\end{aligned} \tag{2.23}$$

It turns out that the set of possible r.f. field-induced phenomena, as summarized in Table I, is similar to that for the region L, except the pumpings by the r.f. field parallel to the static field do not vanish in the limit of $E = 0$.

3. Spin waves in systems with cubic anisotropy

Our method of spin operators construction can also be applied to weak-exchange -systems with other anisotropy fields. Here we shall discuss the case of purely cubic anisotropy, when the single-ion hamiltonian reads

$$\begin{aligned}\mathscr{H}_i &= A[(S_i^x)^4+(S_i^y)^4+(S_i^z)^4]-HS_i^z \\ &= \tfrac{1}{8}A[(S_i^+)^4+(S_i^-)^4+(S_i^+)^2(S_i^-)^2+(S_i^-)^2(S_i^+)^2 \\ &\quad +(S_i^+S_i^-+S_i^-S_i^+)^2]+A(S_i^z)^4-HS_i^z. \end{aligned} \tag{3.1}$$

if this is a spin 2 system, the single-ion hamiltonian has the five following eigenstates:

$$\begin{aligned}\mathscr{H}_i|+\rangle_i &= [21A+(9A^2+4H^2)^{1/2}]\,|+\rangle_i, \\ \mathscr{H}_i|0\rangle_i &= 24A|0\rangle, \\ \mathscr{H}_i|-1\rangle_i &= (18A+H)\,|-1\rangle_i, \\ \mathscr{H}_i|-\rangle_i &= [21A-(9A^2+4H^2)^{1/2}]\,|-\rangle_i, \\ \mathscr{H}_i|+1\rangle_i &= (18A-H)\,|+1\rangle_i, \end{aligned} \tag{3.2}$$

where

$$|+\rangle_i = \frac{1}{\sqrt{2}}\mathscr{N}'\{[(9A^2+4H^2)^{1/2}+2H]^{1/2}|-2\rangle_i+\eta'[(9A^2+4H^2)^{1/2}-2H]^{1/2}|+2\rangle_i\},$$

$$|-\rangle_i = \frac{1}{\sqrt{2}}\mathscr{N}'\{[(9A^2+4H^2)^{1/2}+2H]^{1/2}|+2\rangle_i-\eta'[(9A^2+4H^2)^{1/2}-2H]^{1/2}|-2\rangle_i\}, \tag{3.3}$$

and

$$\mathscr{N}' = (9A^2+4H^2)^{-1/4}.$$

$$\eta' = \begin{cases}+1 & A \geqslant 0 \\ -1 & A < 0.\end{cases}$$

There are two possible structures of the single-ion ground state:
(1) For positive A and for $0 < H < H_c'^{(0)} = 2A$, the lowest energy state is $|+1\rangle_i$ and the next states are in the sequence $|-\rangle_i$, $|-1\rangle_i$, $|0\rangle_i$, $|+1\rangle_i$. If the product of states $|+1\rangle_i$ is an approximate ground state of the system, we shall speak of region S'.
(2) For positive A and $H > H_c'^{(0)}$ and for negative A, $H > 0$, $|-\rangle_i$ is the ground state and other states can switch their relative positions under the influence of the Zeeman field. When the ground state of the system is composed of the states $|-\rangle_i$, we refer to region L'. Again, if A is positive, regions S' and L' are separated by an intermediate region of width proportional to J.

A. Region S'

Consider first region S'. Let

$$|+1\rangle_i = |0,0,0,0\rangle_i,$$
$$|-\rangle_i = a_i^+|0,0,0,0\rangle_i,$$
$$|-1\rangle_i = b_i^+|0,0,0,0\rangle_i,$$
$$|0\rangle_i = c_i^+|0,0,0,0\rangle_i,$$
$$|+\rangle_i = d_i^+|0,0,0,0\rangle_i, \tag{3.4}$$

where a_i, b_i, c_i, d_i are four sets of Bose operators and $|0,0,0,0\rangle_i$ is the joint vacuum state. The matrix-elements matching method yields

$$S_i^z = 1+[(\mathcal{N}')^2 4H-1]a_i^+a_i-2b_i^+b_i-c_i^+c_i$$
$$-[(\mathcal{N}')^2 4H+1]d_i^+d_i+6A(\mathcal{N}')^2(a_i^+d_i+d_i^+a_i), \tag{3.5a}$$
$$S_i^+ = \sqrt{2}\,\mathcal{N}'[(9A^2+4H^2)^{1/2}+2H]^{1/2}$$
$$\times(a_i^+ +b_i^+d_i-a_i^+a_i^+a_i-a_i^+b_i^+b_i-a_i^+c_i^+c_i-a_i^+d_i^+d_i)$$
$$+\sqrt{2}\,\eta'\mathcal{N}'[(9A^2+4H^2)^{1/2}-2H]^{1/2}$$
$$\times(d_i^+ -b_i^+a_i-d_i^+a_i^+a_i-d_i^+b_i^+b_i-d_i^+c_i^+c_i-d_i^+d_i^+d_i)$$
$$+\sqrt{6}\,(c_i+c_i^+b_i-a_i^+a_ic_i-b_i^+b_ic_i-c_i^+c_ic_i-d_i^+d_ic_i)+\ldots, \tag{3.5c}$$
$$S_i^- = (S_i^+)^\dagger. \tag{3.5c}$$

With the use of Eqs. (3.5) we arrive at the following hamiltonian for the system

$$\mathcal{H} = N(18A-H-2Jz)$$
$$+\sum_k(\varepsilon_k^a a_k^+a_k+\varepsilon_k^b b_k^+b_k+\varepsilon_k^c c_k^+c_k+\varepsilon_k^d d_k^+d_k)$$
$$-2J\sum_k\{\sqrt{3}\,\mathcal{N}'[(9A^2+4H^2)^{1/2}+2H]^{1/2}\gamma(k)\,(a_k^+c_{-k}^+ +a_kc_{-k})$$
$$+\sqrt{3}\,\mathcal{N}'\eta'[(9A^2+4H^2)^{1/2}-2H]^{1/2}\gamma(k)\,(d_k^+c_{-k}^+ +d_kc_{-k})$$
$$+(\mathcal{N}')^2 6Az(a_k^+d_k+d_k^+a_k)\}+\ldots, \tag{3.6}$$

where

$$\varepsilon_k^a = 3A+H-(9A^2+4H^2)^{1/2}-2J\{z[(\mathcal{N}')^2 4H-1]+(\mathcal{N}')^2[(9A^2+4H^2)^{1/2}+2H]\gamma(k)\},$$
$$\varepsilon_k^b = 2H+4Jz,$$
$$\varepsilon_k^c = 6A+H-2J[3\gamma(k)-z],$$
$$\varepsilon_k^d = 3A+H+(9A^2+4H^2)^{1/2}-2J\{-z[(\mathcal{N}')^2 4H+1]+(\mathcal{N}')^2[(9A^2+4H^2)^{1/2}-2H]\gamma(k)\}. \tag{3.7}$$

The energies ε_k^a, ε_k^c, ε_k^d are the magnon energies, if terms quadratic with respect to J are neglected. The energy ε_k^b is exact. The energy ε_0^a vanishes at

$$H'_{c1} = 2A - 8Jz + O(J^2). \tag{3.8}$$

Beyond this field the system is no longer in the region S'.

Note that at $H = 0$ (with $A > 0$) the a-magnons are soft regardless of the magnitude of J, and then our theory does not work. This reflects the degeneracy of the single-ion ground state: it is a mixture of the states $|-1\rangle_i$, $|+1\rangle_i$, and $|-\rangle_i$. Therefore, the system is then in an intermediate phase. Since the degenerate single-ion ground state becomes magnetic under the influence of exchange interactions, this intermediate phase is ferromagnetically ordered. For $s > 2$, however, the matrix-elements matching theory will apply even in this limit. For example, in the $s = 4$ cubic system analyzed by Grover [5], the ground state is a singlet at $H = 0$, and for small enough J's such systems are paramagnetic.

The hamiltonian (3.6) becomes diagonal under the transformation

$$\begin{aligned} a_k &= e_k + J\gamma(k) m_1 f_{-k} + 6Jz(\mathcal{N}')^2 A(3A+H)^{-1} g_k + O(J^2), \\ b_k &= b_k, \\ c_k &= f_k + J\gamma(k) m_1 e^{+}_{-k} + O(J^2), \\ d_k &= g_k + J\gamma(k) m_2 f^{+}_{-k} + 6Jz(\mathcal{N}')^2 A(3A+H)^{-1} e_k + O(J^2), \end{aligned} \tag{3.9}$$

with

$$m_1 = 2\sqrt{3}\,\mathcal{N}'[(9A^2+4H^2)^{1/2}+2H]^{1/2}[9A+2H-(9A^2+4H^2)^{1/2}]^{-1},$$

$$m_2 = 2\sqrt{3}\,\mathcal{N}'[(9A^2+4H^2)^{1/2}-2H]^{1/2}[3A+H+(9A^2+4H^2)^{1/2}]^{-1}.$$

Operators e_k, f_k, g_k satisfy boson commutation relations with an error which is proportional to $(J/A)^2$. Equations (3.9) allow us to find that $\sum_i S_i^z$ contains the following terms:

(1) $e_k^+ e_k$, $b_k^+ b_k$, $f_k^+ f_k$, $g_k^+ g_k$,

(2) $(e_k^+ g_k + g_k^+ e_k)$,

(3) $(f^+_{-k} e_k^+ + e_k f_{-k})$,

(4) $(g_k^+ f^+_{-k} + g_k f_{-k})$.

The last two terms are proportional to J. Term (2) corresponds to an incoherent resonance [7] which occurs at the frequency

$$\varepsilon_k^d - \varepsilon_k^a = 2(9A^2+4H^2)^{1/2} + 8JH(\mathcal{N}')^2[2z+\gamma(k)] + O(J^2). \tag{3.10}$$

Terms (3) and (4) correspond to pumping. In both these processes f-magnons are pumped. None of the effects contributes to the production of b's.

On the other hand $\sum_i S_i^x$ involves

$$
\begin{aligned}
&(5)\ (e_0^+ + e_0), (f_0^+ + f_0), (g_0^+ + g_0),\\
&(6)\ (b_k^+ f_{-k}^+ + b_k f_{-k}),\\
&(7)\ (b_k^+ e_{-k}^+ + b_k e_{-k}),\\
&(8)\ (f_k^+ b_k + b_k^+ f_k),\\
&(9)\ (b_k^+ g_k + g_k^+ b_k),\\
&(10)\ (b_k^+ e_k + e_k^+ b_k).
\end{aligned}
$$

Terms $(f_0^+ + f_0)$, (6), and (7) are proportional to J. Now b, f, and e-magnons can be pumped, if appropriate conditions for frequencies are met. There are also two strong coherent resonances at frequency $\omega = \varepsilon_0^a$ and at $\omega = \varepsilon_0^d$, and a weak one at $\omega = \varepsilon_0^c$. Finally three incoherent resonances are possible, namely at frequencies $\omega = \varepsilon_k^c - \varepsilon_k^b$, $\varepsilon_k^d - \varepsilon_k^b$, $\varepsilon_k^b - \varepsilon_k^a$. All of these incoherent resonances involve b-magnons. We shall not discuss the above mentioned processes in greater detail, as we cannot establish the form of the quartic terms in the hamiltonian.

B. Region L'

In this region we put

$$
\begin{aligned}
|-\rangle_i &= |0, 0, 0, 0\rangle_i,\\
|+1\rangle_i &= a_i^+ |0, 0, 0, 0\rangle_i,\\
|-1\rangle_i &= b_i^+ |0, 0, 0, 0\rangle_i,\\
|0\rangle_i &= c_i^+ |0, 0, 0, 0\rangle_i,\\
|+\rangle_i &= d_i^+ |0, 0, 0, 0\rangle_i,
\end{aligned}
\tag{3.11}
$$

and find

$$
\begin{aligned}
S_i^z = {} & 4H(\mathcal{N}')^2(1 - c_i^+ c_i - 2d_i^+ d_i) + [1 - 4H(\mathcal{N}')^2] a_i^+ a_i\\
& - [1 + 4H(\mathcal{N}')^2] b_i^+ b_i + 6A(\mathcal{N}')^2 (d_i^+ + d_i - d_i^+ a_i^+ a_i\\
& - a_i^+ a_i d_i - d_i^+ b_i^+ b_i - b_i^+ b_i d_i - d_i^+ c_i^+ c_i - c_i^+ c_i d_i) + \ldots,
\end{aligned}
\tag{3.12a}
$$

$$
\begin{aligned}
S_i^+ = {} & \sqrt{2}\,\mathcal{N}'[(9A^2 + 4H^2)^{1/2} + 2H]^{1/2}\\
& \times (a_i + b_i^+ d_i - a_i^+ a_i a_i - b_i^+ b_i a_i - c_i^+ c_i a_i - d_i^+ d_i a_i)\\
& + \sqrt{2}\,\eta' \mathcal{N}'[(9A^2 + 4H^2)^{1/2} - 2H]^{1/2}\\
& \times (-b_i^+ + d_i^+ a_i + b_i^+ a_i^+ a_i + b_i^+ b_i^+ b_i + b_i^+ c_i^+ c_i + b_i^+ d_i^+ d_i)\\
& + 6^{1/2}(a_i^+ c_i + c_i^+ b_i) + \ldots,
\end{aligned}
\tag{3.12b}
$$

$$
S_i^- = (S_i^+)^\dagger. \tag{3.12c}
$$

In the harmonic approximation the hamiltonian becomes

$$\begin{aligned}\mathscr{H} &= N[21A-(9A^2+4H^2)^{1/2}-16JzH^2(\mathscr{N}')^4] \\ &\quad -48JAHz(\mathscr{N}')^4N^{1/2}(d_0^+ + d_0) \\ &\quad +\sum_k (\varepsilon_k'^a a_k^+ a_k + \varepsilon_k'^b b_k^+ b_k + \varepsilon_k'^c c_k^+ c_k + \varepsilon_k'^d d_k^+ d_k) \\ &\quad +6AJ(\mathscr{N}')^2 \sum_k \gamma(k)\,(a_k^+ b_{-k}^+ + a_k b_{-k}) + \dots, \end{aligned} \tag{3.13}$$

where

$$\begin{aligned}\varepsilon_k'^{a,b} &= (9A^2+4H^2)^{1/2} \mp H - 3A \\ &\quad -2J(\mathscr{N}')^2\{\gamma(k)\,[(9A^2+4H^2)^{1/2} \pm 2H] \pm 4Hz[1\mp 4H(\mathscr{N}')^2]\},\end{aligned} \tag{3.14a}$$

$$\varepsilon_k'^c = (9A^2+4H^2)^{1/2}+3A+32JH^2(\mathscr{N}')^4 z, \tag{3.14b}$$

$$\varepsilon_k'^d = 2(9A^2+4H^2)^{1/2}+64JH^2(\mathscr{N}')^4 z. \tag{3.14c}$$

The linear term in (3.13) is eliminated by applying the transformation:

$$d_k = d_k' + \delta_{k,0} JN^{1/2} 24AHz(\mathscr{N}')^6 + O(J^2). \tag{3.15}$$

The energies (3.14) correspond to the four branches of spin-wave excitations, except that $\varepsilon_k'^a$ and $\varepsilon_k'^b$ do not include corrections proportional to J^2.

When A is positive the energy $\varepsilon_k'^a$ vanishes at

$$H_{c2}' = 2A + \tfrac{14}{5}\, Jz \tag{3.16}$$

and this determines the lower boundary of the region L'.

With negative A the $k = 0$ c-magnons become soft when H vanishes. The theory does not apply to the $H = 0$ situation since then the single-ion ground state is degenerate: the states $|0\rangle_i$ and $|-\rangle_i$ correspond to the same energy eigenvalue of $-24|A|$. At $H = 0$ such systems are in an intermediate phase with no magnetic moment along the cubic edges. Again, higher spin values may yield either a paramagnetic or ferromagnetic configuration, depending on the strength of J.

The hamiltonian (3.13) is already diagonal with respect to c's and d's. It is completely diagonalized by the transformation

$$\begin{aligned} a_k &= a_k' - 3J\gamma(k)A(\mathscr{N}')^2[(9A^2+4H^2)^{1/2}-3A]^{-1} b_{-k}'^+, \\ b_k &= b_k' - 3J\gamma(k)A(\mathscr{N}')^2[(9A^2+4H^2)^{1/2}-3A]^{-1} a_{-k}'^+. \end{aligned} \tag{3.17}$$

On the basis of Eqs. (3.17) and (3.12) we conclude that an r.f. field in the z-direction can produce pumping of pairs $a_k'^+ b_k'^+$ and a coherent resonance at $\omega = \varepsilon_0'^d$. An r.f. field applied in the x-direction should give coherent resonances at $\omega = \varepsilon_0'^a$ and $\omega = \varepsilon_0'^b$, incoherent resonances at $\omega = \varepsilon_k'^d - \varepsilon_k'^b$ and $\omega = \varepsilon_k'^d - \varepsilon_k'^a$, and finally it should produce pumping of pairs $d_{-k}'^+ b_k'^+$ and $d_{-k}'^+ a_k'^+$. The rate of these pumpings should be enhanced by the presence of the corresponding virtual processes [2].

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[7] This is still an incoherent resonance because it involves a mixture of wave-vectors. Even if the spectral width of the r.f. field is a small fraction of *J*, modes from some finite domain of wave-vectors would be excited.