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Spin waves in systems with weak exchange fields*

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Multiboson spin waves are constructed to represent the low-temperature collective states of paramagnets and ferromagnets 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 single-ion spin operators are constructed by a matrix-elements-matching method. The method allows the determination of magnon frequencies and of possible effects of applied oscillatory magnetic fields, and also allows the estimation of boundaries between different magnetic phases. However, it does not establish the form of the single-ion contribution to four-magnon interactions. In addition, it is not applicable to transitional phases where the exchange cannot be treated as a perturbation. The spin-1 system with positive and negative uniaxial and orthorhombic anisotropies in various magnitudes of parallel static magnetic fields is discussed in detail. Results of Ishikawa and Oguchi are obtained and generalized. The presence of orthorhombic anisotropy is predicted to make possible the "parallel" pumping of various magnon pairs, and also to give rise to parallel incoherent resonance absorption between excited states, as well as the usual "perpendicular" coherent $k = 0$ ground-state resonance excitation. Magnon relaxation times are estimated in the case of a paramagnet with hard-axis anisotropy in sufficiently small magnetic fields. Typical materials to which this theory applies are hydrated nickel salts.

I. INTRODUCTION

Most of the existent low-temperature theories of magnetic materials deal either with strongly exchange-coupled spin systems or with paramagnets in which there is no interaction between spins. In the first case the crystal field created by each spin's surrounding atoms, which leads to an effective anisotropy field contribution to the Hamiltonian, is treated as a perturbation to the exchange field. In the second case the crystal field is usually considered so extremely dominant that any exchange coupling is completely neglected.

In real paramagnets, even those of very-large single-ion crystal fields, some interactions between neighboring spins will always be present and will allow for the appearance of collective excitations, i.e., spin waves. The aim of this paper is to supply a method of spin-wave analysis of single-ion-anisotropy dominated systems, both ordered and paramagnetic. Such analysis is of importance since, when compared with experimental resonance data, it enables one to determine the crystal-field and exchange parameters. The main difficulty in constructing this kind of theory is in finding a way to transform a spin Hamiltonian into a magnon one.

We shall discuss systems of N spins described by a Hamiltonian of the form

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

We assume that the spins are coupled by a weak, ferromagneticlike, exchange interaction, of strength $J > 0$, between the z nearest neighbors. The crystal is in a crystal-field dominated regime if J is much smaller than one of the anisotropy constants in the single-ion Hamiltonian $\mathcal{H}_i$. We will restrict ourselves to systems in which nearest-neighbor dipole-dipole forces are much weaker than exchange and to situations in which long-range dipole-dipole forces are either not of importance or can be incorporated into an effective applied field H. We are also always assuming that $k_B T$ is small compared to the energy separation of the single-ion ground and excited states. This will allow analysis of most processes from study of that part of the spin-wave Hamiltonian quadratic in boson operators, and the use of perturbation theory to estimate relaxation times arising from quartic terms.

Two types of anisotropy fields will be considered: uniaxial (either easy or hard axis) and orthorhombic. The simplest model which is able to display the role of the first two kinds of fields is a system of spins each with $s = 1$, since the lowest spin that can distinguish uniaxial and/or orthorhombic symmetry is one.

A spin-wave theory of spin-one paramagnets with the hard-axis anisotropy field has been already worked out by Tachiki, Yamada, and Maekawa.[1] These authors, however, take into account only the lowest two single-ion states, which is not right in a domain of anisotropy and magnetic fields where the third state is close to the second one.

The same system has been analyzed by Ishikawa and Oguchi[2] on the basis of a rigorous two-boson spin representation, which was first introduced by Homma, Okada, and Matsuda[3] in order to investigate rotational excitations (librons) in orthohydrogen molecules. Their representation is applicable only when the spins have the value of one, the anisotropy has the hard-axis form, the applied magnetic field is parallel to the anisotropy axis and, moreover, when the applied field is sufficiently small.

In this paper we transform a spin Hamiltonian into a magnon Hamiltonian in a two-step procedure. First we introduce sets of Bose particles which are excitable to individual single-ion levels and then we construct boson representations of spin operators by matching to the spin matrix elements between the single-ion eigenstates. This method can be applied to any kind of anisotropy, any value of spin, and essentially any value and direction of the magnetic field.

The method fails only in a small, of the order of J, region of values of the magnetic field where the exchange can no longer be treated as a perturbation. This happens when the anisotropy field becomes compensated by the applied field. Such region will be called the intermediate one, since it separates two phases with two different single-ion ground-state orderings, whenever the anisotropy field allows for such a transition. For instance, in the case of hard-axis anisotropy at sufficiently small magnetic fields applied parallel to the axis (which shall be called region S), the spins ($s=1$) in the ground state possess zero magnetic moment, i.e., are not aligned in any direction. But for sufficiently large fields (which shall be called region L) the spins point in the field direction. The intermediate region, which separates regions S and L, has been investigated by Tsuneto and Murao[4] in the molecular-field approximation and by Tachiki *et al.*[1] within the framework of their approximate spin-wave theory. Both these calculations indicate an onset of a transverse magnetization there. A more precise description of the intermediate region remains as an open problem since neither methods in which the exchange field is treated as a perturbation of the single-ion field, nor the methods (like that of Holstein and Primakoff) which take the exchange field as the dominant one, are applicable in this region. When the anisotropy has an easy axis no transitional phase occurs.

The second drawback of the method applied in this article is that it does not allow determination of the form of the important single-ion contributions to the quartic terms of the magnon Hamiltonian. These can be found only in situations where Homma *et al.*'s[3] representation is applicable, if the relationship between the two approaches is established; and therefore only then are we in position to discuss relaxation processes. In general the method is sufficient to determine spin-wave frequencies and to delineate possible effects of an applied rf field.

The method of matching matrix elements has been already employed by Grover[5] for the case of systems with hexagonal and cubic anisotropy fields. He takes into account only the lowest two single-ion states, restricts his theory to the case of vanishing applied field, and, moreover, he does not discuss the form of higher order than quadratic terms in the Hamiltonian. A one-boson method of matching matrix elements has been applied by Lindgård and Danielson and by Lindgård and Kowalska for systems with dominant exchange energy.[6] Their aim has been to find a modification of the Holstein-Primakoff representation which can account for substantial anisotropy-field distortions of the usual exchange-field results.

The plan of this article is as follows. In Sec. II we analyze spin-one systems with uniaxial and orthorhombic anisotropy in the presence of a magnetic field applied parallel to the uniaxial anisotropy field. The uniaxial anisotropy constant D and the orthorhombic anisotropy constant E can have any sign and any relative magnitude. It turns out that, when no field is applied, there are two possible collective ground-state arrangements and hence these systems can be either paramagnetic or ferromagnetic.

If D is of the hard-axis type, if $|E|<D$, and if this is a region of small (S) magnetic fields, the ground state is a singlet. In this situation we just generalize Ishikawa and Oguchi's[2] two-bosons results for a paramagnet with nonzero E and we compare the two approaches. Subsequently we set up a theory for systems in which the singlet is not the ground state (region L). This happens when the system is either a paramagnet in the region of large magnetic fields or if it is a ferromagnet.

In Sec. III possible effects of oscillatory magnetic fields are discussed. This subject does not seem to have been explored before. It turns out that, due to interplay between the orthorhombic anisotropy and the exchange, "parallel" pumping of various magnon pairs should be possible in sufficiently large oscillatory magnetic fields. The presence of orthorhombic anisotropy also breaks down a selection rule and allows an oscillatory field applied parallel to the uniaxis to produce incoherent resonant absorption between excited levels of the system. The usual coherent "perpendicular" resonance excitation of $k=0$ magnons out of the ground state can also take place, within certain restrictions. The conditions governing the excitation of these three possible absorption processes, namely of pumping, incoherent resonance, and coherent resonance, are listed and classified for the regions S and L.

In Sec. IV the pumping rates and threshold oscillatory fields are calculated for region S. Both direct pumping and pumping by way of virtual incoherent resonance excitation are discussed. In Sec. V our expectations about pumping rates in region L are outlined. Magnon relaxation times in region S are estimated in Sec. VI. In Sec. VII are listed a few typical materials with weak exchange couplings. That section also mentions some available extensions of the above theory to other situations and systems.

II. MAGNETIC FIELD PARALLEL TO THE UNIAXIAL ANISOTROPY FIELD

Consider a system with uniaxial anisotropy constant D and orthorhombic anisotropy constant E. The single-ion Hamiltonian reads

$$\begin{aligned}\mathcal{H}_i &= D(S_i^z)^2 + E[(S_i^x)^2 - (S_i^y)^2] - HS_i^z \\ &= D(S_i^z)^2 + \tfrac{1}{2}E[(S_i^+)^2 + (S_i^-)^2] - HS_i^z ,\end{aligned} \tag{2.1}$$

where H denotes the applied magnetic field multiplied by the Bohr magneton and by the Landé g factor.

If the spin $s=1$, then the single-ion Hamiltonian $\mathcal{H}_i$ has the three following eigenstates:

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

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

$$\mathcal{H}_i|0\rangle_i = 0|0\rangle_i . \tag{2.2c}$$

If the eigenstates of the operator S_i^z are denoted as $|+1\rangle_i, |-1\rangle_i$ and $|0\rangle_i$, then

$$\begin{aligned}|+\rangle_i = (1/\sqrt{2})\mathfrak{N}\{&[(H^2+E^2)^{1/2}+H]^{1/2}|-1\rangle_i \\ &+\eta[(H^2+E^2)^{1/2}-H]^{1/2}|+1\rangle_i\} ,\end{aligned} \tag{2.3a}$$

$$\begin{aligned}|-\rangle_i = (1/\sqrt{2})\mathfrak{N}\{&-\eta[(H^2+E^2)^{1/2}-H]^{1/2}|-1\rangle_i \\ &+[(H^2+E^2)^{1/2}+H]^{1/2}|+1\rangle_i\} ,\end{aligned} \tag{2.3b}$$

whereas the eigenstates corresponding to vanishing energy and to vanishing z component of the spin coincide. In the formulas (2.3)

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

and

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

In crystals without any orthorhombic anisotropy the states $|+\rangle_i$ and $|-\rangle_i$ are no longer mixtures of eigenstates of S_i^z. Then they are simply $|+\rangle_i = |-1\rangle_i, |-\rangle_i = |+1\rangle_i$. The three possible patterns of the single-ion levels[7] can be sketched as shown in Fig. 1, where the symbol $H_c^{(0)}$ is defined as $(D^2-E^2)^{1/2}$.

If the exchange field is thought of as a small perturbation, the products of these single-ion states remain the approximate eigenstates of the total Hamiltonian (1.1), except when $|H - H_c^{(0)}| = O(J)$, i.e., is of the order of J, as explained below.

Consider first a hard-axis material, i.e., one with $D>0$. Let the constant E attain any real value. For a given system the relative magnitudes of E and D can be manipulated, for instance, by applying an electric field or by chemical substitutional changes. If $|E|<D$ and if, as we shall see in Sec. II A, J being small does not exceed $(D-|E|)/4z$, then such systems are paramagnetic, since at magnetic fields H, which are smaller than some critical field $H_{c1} = H_c^{(0)} - O(J)$, the lowest-energy state is $|0\rangle_i$. It means that in the ground state the spins are not aligned in any direction. This domain of parameters H, D, E we shall call region S (small magnetic fields). In the region S the configuration of the eigenstates of the total Hamiltonian looks like in Fig. 1(a). For magnetic fields larger than a critical field $H_{c2} = H_c^{(0)} + O(J)$ the approximate eigenstates of the total Hamiltonian form the pattern as in Fig. 1(b), and we speak of region L. Now the ground state is formed out of the states $|-\rangle_i$. If $E=0$, this is a state in which spins are aligned along the z axis.

Regions L and S are separated by a small intermediate region in which exchange effects become important. For reasons explained in the Introduction we will exclude the intermediate region from our discussion.

Consider now hard-axis materials with $|E|\rangle D$. For any magnetic field such systems are in region

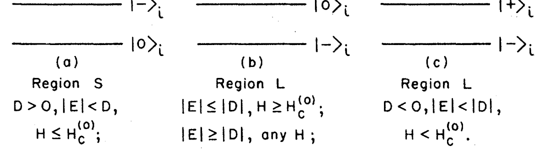

FIG. 1. Patterns of the single-ion energy levels, $H \geq 0$. Here $H_c^{(0)} \equiv (D^2 - E^2)^{1/2}$.

L. As we shall see in Sec. II D, if $6Jz<(|E|-D)$ these systems are paramagnets, if $2Jz\gtrsim|E|$, they are ferromagnets (and then J cannot be treated as a small parameter unless a sufficiently strong magnetic field is applied). For Jz between these two values we expect the systems to be in the intermediate region.

In the case of easy-axis systems $|-\rangle_i$ remains the ground state at any H. Note that the higher, $|+\rangle_i$ and $|0\rangle_i$, states switch their relative positions at $H=H_c^{(0)}$. This, however, does not lead to any physical transitions. Therefore a theory for the region L applies also to the level pattern as in Fig. 1(c). If $2Jz>|E|$, then, as shall be shown, the easy-axis systems are ferromagnets. In particular, when $E=0$, a ferromagnetic order is imposed by an exchange coupling of even infinitesimal strength. The crystals become paramagnetic if $2Jz<|E|$, with $|E|<|D|$, or $6Jz<(|E|+|D|)$, with $|E|>|D|$.

A. Paramagnets in region S: Homma-Okada-Matsuda representation

The spin-wave analysis of the paramagnet in the region S has been worked out by Ishikawa and Oguchi[2] for the case $E=0$. These authors employed the following Homma *et al.*'s[3] representation of the spin operators:

$$S_i^z=\tilde{a}_i^\dagger\tilde{a}_i-\tilde{b}_i^\dagger\tilde{b}_i\,, \tag{2.4a}$$

$$\begin{aligned}S_i^+&=\sqrt{2}(1-\tilde{b}_i^\dagger\tilde{b}_i)(\tilde{a}_i^\dagger+\tilde{b}_i)(1-\tilde{a}_i^\dagger\tilde{a}_i)\\&=\sqrt{2}(\tilde{a}_i^\dagger+\tilde{b}_i-\tilde{b}_i^\dagger\tilde{a}_i^\dagger\tilde{b}_i-\tilde{b}_i^\dagger\tilde{b}_i\tilde{b}_i\\&\qquad-\tilde{a}_i^\dagger\tilde{a}_i^\dagger\tilde{a}_i-\tilde{a}_i^\dagger\tilde{b}_i\tilde{a}_i+\cdots)\,,\end{aligned} \tag{2.4b}$$

$$S_i^-=(S_i^+)^\dagger\,, \tag{2.4c}$$

which is exact for $s=1$. In the Eqs. (2.4) $\tilde{a}_i^\dagger$, $\tilde{b}_i^\dagger$ are two sets of Bose operators that excite the $|+1\rangle_i$ and $|-1\rangle_i$ states from the states $|0\rangle_i\equiv|0,0\rangle_i$, namely $|+1\rangle_i=\tilde{a}_i^\dagger|0,0\rangle_i$, and $|-1\rangle_i=\tilde{b}_i^\dagger|0,0\rangle_1$. From Eq. (2.4) we get

$$\begin{aligned}(S_i^z)^2=&\,\tilde{a}_i^\dagger\tilde{a}_i+\tilde{b}_i^\dagger\tilde{b}_i+\tilde{a}_i^\dagger\tilde{a}_i^\dagger\tilde{a}_i\tilde{a}_i\\&+\tilde{b}_i^\dagger\tilde{b}_i^\dagger\tilde{b}_i\tilde{b}_i-2\tilde{b}_i^\dagger\tilde{a}_i^\dagger\tilde{b}_i\tilde{a}_i\,,\end{aligned} \tag{2.5a}$$

$$(S_i^+)^2=2\tilde{a}_i^\dagger\tilde{b}_i-2\tilde{b}_i^\dagger\tilde{a}_i^\dagger\tilde{a}_i^\dagger\tilde{b}_i-2\tilde{a}_i^\dagger\tilde{b}_i^\dagger\tilde{b}_i\tilde{b}_i+\cdots\,, \tag{2.5b}$$

$$(S_i^-)^2=[(S_i^+)^2]^\dagger\,. \tag{2.5c}$$

Ishikawa and Oguchi have proved that, if $E=0$, there are two branches of spin-waves excitations, with energies

$$\epsilon_k^\pm=[D^2-4JD\gamma(k)]^{1/2}\pm H\,, \tag{2.6}$$

where

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

and $\sum_\delta$ denotes the summation over the z-nearest neighbors. The critical field is defined by the condition $\epsilon_0^-=0$ (a soft-mode transition), which yields

$$H_{c1}=D(1-4Jz/D)^{1/2}\,. \tag{2.7}$$

Equation (2.7) coincides with the result of Tachiki *et al.*[1] as their replacement of the three-level system by a two-level one becomes justified for H close to H_{c1}.

Homma *et al.*'s representation is also applicable in the case of a crystal with orthorhombic anisotropy, provided the system is in the region S. With the use of Eqs. (2.4) and (2.5) we can transform (1.1) into a magnon Hamiltonian of the form

$$\begin{aligned}\mathcal{H}=&-4J\sum_{\langle ij\rangle}(\tilde{a}_i^\dagger\tilde{a}_j+\tilde{b}_i\tilde{a}_j+\tilde{a}_i^\dagger\tilde{b}_j^\dagger+\tilde{b}_j^\dagger\tilde{b}_i)\\&+D\sum_i(\tilde{a}_i^\dagger\tilde{a}_i+\tilde{b}_i^\dagger\tilde{b}_i)+E\sum_i(\tilde{a}_i^\dagger\tilde{b}_i+\tilde{b}_i^\dagger\tilde{a}_i)\\&-H\sum_i(\tilde{a}_i^\dagger\tilde{a}_i-\tilde{b}_i^\dagger\tilde{b}_i)+\cdots\,.\end{aligned} \tag{2.8}$$

The quartic and higher-order terms have been neglected for the moment. At first sight this seems to be a good approximation, since all of the dropped terms, except for those which originate from the $S_i^zS_j^z$ exchange interaction, either identically vanish on the three allowed states or correspond to a multiple excitation and deexcitation of bosons at the same site, which does not have any direct meaning when one thinks about spin reversals. They just describe interactions of the spin excitations. However, the neglected terms produce zero-point corrections to the frequency, which, as we shall see in a similar discussion in Sec. III, grow when H approaches H_{c1}. The quartic terms are also crucial for the discussion of the relaxation processes in the system.

If we introduce Fourier transformed operators

$$\tilde{a}_k=N^{-1/2}\sum_j e^{-ikR_j}\tilde{a}_j\,,$$

etc., we can write Eq. (2.8) as

$$\begin{aligned}\mathcal{H}=\sum_k[&\mathcal{A}_k^-\tilde{a}_k^\dagger\tilde{a}_k+\mathcal{A}_k^+\tilde{b}_k^\dagger\tilde{b}_k-2J\gamma(k)(\tilde{a}_k^\dagger\tilde{b}_{-k}^\dagger+\tilde{a}_k\tilde{b}_{-k})\\&+E(\tilde{a}_k^\dagger\tilde{b}_k+\tilde{a}_k\tilde{b}_k^\dagger)]+\cdots\,,\end{aligned} \tag{2.9}$$

where

$$\mathcal{A}_k^\pm=D\pm H-2J\gamma(k)\,.$$

With the use of the equation of motion method we get the following two frequencies:

$$\begin{aligned}\epsilon_k^\pm=(&E^2+D^2+H^2-4JD\gamma(k)\pm2\{H^2[D^2-4DJ\gamma(k)]\\&+E^2[D-2J\gamma(k)]^2\}^{1/2})^{1/2}\,.\end{aligned} \tag{2.10}$$

The expansion of these energies to order J will be used later [see Eq. (3.2)]. In the limit of $E\to0$ Eq. (2.10) coincides with Eq. (2.6). The value of the critical field is modified to

$$H_{c1}=[D^2-E^2-4(D+|E|)Jz]^{1/2}. \quad (2.11)$$

The effect of the orthorhombic anisotropy is to lower H_{c1} since the E interaction pulls the magnons out of the ground state. Owing to the presence of the zero-point corrections we cannot expect the terms proportional to J in H_{c1} to be accurate; rather they have the qualitative meaning that $H_{c1} = H_c^{(0)} - O(J)$.

Note that Eq. (2.10) requires at $H=0$ that the frequency ϵ_0^- becomes imaginary if $J>(D-|E|)/4z$. This supplies a criterion for the maximal size of the exchange constant.[8] If J is smaller than $(D-|E|)/4z$, the perturbational treatment of the exchange is justified. For J slightly bigger than this limiting value, or more precisely when $(D-|E|)/4z<J<(D+|E|)/4z$, the Hamiltonian becomes two-level-like and we expect the system to be in its intermediate region with a transverse magnetization present, since the $|+\rangle_i$ is only slightly admixed.[1,4] Systems with even bigger exchange, i.e., satisfying the inequality $J \gtrsim (D+|E|)/4z$ are ferromagnets. Modified Holstein-Primakoff representation[6] is applicable then.

B. Operator-matching method

Ishikawa and Oguchi's method is not applicable to the region L, since $|0\rangle_i$ is not the ground state.

An alternative approach to both regions L and S is furnished by introduction of Bose operators which are excitable to eigenstates of the single-ion Hamiltonian. Again two sets of bosons have to be used. One set is not enough, since, unlike the case of a crystal with anisotropy fields much smaller than the exchange field, the energy levels of the system are not in general equidistant. Especially when the higher two levels are close to each other the efforts to describe the system by one set of bosons are artificial and become more of a patchwork. Moreover in a one-boson theory the interactions with the highest level would automatically be transferred to the quartic terms.

Just to reproduce the single-ion part of the Hamiltonian, two sets of fermions would be much more adequate. This, however, would require magnons at different sites to anticommute rather than to commute. The price paid for use of commuting bosons is the occurence of quartic and higher-order terms in the single-ion Hamiltonian, since the bosons are not forbidden to be excited more than once at the same site.

The Bose operators should reproduce the following behavior of the spin operators:

$$S_i^z|+\rangle_i=\mathfrak{N}^2(E|-\rangle_i-H|+\rangle_i), \quad (2.12a)$$

$$S_i^z|-\rangle_i=\mathfrak{N}^2(E|+\rangle_i+H|-\rangle_i), \quad (2.12b)$$

$$S_i^z|0\rangle_i=0; \quad (2.12c)$$

$$S_i^+|+\rangle_i=\mathfrak{N}[(H^2+E^2)^{1/2}+H]^{1/2}|0\rangle_i, \quad (2.13a)$$

$$S_i^+|-\rangle_i=-\eta\mathfrak{N}[(H^2+E^2)^{1/2}-H]^{1/2}|0\rangle_i, \quad (2.13b)$$

$$S_i^+|0\rangle_i=\mathfrak{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}|-\rangle_i + \eta[(H^2+E^2)^{1/2}-H]^{1/2}|+\rangle_i\}; \quad (2.13c)$$

$$S_i^-|+\rangle_i=\eta\mathfrak{N}[(H^2+E^2)^{1/2}-H]^{1/2}|0\rangle_i, \quad (2,14a)$$

$$S_i^-|-\rangle_i=\mathfrak{N}[(H^2+E^2)^{1/2}+H]^{1/2}|0\rangle_i, \quad (2.14b)$$

$$S_i^-|0\rangle_i=\mathfrak{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}|+\rangle_i - \eta[(H^2+E^2)^{1/2}-H]^{1/2}|-\rangle_i. \quad (2.14c)$$

In the harmonic approximation the spin operators should have the following form:

$$S_i^z=\alpha+\beta b_i+\beta^* b_i^\dagger+\gamma a_i+\gamma^* a_i^\dagger+\delta a_i^\dagger b_i + \delta^* b_i^\dagger a_i+\epsilon a_i^\dagger a_i+\xi b_i^\dagger b_i+\eta a_i^\dagger a_i^\dagger+\eta^* a_i a_i + \theta b_i^\dagger b_i^\dagger+\theta^* b_i b_i+l a_i^\dagger b_i^\dagger+l^* a_i b_i+\cdots, \quad (2.15a)$$

$$S_i^+=\kappa+\lambda b_i+\lambda' b_i^\dagger+\mu a_i+\mu' a_i^\dagger+\nu a_i^\dagger b_i+\nu' b_i^\dagger a_i + \xi a_i^\dagger a_i+\sigma b_i^\dagger b_i+\pi a_i^\dagger a_i^\dagger+\pi' a_i a_i + \rho b_i^\dagger b_i^\dagger+\rho' b_i b_i+\sigma a_i^\dagger b_i^\dagger+\sigma' a_i b_i+\cdots, \quad (2.15b)$$

$$S_i^-=(S_i^+)^\dagger, \quad (2.15c)$$

where a_i, b_i are boson operators, and $\alpha, \beta, \ldots, \sigma'$ are constants to be determined. In a similar way one can find the subsequent third-order terms in the expansion. Analogously the structure of $(S_i^z)^2$, $(S_i^+)^2$, $(S_i^-)^2$, etc., can be established. The commutation rules are not helpful here as terms of arbitrary high order, when commuted, may produce constant, linear, and quadratic contributions.

This method, however, fails when one wants to find quartic and higher-order terms in S_i^z, S_i^+, S_i^-, $(S_i^z)^2, (S_i^-)^2$, since all such terms, which could enter the expressions for these operators with nonzero coefficients, either themselves or their Hermitian conjugates identically vanish in the allowed subspace of states. This deficiency can be removed only in the region S, where one can employ a relationship between our approach and that of Ishikawa and Oguchi [see Eq. (2.19)]. Consider first the region of magnetic fields smaller than $H_{c1}=H_c^{(0)}-O(J)$.

C. Operator-matching method: Region S

The scheme of the single-ion energy levels is as in Fig. 1(a). Let

$$|0\rangle_i=|0,0\rangle_i, \quad |-\rangle_i=a_i^\dagger|0,0\rangle_i, \quad |+\rangle_i=b_i^\dagger|0,0\rangle_i, \quad (2.16)$$

where $|0,0\rangle_i$ is the joint vacuum state for the two bosons. If we could impose the constraint $(a_i^\dagger)^2 = (b_i^\dagger)^2=0$, then the single-ion Hamiltonian would be simply

$$\mathcal{H}_i = [D + (H^2+E^2)^{1/2}]b_i^\dagger b_i + [D - (H^2+E^2)^{1/2}]a_i^\dagger a_i . \qquad (2.17)$$

Since in the case of bosons this constraint is artificial, Eq. (2.17) remains as an approximation and some quartic and higher-order terms have to be added. With the use of (2.12)–(2.15) we obtain

$$S_i^z = \mathfrak{N}^2[H(a_i^\dagger a_i - b_i^\dagger b_i) + E(a_i^\dagger b_i + b_i^\dagger a_i)], \qquad (2.18a)$$

$$S_i^+ = \mathfrak{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}(a_i^\dagger + b_i - a_i^\dagger a_i b_i - b_i^\dagger a_i^\dagger b_i - b_i^\dagger b_i b_i - a_i^\dagger a_i^\dagger a_i) + \eta[(H^2+E^2)^{1/2}-H]^{1/2}(b_i^\dagger - a_i - a_i^\dagger b_i^\dagger a_i + b_i^\dagger b_i a_i - b_i^\dagger b_i^\dagger b_i + a_i^\dagger a_i a_i)\} + \cdots, \qquad (2.18b)$$

$$S_i^- = (S_i^+)^\dagger, \qquad (2.18c)$$

which allows us to find the exchange part of the Hamiltonian.

Note that in the limit $E \to 0$ Eq. (2.18) coincides with (2.4). Note also that we would get exactly the same equations for S_i^z, S_i^+, S_i^- as (2.18) if we substituted

$$\tilde{a}_i = (\mathfrak{N}/\sqrt{2})\{\eta[(H^2+E^2)^{1/2}-H]^{1/2}b_i + [(H^2+E^2)^{1/2}+H]^{1/2}a_i\}, \qquad (2.19a)$$

$$\tilde{b}_i = (\mathfrak{N}/\sqrt{2})\{[(H^2+E^2)^{1/2}+H]^{1/2}b_i - \eta[(H^2+E^2)^{1/2}-H]^{1/2}a_i\} \qquad (2.19b)$$

into (2.4). As expected, the harmonic part of the single-ion Hamiltonian (2.9) becomes diagonal if written in terms of a_i's and b_i's. The above relation, together with Eq. (2.5), allows us to find the quartic contributions to $(S_i^z)^2$, $(S_i^+)^2$, and $(S_i^-)^2$. In this way we obtain the following form for the total Hamiltonian in the region S:

$$\mathcal{H} = \sum_k \{\mathcal{B}_k^- a_k^\dagger a_k + \mathcal{B}_k^+ b_k^\dagger b_k - 2J\mathfrak{N}^2\gamma(k)[H(a_k^\dagger b_{-k}^\dagger + a_k b_{-k}) + \tfrac{1}{2}E(b_k^\dagger b_{-k}^\dagger + b_k b_{-k} - a_k^\dagger a_{-k}^\dagger - a_k a_{-k})]\} + \mathcal{H}_{\mathrm{IV}}, \qquad (2.20)$$

where

$$\mathcal{B}_k^\pm = D \pm (H^2+E^2)^{1/2} - 2J\gamma(k)$$

and

$$\begin{aligned}\mathcal{H}_{\mathrm{IV}} = N^{-1}\sum_{k_1k_2k_3k_4} \delta(k_1+k_2-k_3-k_4)(&\phi_{12,34}b_{k_1}^\dagger a_{k_2}^\dagger b_{k_3}a_{k_4} + \psi_{12,34}^a a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3}a_{k_4} + \psi_{12,34}^b b_{k_1}^\dagger b_{k_2}^\dagger b_{k_3}b_{k_4}\\
&+ (a_{k_1}^\dagger a_{k_2}^\dagger b_{k_3}b_{k_4} + \text{H.c.})[\mathfrak{N}^4E^2(D+\tfrac{1}{2}H) - J\mathfrak{N}^4E^2\gamma(k_2-k_4)]\\
&+ (b_{k_1}^\dagger b_{k_2}^\dagger b_{k_3}a_{k_4} - a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3}b_{k_4} + \text{H.c.})\{\mathfrak{N}^4E[\tfrac{1}{2}E^2 + 2HJ\gamma(k_2-k_4)]\}\\
&+ (b_{k_1}^\dagger a_{k_2}^\dagger a_{-k_3}^\dagger a_{k_4} + \text{H.c.})\{-\tfrac{1}{4}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2} - 3H] + 2J\mathfrak{N}^2H[\gamma(k_1)+\gamma(k_2)]\}\\
&+ (a_{k_1}^\dagger b_{k_2}^\dagger b_{-k_3}^\dagger b_{k_4} + \text{H.c.})\{-\tfrac{1}{4}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2} + 3H] + 2J\mathfrak{N}^2H[\gamma(k_1)+\gamma(k_2)]\}\\
&+ (b_{k_1}^\dagger b_{k_2}^\dagger b_{-k_3}^\dagger b_{k_4} + \text{H.c.})[-\tfrac{1}{4}\mathfrak{N}^4E^3 + 2\mathfrak{N}^2EJ\gamma(k_1)] + (a_{k_1}^\dagger a_{k_2}^\dagger a_{-k_3}^\dagger a_{k_4} + \text{H.c.})[-\tfrac{1}{4}\mathfrak{N}^4E^3 - 2\mathfrak{N}^2EJ\gamma(k_1)]\\
&+ (b_{k_1}^\dagger b_{k_2}^\dagger a_{-k_3}^\dagger a_{k_4} + \text{H.c.})\{\tfrac{1}{4}E\mathfrak{N}^4[E^2 + 2H(H^2+E^2)^{1/2} - 2H^2] + 2\mathfrak{N}^2EJ\gamma(k_1)\}\\
&+ (a_{k_1}^\dagger a_{k_2}^\dagger b_{-k_3}^\dagger b_{k_4} + \text{H.c.})\{-\tfrac{1}{4}E\mathfrak{N}^4[2H^2 + 2H(H^2+E^2)^{1/2} - E^2] - 2\mathfrak{N}^2EJ\gamma(k_1)\}\\
&+ (b_{k_1}^\dagger b_{k_2}^\dagger b_{-k_3}^\dagger a_{k_4} + \text{H.c.})\tfrac{1}{4}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2} - H] + (a_{k_1}^\dagger a_{k_2}^\dagger a_{-k_3}^\dagger b_{k_4} + \text{H.c.})\tfrac{1}{4}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2} + H]),\end{aligned} \qquad (2.21)$$

with

$$\phi_{12,34} = 2\mathfrak{N}^4[D(E^2-H^2) + E^2H] + 2J[\gamma(k_1)+\gamma(k_2)+\gamma(k_3)+\gamma(k_4) + \mathfrak{N}^4H^2\gamma(k_1-k_3) - \mathfrak{N}^4E^2\gamma(k_1-k_4)], \qquad (2.22)$$

$$\psi_{12,34}^{a,b} = \mathfrak{N}^4\{H^2D \pm \tfrac{1}{2}E^2[(H^2+E^2)^{1/2} \mp H]\} + 2J[\gamma(k_1) + \gamma(k_4) - H^2\mathfrak{N}^4\gamma(k_2-k_4)]. \qquad (2.23)$$

In the harmonic approximation the $S_i^zS_j^z$ interaction again drops out and the frequencies are given by Eq. (2.10) which checks our method.

Now we are ready to apply our two-boson theory to the case of magnetic fields larger than $H_{c2} = H_c^{(0)} + O(J)$.

D. Operator-matching method: Region L

The single-ion level pattern is now as in Fig. 1(b) or 1(c). Since the ground state is $|-\rangle_i$, let

$$|-\rangle_i = |0,0\rangle_i, \quad |0\rangle_i = a_i^\dagger |0,0\rangle_i,$$
$$|+\rangle_i = b_i^\dagger |0,0\rangle_i .$$

With the use of Eqs. (2.12)–(2.15) we now obtain

$$S_i^z = \mathfrak{N}^2\{H + E(b_i^\dagger + b_i) - Ha_i^\dagger a_i - 2Hb_i^\dagger b_i - E(b_i^\dagger a_i^\dagger a_i + a_i^\dagger a_i b_i + b_i^\dagger b_i^\dagger b_i + b_i^\dagger b_i b_i)\} + \cdots , \tag{2.24a}$$

$$S_i^+ = \mathfrak{N}\{[(H^2+E^2)^{1/2}+H]^{1/2}(a_i + a_i^\dagger b_i - b_i^\dagger a_i b_i - a_i^\dagger a_i a_i) + \eta[(H^2+E^2)^{1/2} - H]^{1/2} \times (-a_i^\dagger + b_i^\dagger a_i + a_i^\dagger b_i^\dagger b_i + a_i^\dagger a_i^\dagger a_i)\} + \cdots , \tag{2.24b}$$

$$S_i^- = (S_i^+)^\dagger , \tag{2.24c}$$

Subsequent terms in the expansion of S_i^z, S_i^+, S_i^- cannot be found by means of our method.

In order to find the degree to which one can rely on the representation (2.24), when truncated to the quadratic terms, let us check the commutation relations for the spin operators. For instance $[S_i^+, S_i^-]$ reproduces $2S_i^z$ except for the coefficient in front of $b_i^\dagger b_i$, which turns out to be twice too small. For the domain of parameters which corresponds to the Fig. 1(b) the error thus introduced is insignificant. However, since (unlike the region S situation) the harmonic contribution to S_i^z does enter the harmonic exchange Hamiltonian, the b particles are, at low temperatures, only slightly excited. The error increases in the region corresponding to Fig. 1(c). Then b particles are more excited than a particles. On the other hand $[S_i^z, S_i^+]$ reproduces the linear terms of S_i^+ correctly, whereas the coefficients in front of $a_i^\dagger b_i$ and $b_i^\dagger a_i$ are exact in the limit $|E| \ll H$ only. These terms don't enter the harmonic part of the Hamiltonian anyway.

The single-ion Hamiltonian becomes

$$\mathcal{H}_i = D - (H^2+E^2)^{1/2} + 2(H^2+E^2)^{1/2} b_i^\dagger b_i + [(H^2+E^2)^{1/2} - D] a_i^\dagger a_i^\dagger \cdots , \tag{2.25}$$

because the energy difference between the states $|-\rangle_i$ and $|+\rangle_i$ is equal to $2(H^2+E^2)^{1/2}$, whereas between the states $|-\rangle_i$ and $|0\rangle_i$ it is $[(H^2+E^2)^{1/2} - D]$. Analysis of the structure of the operators $(S_i^z)^2$, $(S_i^+)^2$, and $(S_i^-)^2$ indicates that, the third-order terms of $-HS_i^z$ are canceled by the cubic terms of the anisotropy-field Hamiltonian. What the expression (2.25) does not include are some undetermined quartic contributions.

The total Hamiltonian for the region L reads

$$\mathcal{H} = N[D - (H^2+E^2)^{1/2} - Jz\mathfrak{N}^4 H^2] - 2Jz\mathfrak{N}^4 EHN^{1/2}(b_0^\dagger + b_0) + \sum_k [\mathcal{A}(k) a_k^\dagger a_k + JE\mathfrak{N}^2\gamma(k)(a_k^\dagger a_{-k}^\dagger + a_k a_{-k})]$$
$$+ \sum_k [\mathcal{B}(k) b_k^\dagger b_k - JE^2\mathfrak{N}^4\gamma(k)(b_k^\dagger b_{-k}^\dagger + b_k b_{-k})] + 2JH\mathfrak{N}^2 N^{-1/2} \sum_{k_1 k_2} \{-\gamma(k_1)(b_{k_1+k_2}^\dagger a_{k_1} a_{k_2} + a_{k_1}^\dagger a_{k_2}^\dagger b_{k_1+k_2})$$
$$+ \mathfrak{N}^2 E[z + \gamma(k_1)](b_{k_1}^\dagger a_{k_2}^\dagger a_{k_1+k_2} + a_{k_1+k_2}^\dagger b_{k_1} a_{k_2}) + \mathfrak{N}^2 E[z + 2\gamma(k_1)](b_{k_1}^\dagger b_{k_2}^\dagger b_{k_1+k_2} + b_{k_1+k_2}^\dagger b_{k_1} b_{k_2})\} + \cdots , \tag{2.26}$$

with

$$\mathcal{A}(k) = (H^2+E^2)^{1/2} - D + 2J\mathfrak{N}^4 H^2 z - 2J\gamma(k) , \tag{2.27}$$

$$\mathcal{B}(k) = 2(H^2+E^2)^{1/2} + 2J\mathfrak{N}^4[2H^2 z - E^2\gamma(k)] . \tag{2.28}$$

In order to eliminate the linear term $(b_0 + b_0^\dagger)$, let us perform the following unitary transformation of the Hamiltonian:

$$b_k = \beta_k + \delta_{k,0} N^{1/2} JEHz[(H^2+E^2)^{3/2} + 2Jz(H^2 - E^2)]^{-1}$$
$$= \beta_k + \delta_{k,0} N^{1/2} JEHz\mathfrak{N}^6 + O(J^2) . \tag{2.29}$$

If we limited ourselves to the linear and harmonic terms of Eq. (2.26), then under the influence of (2.29) the linear term would vanish. However this very transformation, when applied to higher-order terms, produces new linear and quadratic expressions. Now, the quartic and higher-order single-ion terms either contain less than two b operators or at least two such operators. In the first case no new linear or harmonic term is produced. In the second case we obtain negligible corrections which are proportional to J^2 and higher powers of J. On the other hand no triple- or higher-order exchange process generates lower-order terms which are proportional to J.

We conclude that

$$\mathcal{H} = N[D - (H^2+E^2)^{1/2} - Jz\mathfrak{N}^4 H^2] + \sum_k [\mathcal{A}(k) a_k^\dagger a_k + 2JE\mathfrak{N}^2\gamma(k)(a_k^\dagger a_{-k}^\dagger + a_k a_{-k})]$$
$$+ \sum_k [\mathcal{B}(k)\beta_k^\dagger \beta_k - JE^2\mathfrak{N}^4\gamma(k)(\beta_k^\dagger \beta_{-k}^\dagger + \beta_k \beta_{-k})] + O(J^2) + \cdots . \tag{2.30}$$

The transformation (2.29) can also lead to new cubic terms which are proportional to J. So the form of the triple terms in (2.30) becomes undetermined. In the first approximation with respect to J the a particles do not interact with the β particles. Further approximations bring in such interactions.

The energy associated with the a magnons is

$$\epsilon^{\alpha}(k) \simeq [\mathcal{A}^2(k) - 16J^2E^2\mathfrak{N}^4\gamma^2(k)]^{1/2}$$
$$= (H^2+E^2)^{1/2} - D + 2J[\mathfrak{N}^4H^2z - \gamma(k)] + O(J^2)\,. \qquad (2.31)$$

The dispersion relation for the β magnons is D independent and reads

$$\epsilon^{\beta}(k) \simeq \{4(H^2+E^2) + 8J\mathfrak{N}^2[2H^2z - E^2\gamma(k)] + 16J^2z\mathfrak{N}^8H^2[H^2z - E^2\gamma(k)]\}^{1/2}$$
$$= 2(H^2+E^2)^{1/2} + 2J\mathfrak{N}^4[2H^2z - E^2\gamma(k)] + O(J^2)\,. \qquad (2.32)$$

Now the case $D>0$, $|E|<D$ is interesting, since the energy of the less energetic (then) a particles vanishes in the vicinity of $H_c^{(0)}$. In the J term of the equation $\epsilon^{\alpha}(0)=0$ we can substitute $H_c^{(0)}$ instead of H. That way we obtain the following expression for H_{c2}:

$$H_{c2} = [D^2 - E^2 + 4JzE^2D^{-1} + O(J^2)]^{1/2}\,. \qquad (2.33)$$

This allows us to conclude that the width of the intermediate region is of the order of J. For $E=0$, $H_{c2} = D + O(J^2)$ is in agreement with the calculation of Tachiki *et al.*,[1] but as already mentioned, Eq. (2.33) has rather a qualitative meaning.

Let us discuss now the stability conditions for the region L at $H=0$. The $k=0$ a magnons become soft if $6Jz > (|E| - D)$, whereas the frequency $\epsilon^{\beta}(0)$ gives an instability when $2Jz > |E|$. It means that if $|E| \geq D$, the system should be in an intermediate phase for $(|E| - D)/3 < 2Jz < |E|$. Smaller J's give a paramagnetic case. If $2Jz > |E|$, the three states are heavily mixed and the system is a ferromagnet. On the other hand if D is negative and $|E| < |D|$, the crystal is ferromagnetic for $2Jz > |E|$. Depending on the size of J, the $|0\rangle_i$ state may or may not be strongly coupled by exchange, but when this state is only slightly admixed we do not expect to find an intermediate phase with a transversal magnetic moment since it does not influence the spin direction. Note, in particular, that a system with $E=0$ ($D<0$) is an easy axis ferromagnet for arbitrary small J. Then the single-ion ground state is degenerate at $H=0$, and our theory leads to unstable excitations.

Let us consider pattern (*c*) of Fig. 1 with very large negative D, so that the $|0\rangle_i$ level lies too high to be important. With $H=0$ the matrix elements of S_i^z are

$$\langle + |S_i^z| + \rangle = \langle - |S_i^z| - \rangle = 0\,,$$
$$|\langle + |S_i^z| - \rangle| = |\langle - |S_i^z| + \rangle| = 1\,. \qquad (2.34)$$

The situation is then that of two nonmagnetic levels, separated by $\Delta = 2|E|$, and possesing off-diagonal matrix elements of the magnetic moment (Van Vleck paramagnetism). When such a system of ions is coupled by exchange, Bleaney[9] has shown that—in the molecular-field approximation—ferromagnetism results when

$$J > \Delta/4z(\langle + |S_i^z| - \rangle)^2\,. \qquad (2.35)$$

On inspecting Eq. (2.32) we see that the $k=0$ β magnons become soft when J exceeds $|E|/2z$, in agreement with Bleaney's criterion. More precise calculations by Wang and Cooper[10] yield a (~10%) larger value of critical J. It is necessary to take into account the complicated ground state, i.e., the zero-point corrections to our assumed ground state which arise from the interplay between J and E. As we have pointed out below Eq. (2.11), we cannot expect large terms in J to have more than a general qualitative meaning.

We have established that for either sign of D the system, which at $H=0$ is in region L and for which the inequality $2Jz > |E|$ is satisfied, is a ferromagnet. Now, in the presence of a magnetic field our theory, in some instances, is applicable also to these ferromagnets. This is because stability conditions, which include H, allow increase of $2Jz$ beyond $|E|$.

Let us discuss first the situation $D<0$, $|E| < |D|$. Since in our calculations J was thought of as a small parameter, the results of the theory are meaningful if $J \ll |D|$, and if $J \ll H$. Therefore the theory applies to the ferromagnetic region when also $|E|$ is much smaller than $|D|$ and H. In particular consider a system which does not have any orthorhombic anisotropy field, or in other words let, say, $2H^2 \geq E^2$ (in order to guarantee nonimaginary ϵ^{β} for any J) and imagine that E is continuously switched off. In this situation none of the frequencies become soft for any, even vanishing, H. Except for the smallness requirements, J is not restricted then.

Now, if $|E| > |D|$ with D either positive or negative, the theory still applies for the ferromagnetic $2Jz > |E|$ provided the magnetic field is sufficiently strong, namely if $Jz \ll H$. In general the theory works whenever the single-ion ground state is, in comparison with J, significantly separated from the excited states.

III. EFFECTS OF OSCILLATORY MAGNETIC FIELDS

In ferromagnetic materials a uniform rf magnetic field $h\cos\omega t$ can trigger two effects[11,12]: (a) resonant production of $k=0$ magnons, and (b) (beyond a threshold) excitation of pairs of magnons with opposite wave vectors. Since usually the latter effect occurs with the oscillatory field applied parallel to a steady magnetic field, it is known as "parallel pumping."

In magnetic materials with uniaxial and orthorhom-

TABLE I. Possible effects of an oscillatory field.

Region	rf field along	Form of the rf perturbation	Process	Qualifications
S	z	$d_k^\dagger c_k$	Incoherent resonance	$E \neq 0$
(static field along z)		$d_k^\dagger c_{-k}^\dagger$	Parallel pumping of unlike magnons	$J \neq 0, E \neq 0$
		$c_k^\dagger c_{-k}^\dagger, d_k^\dagger d_{-k}^\dagger$	Parallel pumping of like magnons	$J \neq 0, E \neq 0, H \neq 0$
	x	$c_0^\dagger$	Coherent perpendicular resonance	$H \neq 0$ if $E > 0$; none if $E > 0$
		$d_0^\dagger$	Coherent perpendicular resonance	None if $E > 0$; $H \neq 0$ if $E < 0$
L	z	$d_0^\dagger$	Coherent parallel resonance	$E \neq 0$
(static field along z)		$c_k^\dagger c_{-k}^\dagger, d_k^\dagger d_{-k}^\dagger$	Parallel pumping of like magnons	$J \neq 0, E \neq 0, H \neq 0$
	x	$c_0^\dagger$	Coherent perpendicular resonance	$H \neq 0$
		$d_k^\dagger c_k$	Incoherent resonance	None if $E \geq 0$; $H \neq 0$ if $E < 0$
		$c_k^\dagger d_{-k}^\dagger$	"Parallel" pumping of unlike magnons	$J \neq 0, E \neq 0$

bic anisotropies and with weak exchange interactions, we will show that three oscillatory effects can take place: (a) resonant production of $k=0$ magnons, which will be called "coherent resonance"; (b) resonant production of a spectrum of magnons of different k, which will be called "incoherent resonance"; and (c) pumping of pairs of magnons beyond a threshold magnitude of rf perturbation. The presence of dipole-dipole forces is not required for these effects to occur, so we will continue to discuss systems in which dipolar fields are either ignored or incorporated into the uniform field H.

Various possible oscillatory effects under various possible conditions are listed and classified in Table I. They will now be discussed in detail.

A. Region S

Consider first the paramagnet in the region S. For small J the transformation

$$a_k = c_k - J\gamma(k)E\mathfrak{N}^2[D-(H^2+E^2)^{1/2}]^{-1}c_{-k}^\dagger + J\gamma(k)H\mathfrak{N}^2D^{-1}d_{-k}^\dagger + O(J^2)\,. \qquad (3.1a)$$

$$b_k = d_k + J\gamma(k)E\mathfrak{N}^2[D+(H^2+E^2)]^{1/2}]^{-1}d_{-k}^\dagger + J\gamma(k)H\mathfrak{N}^2D^{-1}c_{-k}^\dagger + O(J^2) \qquad (3.1b)$$

brings the quadratic part of Hamiltonian (2.20) to the diagonal form

$$\mathcal{H}_{\rm II} = \sum_k \epsilon_k^c c_k^\dagger c_k + \sum_k \epsilon_k^d d_k^\dagger d_k + O(J^2)\,, \qquad (3.2)$$

where

$$\epsilon_k^c = D-(H^2+E^2)^{1/2} - 2J\gamma(k) + O(J^2)\,, \qquad (3.3)$$

$$\epsilon_k^d = D+(H^2+E^2)^{1/2} - 2J\gamma(k) + O(J^2)\,, \qquad (3.4)$$

These energies agree with (2.10) when that equation is expanded to order J. The operators c_k, d_k satisfy boson commutation relations when terms quadratic in J are discarded.

Now we translate the quartic contributions (2.21) to the Hamiltonian into the language of c_k's and d_k's. Again we reject expressions proportional to J^n, $n \geq 2$. When, however, the normal ordering is introduced, some new quadratic terms appear, which are of the order of J. These are the zero-point corrections. If the theory based on the transformation (3.1) is to work, that is, if we assume that the original harmonic Hamiltonian gives a sufficient account of the dynamics of the system, these new terms have to be thrown out. Since the new terms involve coefficients from the transformation (3.1), which become large when H approaches H_{c1}, such theory fails to be reliable in the vicinity of H_{c1}. For the sake of consistency, the quartic terms with these very coefficients are also considered negligible. As a result the fourth-order terms are approximately given by (2.21), where a_k's and b_k's are replaced by c_k's and d_k's, respectively.

Let us look now at the operator of the z component of magnetization. From Eqs. (2.18a) and (3.1) it has the form

$$\sum_i S_i^z = \sum_k \Big(\mathfrak{N}^2 H(c_k^\dagger c_k - d_k^\dagger d_k) + \mathfrak{N}^2 E(d_k^\dagger c_k + c_k^\dagger d_k) - J\gamma(K)\mathfrak{N}^2 E\{2E[D^2 - (H^2+E^2)]^{-1}(d_{-k}^\dagger c_k^\dagger + c_k d_{-k}) + HD^{-1}[D-(H^2+E^2)^{1/2}]^{-1}(c_k^\dagger c_{-k}^\dagger + c_k c_{-k}) + HD^{-1}[D+(H^2+E^2)^{1/2}]^{-1}(d_k^\dagger d_{-k}^\dagger + d_k d_{-k})\}\Big) + O(J^2) + \cdots . \quad (3.5)$$

Thus magnons of a given wave vector k make the magnetization to oscillate with the following frequencies: (i) $\epsilon_k^d - \epsilon_k^c$, (ii) $\epsilon_{-k}^d + \epsilon_k^c$, (iii) $\epsilon_{-k}^c + \epsilon_k^c$, (iv) $\epsilon_{-k}^d + \epsilon_k^d$. Effects (i)–(iv) are, in the light of the preceding discussion, small outside the critical-field region; but they can be amplified by coupling to the rf field. All of the four motions disappear for a purely uniaxial paramagnet, i.e., when $E=0$, since then the magnetization is conserved. If H vanishes, (i) and (ii) still take place.

If an oscillatory magnetic field is applied in the z direction, a perturbation of the form

$$\delta\mathcal{H} = \delta\mathcal{H}' \cos\omega t \equiv -\cos(\omega t) h \sum_i S_i^z \quad (3.6)$$

has to be added to the Hamiltonian (2.20). The perturbation will trigger one of the four following processes:

(i) If the condition

$$\omega = \epsilon_k^d - \epsilon_k^c = 2(H^2+E^2)^{1/2} + O(J^2)$$

is met, c_k magnons are annihilated and d_k-magnons are created. Since $\epsilon_k^d - \epsilon_k^c$ is independent of k to first order in J, when $\omega = 2(H^2+E^2)^{1/2}$, magnons of all wave vectors participate in these processes. Larger J will provide broadening. This is induced absorption which in effect transfers a portion of the system from the c level to the d level. The reverse processes, induced emissions, also take place. The net power absorbed will be given by subtracting emissions from absorptions and will be proportional to the difference in population between the c and d levels. Since neither c nor d is the ground state, the net absorption will be small. Furthermore, it is unlike the absorption out of the ground state. The latter is produced by a component of rf field which is normal to a component of ground-state magnetic moment (spontaneous, or induced by H); the component of the rf field causes that magnetic moment to precess coherently, i.e., excites $k=0$ magnons. Instead, the absorption between excited states c and d is produced by a component of rf which ordinarily cannot connect the two states, but which here is allowed because of a perturbation mixing the two states, in this case due to E. Furthermore, the absorption takes place across a spread of k vectors and therefore cannot be described as a coherent motion. We call it *incoherent resonance*.

(ii) If the condition

$\omega = \epsilon_k^c + \epsilon_{-k}^d = 2D - 4J\gamma(k) + O(J^2)$ is satisified, two particles c_k and d_{-k} are produced. This is nothing else but a "parallel" pumping, which is allowed by an interplay between the exchange and orthorhombic anisotropy fields. Only one wave vector is involved now, provided the incident radiation is sufficiently monochromatic, i.e., provided the spectral width of the rf field is much smaller than J. This process requires a threshold size rf, which we calculate in Sec. IV. In this process it might seem that two different atomic levels are being simultaneously excited. However, the excitation is of magnons belonging to the whole crystal, i.e., of levels shared by all of the atoms. We make the usual small-magnon-numbers approximation that these modes do not interfere kinematically with each other.

(iii) For

$$\omega = \epsilon_k^c + \epsilon_{-k}^c = 2D - 2(H^2+E^2)^{1/2} - 4J\gamma(k) + O(J^2),$$

pumping of c magnons occurs; and (iv) for

$$\omega = \epsilon_k^d + \epsilon_{-k}^d = 2D + 2(H^2+E^2)^{1/2} - 4J\gamma(k) + O(J^2)$$

we have pumping of d magnons. Processes (iii) and (iv) disappear in the limit of $H \to 0$.

Unlike the usual ferromagnetic case,[11,12] the pumping phenomena here are not, at least in the first approximation, due to a simple elliptical motion of the transverse components of the spins. To see this, let us write down the expressions for S_i^x and S_i^y for $H=0$ and $E>0$:

$$S_i^x = N^{-1/2} \sum_k e^{-ikr_i}[d_k^\dagger + d_{-k} + J\gamma(k)(D+E)^{-1}(d_k^\dagger + d_{-k})] + O(J^2) + \cdots ,$$

$$S_i^y = N^{-1/2} \sum_k e^{-ikr_i}[c_{-k} - c_k^\dagger + J\gamma(k)(D-E)^{-1}(c_{-k} + c_k^\dagger)] + O(J^2) + \cdots .$$

where the dots represent triple and higher-order interactions. Thus for a given wave-vector, S_i^x participates in a motion with frequency ϵ_k^d, whereas S_i^y—with ϵ_k^c; thus Lisajous curves are followed.

Before we discuss the pumping rates for (ii)–(iv), let us review what happens in other configurations of the static and of the oscillatory magnetic fields.

If, in the region S, the rf field is applied along the x axis, it will couple to

$$\sum_i S_i^x = \tfrac{1}{2}N^{1/2}(V_-\{1 - JzE\mathfrak{N}^2[D-(H^2+E^2)^{1/2}]^{-1}\} + V_+ Jz\mathfrak{N}^2HD^{-1})(c_0^\dagger + c_0) + \tfrac{1}{2}N^{1/2}(V_+\{1+JzE\mathfrak{N}^2[D+(H^2+E^2)^{1/2}]^{-1}\} + V_- Jz\mathfrak{N}^2HD^{-1})(d_0^\dagger + d_0)\,, \tag{3.7}$$

where

$$V_\pm = (1+H\mathfrak{N}^2)^{1/2} \pm \eta(1-H\mathfrak{N}^2)^{1/2}\,.$$

Now two *coherent resonances* can take place, i.e., $k=0$ magnons are excited only. The c process disappears for $H=0$ (if $E>0$). On the other hand if $\omega=\epsilon_0^d$, at any H, the d_0 particles are produced ($E>0$). Similar effects are to be observed when the rf field points in the y direction. The only difference is that in the limit of $H\to 0$ the c_0 particles would be produced only.

B. Region L

Consider now systems in the region L. The Hamiltonian (2.30) becomes diagonal under the transformation

$$\alpha_k = c_k - J\gamma(k)E\mathfrak{N}^2[(H^2+E^2)^{1/2}-D]^{-1}c_{-k}^\dagger + O(J^2)\,,$$
$$\beta_k = d_k + \tfrac{1}{2}J\gamma(k)E^2\mathfrak{N}^6 d_{-k}^\dagger + O(J^2)\,. \tag{3.8}$$

This yields

$$\sum_i S_i^z = NH\mathfrak{N}^2(1+\mathfrak{N}^6E^2zJ) + N^{1/2}W_1(d_0^\dagger + d_0) - \mathfrak{N}^2H\sum_k (c_k^\dagger c_k + 2d_k^\dagger d_k - J\gamma(k)\mathfrak{N}^2E \times\{[(H^2+E^2)^{1/2}-D]^{-1}(c_k^\dagger c_{-k}^\dagger + c_kc_{-k}) - \mathfrak{N}^4(d_k^\dagger d_{-k}^\dagger + d_kd_{-k})\}) + O(J^2) + \cdots\,, \tag{3.9}$$

and

$$\sum_i S_i^x = N^{1/2}W_2(c_0^\dagger + c_0) + \tfrac{1}{2}V_+\sum_k \{d_k^\dagger c_k + c_k^\dagger d_k - \tfrac{1}{2}J\gamma(k)\mathfrak{N}^6E^2[(H^2+E^2)^{1/2}+D][(H^2+E^2)^{1/2}-D]^{-1} \times (c_k^\dagger d_{-k}^\dagger + c_kd_{-k})\} + O(J^2) + \cdots\,, \tag{3.10}$$

where

$$W_1 = \mathfrak{N}^2E(1+\tfrac{1}{2}JzE\mathfrak{N}^4 - 2Jz\mathfrak{N}^6H^2)\,,$$
$$W_2 = \tfrac{1}{2}(V_-\{1 - Jz\mathfrak{N}^2E[(H^2+E^2)^{1/2}-D]^{-1}\} + V_+JzEH\mathfrak{N}^6)\,.$$

In the region L a coherent resonance occurs for the rf field applied in either z or x direction. In the former case it vanishes for $E=0$, whereas in the latter it vanishes for $H=0$. For the rf field applied along the z axis, pumping of particles of the same kind takes place, provided H is nonzero. There is no trace of the incoherent resonance in such configuration of the fields. If the rf field points in the x direction, the pumping of particles of different kinds is possible, provided E does not vanish. An onset of the incoherent resonance is also to be observed then.

IV. PUMPING RATES IN THE REGION S

Consider at the beginning the pumping of two particles $c_k^\dagger$, $d_{-k}^\dagger$, which happens when $\omega = 2D - 4J\gamma(k)$. This is process (ii) of Sec. III A. As in the case of an antiferromagnet in the spin-flop phase,[13,14] there are two mechanisms which contribute to the pumping rate: (a) direct pumping due to the $d_{-k}^\dagger c_k^\dagger$ term in the perturbation (3.6); and (b) indirect pumping via creation of a virtual "particle" $d_{k'}^\dagger c_{k'}$ and subsequent anihilation of that "particle" and creation of the pair $c_k^\dagger d_{-k}^\dagger$, via a quartic term in $c_k^\dagger d_{-k}^\dagger c_{k'}^\dagger d_{k'}$. The perturbation (3.6) can supply such "particles" since they appear as nonvirtual when conditions for the incoherent resonance are met. In contradistinction to the pumping rate in the antiferromagnet, the virtual "particle" is a two-body object now, which moreover carries a nonzero momentum.

(a) Let us evaluate first the rate of the direct pumping. According to the standard time-dependent first-order perturbation theory,[15] the transition probability per unit time that the number of particles c_k, n_k^c, and the number of particles d_{-k}, n_{-k}^d, both increase by one is

$$\tfrac{2}{4}\pi|\langle n_k^c+1, n_{-k}^d+1|\delta\mathcal{H}'|n_k^c, n_{-k}^d\rangle|^2\delta(\omega-2D+4J\gamma(k)) = \tfrac{2}{4}\pi 4J^2\gamma^2(k)h^2\mathfrak{N}^4E^4[D^2-(H^2+E^2)]^{-2} \times (n_k^c+1)(n_{-k}^d+1)\delta(\omega-2D+4J\gamma(k))\,. \tag{4.1}$$

The transition probability of the inverse process, in which n_k^c and n_{-k}^d decrease by one, is now subtracted from the above; and the net rate of growth is

$$(\dot{n}_k^c)_{\text{growth},a}=(\dot{n}_{-k}^d)_{\text{growth},a}=2\pi J^2\gamma^2(k)h^2\mathfrak{N}^4E^4\times[D^2-(H^2+E^2)]^{-2}(n_k^c+n_{-k}^d+1)\times\delta(\omega-2D+4J\gamma(k)). \quad (4.2)$$

The larger h, E, H, J the more effective the direct pumping is, and it vanishes when $\gamma(k)=0$.

(b) Let us proceed now to the calculation of the indirect pumping rate. Note first that the quartic Hamiltonian $\mathcal{H}_{\text{IV}}$ can also be considered as a perturbation. In particular the term

$$\mathcal{H}_{\text{IV}}^{cd}=-(4N)^{-1}\mathfrak{N}^4E[2H^2+2H(H^2+E^2)^{1/2}-E^2]\times 2\sum_{k''}(d_{-k}^\dagger c_k^\dagger c_{k''}^\dagger d_{k''}+d_{k''}^\dagger c_{k''}c_k d_{-k}) \quad (4.3)$$

can destroy the virtual "particle" $d_k^\dagger c_k$ and produce magnons which appear as a result of the direct process. The factor of 2 in (4.3) counts the number of ways interactions ($c_{k_1}^\dagger c_{k_2}^\dagger d_{-k_3}^\dagger d_{k_4}$ +H.c.) are in the desired configuration of wave-vectors. Coefficient proportional to J has been neglected in (4.3). The second-order perturbation theory gives the following expression for the transition rate in such process:

$$\tfrac{2}{4}\pi\Big|\sum_{k'\neq k}(\epsilon_{k'}^d-\epsilon_{k'}^c)^{-1}\langle n_{-k}^d+1, n_k^c+1, n_{k'}^d, n_{k'}^c|\mathcal{H}_{\text{IV}}^{cd}|n_{-k}^d, n_k^c, n_{k'}^d+1, n_{k'}^c-1\rangle\times\langle n_{-k}^d, n_k^c, n_{k'}^d+1, n_{k'}^c-1|\delta\mathcal{H}'|n_{-k}^d, n_k^c, n_{k'}^d, n_{k'}^c\rangle\Big|^2\delta(\omega-2D+4J\gamma(k)). \quad (4.4)$$

Only the term

$$\mathfrak{N}^2hE\sum_{k''}(d_{k''}^\dagger c_{k''}+c_{k''}^\dagger d_{k''})$$

contributes to the matrix elements of $\delta\mathcal{H}'$ in (4.4). If the transition rate for the inverse process is subtracted from (4.4), for the net rate we get

$$(\dot{n}_k^c)_{\text{growth},b}=(\dot{n}_{-k}^d)_{\text{growth},b}=\tfrac{2}{64}\pi(H^2+E^2)^{-1}(\mathfrak{N}^2hE)^2\{N^{-1}\mathfrak{N}^4E[2H^2+2H(H^2+E^2)^{1/2}-E^2]\}^2\times\left(\Big|\sum_{k'\neq k}(n_{k'}^d+1)n_{k'}^c(n_{-k}^d+1)^{1/2}(n_k^c+1)^{1/2}\Big|^2-\Big|\sum_{k'\neq k}n_{k'}^d(n_{k'}^c+1)(n_k^c)^{1/2}(n_{-k}^d)^{1/2}\Big|^2\right)\delta(\omega-2D+4J\gamma(k)). \quad (4.5)$$

The k' wave vector which is equal to k gives a nonzero contribution to (4.5) when $k=0$ only. The case $k=0$ is excluded from our discussion from now on.

Assume that the occupation numbers of magnons with wave vectors k' are almost at their equilibrium values, corresponding to temperatures $T \gg J/k_B$. This allows us to take $n_{k'}^c$ and $n_{k'}^d$ as Bose factors, in which J dependence, and therefore k' dependence, is disregarded:

$$n_{k'}^c\approx\bar{n}^c\equiv(\exp\{[D-(H^2+E^2)^{1/2}]/k_BT\}-1)^{-1}, \quad (4.6)$$

$$n_{k'}^d\approx\bar{n}^d\equiv(\exp\{[D+(H^2+E^2)^{1/2}]/k_BT\}-1)^{-1}. \quad (4.7)$$

At temperatures $k_BT \ll D+(H^2+E^2)^{1/2}$ the term $\bar{n}^d\bar{n}^c$ is much smaller than $\bar{n}^c$, so

$$\sum_{k'\neq k}(\bar{n}^d+1)\bar{n}^c\approx(N-1)\bar{n}^c\approx N\bar{n}^c.$$

Equation (4.5) becomes

$$(\dot{n}_k^c)_{\text{growth},b}=(\dot{n}_{-k}^d)_{\text{growth},b}\approx\tfrac{2}{64}\pi\mathfrak{N}^{16}E^4h^2[2H^2+2H(H^2+E^2)^{1/2}-E^2]^2\times\{[(\bar{n}^c)^2-(\bar{n}^d)^2]n_k^cn_{-k}^d+(\bar{n}^c)^2(n_{-k}^d+n_k^c+1)\}\times\delta(\omega-2D+4J\gamma(k)). \quad (4.8)$$

Up to the moment when an instability develops in the system, the nonlinear term $n_k^cn_{-k}^d$ is not so decisive as $n_{-k}^d+n_k^c+1$ and will be dropped.

Now, Eqs. (4.2) and (4.8) have been derived under the assumption that the initial and final states were stationary ones. Actually this is not the case. Owing to various relaxation mechanisms, caused, for instance by the quartic terms in the Hamiltonian, occupation numbers of the produced particles decay with some finite lifetimes $1/\Gamma_k^c$ and $1/\Gamma_{-k}^d$, respectively,

$$(\dot{n}_k^c)_{\text{decay}}=-\Gamma_k^c(n_k^c-\bar{n}_k^c), \quad (4.9)$$

$$(\dot{n}_{-k}^d)_{\text{decay}}=-\Gamma_{-k}^d(n_{-k}^d-\bar{n}_{-k}^d), \quad (4.10)$$

where $\bar{n}_k^c$ and $\bar{n}_{-k}^d$ denote the equilibrium occupation numbers. Hence the two-body wave function of the two produced particles is damped with the damping constant $\frac{1}{2}(\Gamma_k^c+\Gamma_{-k}^d)$. As a result transitions occur also in the immediate vicinity of the "resonant" frequency. Following Callen,[16] we replace the δ function in (4.2) and (4.8) by the Lorentzian line-shape function

$$\frac{1}{2\pi}\frac{\Gamma_k^c+\Gamma_{-k}^d}{[\omega-2D+4J\gamma(k)]^2+(\frac{1}{4})(\Gamma_k^c+\Gamma_{-k}^d)^2}\,. \qquad (4.11)$$

We arrive at two coupled linear differential equations for n_k^c and n_{-k}^d:

$$\begin{aligned}\dot{n}_k^c&=(\dot{n}_k^c)_{\text{growth},a}+(\dot{n}_k^c)_{\text{growth},b}+(\dot{n}_k^c)_{\text{decay}}\,,\\ \dot{n}_{-k}^d&=(\dot{n}_{-k}^d)_{\text{growth},a}+(\dot{n}_{-k}^d)_{\text{growth},b}+(\dot{n}_{-k}^d)_{\text{decay}}\,.\end{aligned} \qquad (4.12)$$

For small enough h, the solutions of (4.12) die out exponentially: the system drives back to equilibrium. However at the critical field, h_{cr} given by

$$\begin{aligned}h_{\text{cr}}&=(\Gamma_k^c\Gamma_{-k}^d)^{1/2}(\Gamma_k^c+\Gamma_{-k}^d)^{-1}\\ &\times\{[\omega-2D+4J\gamma(k)]^2+\tfrac{1}{4}(\Gamma_k^c+\Gamma_{-k}^d)^2\}^{1/2}(E\mathfrak{N})^{-2}\\ &\times\{J^2\gamma^2(k)[D^2-(H^2+E^2)]^{-2}+\tfrac{1}{64}\mathfrak{N}^{12}(\bar{n}^c)^2\\ &\times[2H^2+2H(H^2+E^2)^{1/2}-E^2]^2\}^{-1/2}\end{aligned} \qquad (4.13)$$

an onset of instability takes place and an abrupt increase in the power absorption is to be observed. Beyond this field our linear spin-wave theory fails. The critical field is never reached when $E=0$ as the pumping does not appear then. The minimal value of h_{cr} occurs at the center of the line, i.e., when $\omega=2D-4J\gamma(k)$. Then

$$\begin{aligned}h_{\text{cr}}^{\min}&=\tfrac{1}{2}(\Gamma_k^c\Gamma_{-k}^d)^{1/2}(E\mathfrak{N})^{-2}\\ &\times\{J^2\gamma^2(k)[D^2-(H^2+E^2)]^{-2}+\tfrac{1}{64}\mathfrak{N}^{12}(\bar{n}^c)^2\\ &\times[2H^2+2H(H^2+E^2)^{1/2}-E^2]^2\}^{-1/2}\,.\end{aligned} \qquad (4.14)$$

At temperatures $k_BT\ll D-(H^2+E^2)^{1/2}$ the temperature-independent term in (4.14) is the dominant one. On the other hand, at temperatures comparable to $D-(H^2+E^2)^{1/2}$ this term is negligible, provided H is not too close to H_{c1}.

Now, in Sec. VI we will show that, when $k_BT\ll D+(H^2+E^2)^{1/2}$, and if $(H^2+E^2)^{1/2}\neq\frac{1}{2}D+O(J)$, then

$$\Gamma_k^c\propto J^{-1}\mathfrak{N}^8\{H^2D+\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}-H]\}^2\bar{n}^c\,, \qquad (4.15a)$$

$$\Gamma_{-k}^d\propto J^{-1}\mathfrak{N}^8[D(E^2-H^2)+E^2H]^2\bar{n}^c\,. \qquad (4.15b)$$

So in the low-temperature regime

$$\begin{aligned}h_{\text{cr}}^{\min}&\propto\bar{n}^c[2J^2|\gamma(k)|E^2(H^2+E^2)^{3/2}]^{-1}\\ &\times\{H^2D+\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}-H]\}\\ &\times[D(E^2-H^2)+E^2H][D^2-(H^2+E^2)]\,,\end{aligned} \qquad (4.16a)$$

and the pumping is most effective for small (k close to 0) and large (k from vicinities of the Brillouin-zone corners) wave vectors. It becomes impossible when $\gamma(k)=0$. [The k-dependent coefficients of proportionality, which were not written down in (4.15), don't seem to vanish anywhere.]

To simplify our discussion, consider the case $H=0(E\neq0)$. Then (4.16a) simplifies to

$$h_{\text{cr}}^{\min}\propto D(D^2-E^2)[2J^2|\gamma(k)|]^{-1}\bar{n}^c\,. \qquad (4.16b)$$

If the temperature is so low that $\bar{n}^c\approx J^2z/(D^2-E^2)$, then the amplitude of the rf field has to be roughly equal to D in order to trigger the rise in the power absorption. At larger temperatures $h_{\text{cr}}^{\min}$ increases and becomes

$$h_{\text{cr}}^{\min}\propto\frac{D}{4J}\left[\left(\frac{J}{D-|E|}\right)^2\frac{\gamma^2(k)}{(\bar{n}^c)^2}\frac{1}{(D+E)^2}+\frac{1}{(8E)^2}\right]^{-1/2}. \qquad (4.17)$$

In particular when k_BT is comparable to $D-|E|$, and if $J/(D-|E|)\lesssim\frac{1}{8}$, the first term, as already mentioned, is negligible and

$$h_{\text{cr}}^{\min}\propto 2D|E|J^{-1}\,. \qquad (4.18)$$

We conclude that an experimental observation of the parallel pumping of c and d magnons would require large amplitudes of the rf field, unless the experiment is done at very low temperatures.

Let us discuss now the pumping of magnons of the same kind [processes (iii) and (iv) of Sec. III A]. In the case of c magnons the quartic interaction involved in pumping via virtual particles is

$$\begin{aligned}\mathcal{H}_{\text{IV}}^{cc}&=(4N)^{-1}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2}+H]\\ &\times3\sum_{k''}(c_k^\dagger c_{-k}^\dagger c_{k''}^\dagger d_{k''}+d_{k''}^\dagger c_{k''}c_kc_{-k})\end{aligned} \qquad (4.19a)$$

and, in the case of d magnons,

$$\begin{aligned}\mathcal{H}_{\text{IV}}^{dd}&=(4N)^{-1}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2}-H]\\ &\times3\sum_{k''}(d_k^\dagger d_{-k}^\dagger c_{k''}^\dagger d_{k''}+d_{k''}^\dagger c_{k''}d_kd_{-k})\,.\end{aligned} \qquad (4.19b)$$

The factor of 3 appears because of the counting reasons. In place of (4.14) we obtain this time

$$\begin{aligned}h_{\text{cr};c,d}^{\min}&=(\Gamma_k^{c,d}\Gamma_{-k}^{c,d})^{1/2}(E\mathfrak{N}^2)^{-1}\\ &\times\{J^2\gamma^2(k)H^2D^{-2}[D\mp(H^2+E^2)^{1/2}]^{-2}\\ &+\tfrac{9}{64}\mathfrak{N}^{12}(\bar{n}^c)^2E^2[(H^2+E^2)^{1/2}\pm H]^2\}^{-1/2}\,.\end{aligned} \qquad (4.20)$$

At low temperatures the second term can be discarded, and

$$\begin{aligned}h_{\text{cr},c}^{\min}&\propto\bar{n}^c[J^2|\gamma(k)|\,|E|H(H^2+E^2)^{1/2}]^{-1}D[D-(H^2+E^2)^{1/2}]\\ &\times\{H^2D+\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}-H]\}^2\,,\end{aligned} \qquad (4.21)$$

$$h_{cr,d}^{min} \propto \bar{n}^c [J^2|\gamma(k)|\, |E|\, H(H^2+E^2)^{1/2}]^{-1} D[D+(H^2+E^2)^{1/2}] \times \{D(E^2-H^2)+E^2H\}^2 \quad (4.22)$$

For vanishing H or E, h_{cr}^{min} becomes infinite. At temperatures comparable to $[D-(H^2+E^2)^{1/2}]/k_B$ the first term is negligible and

$$h_{cr,c}^{min} \propto 8\{3JE^2[(H^2+E^2)^{1/2}+H](H^2+E^2)^{1/2}\}^{-1} \times \{H^2D+\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}-H]\}^2 \underset{H=0}{=} \tfrac{2}{3}E^2J^{-1}, \quad (4.23)$$

$$h_{cr,d}^{min} \propto 8\{3JE^2[(H^2+E^2)^{1/2}-H](H^2+E^2)^{1/2}\}^{-1} \times \{D(E^2-H^2)+E^2H\}^2 \underset{H=0}{=} \tfrac{8}{3}D^2J^{-1}. \quad (4.24)$$

Note that in the limit $H=0$ the virtual processes, unlike the direct ones, still pump magnons into the system. Finally, it is easier to pump the less energetic c magnons than the d magnons.

V. PUMPING IN THE REGION L

In the region L a parallel rf field can both pump particles of the same kind, and supply a coherent resonance with d_0 magnons [see Eq. (3.9)]. The critical field for parallel pumping of d magnons is smaller than an analysis of the direct process would indicate. This is caused by triple interactions in which a virtual d_0 magnon is annihilated. This decrease, however, should be a small effect, since the cubic terms in the Hamiltonian are proportional to J. We are not able to find the exact rate of pumping due to the virtual processes, as we do not know the actual form of the cubic terms. The perpendicular-rf-field pumping [Eq. (3.10)] of unlike magnons should be much easier to perform than the parallel pumping in region S, since two kinds of virtual processes increase the pumping rate there: annihilation of a virtual magnon c_0 and annihilation of a virtual "particle" $d_k c_k^\dagger$, both followed by creation of the pair $d_{-k}^\dagger c_k^\dagger$.

In the region L the form of the single-ion quartic terms is not known. Therefore we are not able either to find the rates for the virtual processes or to find the relaxation times. The subsequent analysis for the region S indicates that the known exchange quartic interactions have little influence on the damping.

VI. RELAXATION TIMES IN THE REGION S

In typical systems which we have been discussing, in the region S, the principal source of relaxation will be the four-magnon processes which are caused by anisotropy and exchange fields.

Three-magnon processes, which originate from the bulk dipolar field, will be important only when this field is large compared to both anisotropy and exchange fields, which we have assumed not to be the case. Two-magnon processes, which are due to demagnetizing fields of surface and bulk imperfections, are diminished as samples are made more nearly perfect, and in any event are invoked only to account for those transitions not conserving crystalline momentum and hence possible only at imperfections. Finally we assume weak magnon-phonon interactions, as should usually be the case in systems with weak exchange coupling, unless that coupling has very large spatial derivatives. Thus we concentrate on the four-magnon processes, since they will usually be the processes of greatest intrinsic importance in systems with large single-ion anisotropy and weak exchange.

In the quartic Hamiltonian (2.21) in which a's and b's are replaced by c's and d's, respectively, not all processes are allowed energetically for small J. For instance the interactions $d^\dagger c^\dagger c^\dagger c$, $c^\dagger d^\dagger d^\dagger d$, $d^\dagger d^\dagger d^\dagger d$, $c^\dagger c^\dagger c^\dagger c$, and $d^\dagger d^\dagger d^\dagger c$ are not allowed for any value of H and E. For $(H^2+E^2)^{1/2}=O(J)$, $c^\dagger c^\dagger dd$, $c^\dagger c^\dagger cd$, and $d^\dagger d^\dagger dc$ would be allowed, but this requires E to be small and then the pumping process is inefficient. Finally the interaction $c^\dagger c^\dagger d^\dagger d$ is possible in the vicinity of H_{cl}, but our theory does not claim to be accurate in this region.

If all these forbidden terms are eliminated we arrive at the effective quartic Hamiltonian of the form

$$\mathcal{H}_{IV}^{eff} = N^{-1} \sum_{k_1k_2k_3k_4} \delta(k_1+k_2-k_3-k_4) \times (\phi_{12,34} d_{k_1}^\dagger c_{k_2}^\dagger d_{k_3} c_{k_4} + \psi_{12,34}^a c_{k_1}^\dagger c_{k_2}^\dagger c_{k_3} c_{k_4} + \psi_{12,34}^b d_{k_1}^\dagger d_{k_2}^\dagger d_{k_3} d_{k_4}). \quad (6.1)$$

When $(H^2+E^2)^{1/2}=\frac{1}{2}D+O(J)$ one more channel of interactions turns out to be energetically allowed and has to be added to (6.1), namely

$$\Delta\mathcal{H}_{IV}^{eff} = (4N)^{-1}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2}+H] \times \sum_{k_1k_2k_3k_4} \delta(k_1+k_2+k_3-k_4) \times (c_{k_1}^\dagger c_{k_2}^\dagger c_{k_3}^\dagger d_{k_4} + d_{k_4}^\dagger c_{k_3} c_{k_2} c_{k_1}). \quad (6.2)$$

Consider first the case $(H^2+E^2)^{1/2} \neq \frac{1}{2}D+O(J)$. There are two processes then which can "dissolve" a magnon c_k, previously pumped into the system: (i) annihilation of c_k due to scattering with d-magnons; and (ii) scattering with c magnons only.

Similarly there are two channels in which the d_k magnon can perish: (i′) scattering with c magnons; and (ii′) scattering with d magnons only.

Let us discuss the damping due to (i) first. The transition rate due to this process is

$$(\dot{n}_k^c)_{(1)} = -2\pi N^{-2} \sum_{k_1k_2k_3} |\phi_{12,3k}|^2 \delta(k_1+k_2-k_3-k)\delta(\epsilon_{k_1}^d+\epsilon_{k_2}^c-\epsilon_{k_3}^d-\epsilon_k^c)[n_k^c n_{k_3}^d(n_{k_1}^d+1)(n_{k_2}^c+1)-n_{k_1}^d n_{k_2}^c(n_k^c+1)(n_{k_3}^d+1)]. \quad (6.3)$$

The first term in (6.3) describes the contribution from the process in which magnon c_k is annihilated. The second term is the contribution from the inverse process. Assume now that the occupation numbers $n_{k_3}^d$, $n_{k_1}^d, n_{k_2}^c$ are almost at their equilibrium values: $\bar{n}_{k_3}^d, \bar{n}_{k_1}^d, \bar{n}_{k_2}^c$, respectively, as given by the appropriate Bose factors. This allows us to bring (6.3) to the form

$$(\dot{n}_k^c)_{(1)} = -(n_k^c - \bar{n}_k^c)(\Gamma_k^c)_{(1)}, \quad (6.4)$$

where the summations over the wave-vectors are replaced by integrations over the Brillouin zone, and

$$(\Gamma_k^c)_{(1)} = \pi(2\pi)^{-6}J^{-1}\left(\frac{V}{N}\right)^2 \int_{BZ} d^3k_1 \int_{BZ} d^3k_2 |\phi_{12;k_1+k_2-k,k}|^2 \delta[\gamma(k_1)+\gamma(k_2)-\gamma(k_1+k_2-k)-\gamma(k)] \times[\bar{n}_{k_1+k_2-k}^d(\bar{n}_{k_1}^d+1)+\bar{n}_{k_2}^c(\bar{n}_{k_1+k_2-k}^d-\bar{n}_{k_1}^d)]. \quad (6.5)$$

It seems difficult to calculate the integrals in (6.5) exactly, without employing any numerical methods, since the small-k expansion of γ's is not valid if $k_BT \gg J$. We can however estimate the leading term in the J expansion. If the limit $J \to 0$ is applied to the integrand, the k-dependence of the occupation numbers disappears, whereas $|\phi_{12;k_1+k_2-k,k}|^2$ becomes just a constant. Hence

$$(\Gamma_k^c)_{(1)} \approx 2(2\pi)^{-5}(V/N)^2J^{-1}\mathfrak{N}^8[D(E^2-H^2)+E^2H]^2 \times\bar{n}^d(\bar{n}^d+1)\mathfrak{F}(k), \quad (6.6)$$

with

$$\mathfrak{F}(k) = \int_{BZ} d^3k_1 \int_{BZ} d^3k_2\, \delta(\gamma(k_1)+\gamma(k_2) -\gamma(k_1+k_2-k)-\gamma(k)), \quad (6.7)$$

and $\bar{n}^d$ is defined by Eq. (4.7).

The function $\mathfrak{F}(k)$ is an integral over that subspace of wave vectors k_1 and k_2 for which a collision is allowed. This would be some finite fraction of the Brillouin-zone volume, squared. So in the case of a cubic lattice with lattice constant a, $\mathfrak{F}(k)$ is proportional to $1/a^6$. Thus $V\mathfrak{F}^{1/2}$ is proportional to number of primitive cells. Equation (6.6) can be further simplified by neglecting the square of $\bar{n}^d$.

In a similar way we can analyze the process of damping due to (2) in which the c_k magnons are destroyed. Applying the methods and approximations leading to (6.6) we arrive at the following contribution to the damping constant:

$$(\Gamma_k^c)_{(2)} \approx (2\pi)^{-5}4(V/N)^2J^{-1}\mathfrak{N}^8 \times\{H^2D+\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}-H]\}^2 \times 2\bar{n}^c(\bar{n}^c+1)\mathfrak{F}(k). \quad (6.8)$$

Again $(\bar{n}^c)^2$ is to be neglected. Combining (6.6) with (6.8) we obtain

$$\Gamma_k^c = (\Gamma_k^c)_{(1)} + (\Gamma_k^c)_{(2)} \approx 2(2\pi)^{-5}(V/N)^2J^{-1}\mathfrak{N}^8\mathfrak{F}(k) \times([D(E^2-H^2)+E^2H]^2\bar{n}^d +2\{H^2D+\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}-H]\}^2\bar{n}^c). \quad (6.9)$$

In an analogous way we calculate relaxation via processes (i′) and (ii′) and get

$$\Gamma_k^d \approx 2(2\pi)^{-5}(V/N)^2J^{-1}\mathfrak{N}^8\mathfrak{F}(k) \times([D(E^2-H^2)+E^2H]^2\bar{n}^c +2\{H^2D-\tfrac{1}{2}E^2[(H^2+E^2)^{1/2}+H]\}^2\bar{n}^d). \quad (6.10)$$

At temperatures $Jz \ll k_BT \ll D+(H^2+E^2)^{1/2}$ the $\bar{n}^c$ terms are dominant in the above equations. When $\bar{n}^d$ is neglected we obtain results referred to in Sec. IV [Eq. (4.15)].

The damping of the d magnons significantly increases when $(H^2+E^2)^{1/2}=D/2+O(J)$. Now the d particles can also be relaxed in a splitting process in which three c particles emerge. The transition rate due to splitting is

$$(\dot{n}_k^d)_{\text{splitting}} = -2\pi N^{-2}(3!)^{-1}|\tfrac{6}{4}\mathfrak{N}^4E^2[(H^2+E^2)^{1/2}+H]|^2 \times\sum_{k_1k_2k_3}\delta(k_1+k_2+k_3-k)\delta(\epsilon_{k_1}^c+\epsilon_{k_2}^c+\epsilon_{k_3}^c-\epsilon_k^d) \times[(n_{k_1}^c+1)(n_{k_2}^c+1)(n_{k_3}^c+1)n_k^d -(n_k^d+1)n_{k_1}^c n_{k_2}^c n_{k_3}^c]. \quad (6.11)$$

The factor 1/3! is due to the fact that the change of labels of the three "output" particles does not lead to a different final state. From (6.11) we infer that

$$(\Gamma_k^d)_{\text{splitting}} \approx \tfrac{3}{16}(2\pi)^{-5}(V/N)^2J^{-1}\mathfrak{N}^8E^4 \times[(H^2+E^2)^{1/2}+H]^2\mathfrak{F}'(k)(1+3\bar{n}^c+3\bar{n}^c\bar{n}^c), \quad (6.12)$$

where

$$\mathfrak{F}'(k) = \int_{BZ} d^3k_1 \int_{BZ} d^3k_2\, \delta(\gamma(k_1)+\gamma(k_2) + \gamma(k_1+k_2-k) - \gamma(k)) .$$

In the formula (6.12) there is the large, temperature independent, first term, which drastically increases the value of h_{cr}^{min}.

The confluence process, in which the c-magnons are relaxed, does not contribute to Γ_k^c significantly since it contains terms quadratic in occupation numbers only.

VII. SPECIFIC EXAMPLES AND SOME OTHER SITUATIONS AND SYSTEMS

Materials to which the theory of this paper applies are especially Ni^{++} compounds. These have effective $s=1$, usually have crystal field $D\neq 0$, and frequently have $E\neq 0$.

Paramagnetic resonance studies on five nickel Tutton salts were carried out many years ago by Griffiths and Owen.[17] Typical among these was $Ni(NH_4)_2(SeO_4)_2\cdot 6H_2O$ which has, at 90 K, $D/k = -2.49$ K, $E/k = -1.18$ K, $J/k = +0.018$ K. The value of J was estimated by Stevens[18] from a study of the resonance linewidth as a function of direction of applied field (the direction is necessary to determine the *sign* of J). Stevens concluded that several of the nickel Tutton salts have ferromagnetic coupling of the same magnitude J, which is comparable to the dipole-dipole coupling; there may also be anisotropic exchange. The nickel Tutton salts all have negative D (which is however quite temperature dependent) and therefore they lie in region L.

Other salts in region L are α-$NiSO_4\cdot 6H_2O$, studied by Stout and Hadley[19]; and $NiZrF_6\cdot 6H_2O$, studied by Karnezos, Meier, and Friedberg.[20] Both of these materials appear to have $E=0$.

The salt $NiSnCl_6\cdot 6H_2O$, studied by How and Svare[21] and by Friedberg and co-workers,[22] has, at 4.2 K, $D/k = +0.65$ K, $|E|/k \lesssim 0.07$ K, and hence would be in region S, Fig. 1(a), except that its exchange coupling is negative ($J/k \sim -0.02$ K), i.e., antiferromagnetic. A spin-wave theory appropriate to antiferromagnetic exchange would require division of the spins into two sublattices.

In the last named salt the sign of D changes to negative above 338 K. Walsh[23] has demonstrated, with measurements on $NiSiF_6\cdot 6H_2O$, that hydrostatic pressure of ~6000 kg/cm^2 can change D from negative to positive and also (in two dilute Cr salts[24]) can increase the value of E. The nickel fluosilicate has ferromagnetic exchange.[18,25]

The salt $NiCl_2\cdot 6H_2O$ has $D/k = -1.73$ K, $E/k \sim +0.22$ K, and sufficiently strong antiferromagnetic J ($2zJ/k = -12.4$ K) to satisfy Moriya's criterion for long-range order[8] and to become antiferromagnetic below $T_N = 5.34$ K. The measurements are by Date and Motokawa[26] using antiferromagnetic resonance. They note that J/D is so large that the three paramagnetic resonance lines at 70 K are mixed and merge into a single unresolved line of 7 kOe in breadth. But of course our theory has broken down long before this value of J/D is reached.

The theoretical methods of the preceding sections of this paper have also been applied to the following situations: (i) spin-one systems with uniaxial D and orthorhombic E anisotropies plus weak ferromagnetic exchange, and with the static magnetic field applied perpendicular to the uniaxis; and to (ii) spin-two systems with single-ion cubic anisotropy plus weak ferromagnetic exchange. In this case, to match the five eigenstates of the single-ion Hamiltonian, it is necessary to use four sets of Bose operators. Details of these calculations have been given elsewhere.[27]

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