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SPIN WAVES AND THE PHASE TRANSITION BETWEEN ANTIFERROMAGNETIC PHASE AND SPIN FLOP PHASE

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ABSTRACT

The phase transition between antiferromagnetic phase and spin flop phase for a uniaxial antiferromagnet when the magnetic field is applied in a parallel direction to the easy axis of magnetization is investigated theoretically by means of two different methods of approximation. The one is the harmonic approximation based upon the use of Holstein-Primakoff operators. The ground state energies and the dynamical behaviours of both phases are studied and three kinds of critical fields for the transition are obtained. The physical meaning for them and the relation between them are considered approximately. The other method of approximation based upon the coupled boson treatment is investigated in order to gain the more refined values for them taking into account the effects of dynamical and kinematical interactions between spin waves properly. Thus we have obtained the next approximation for the strength of the critical fields.

§1. Introduction

We consider an antiferromagnet in the Heisenberg model with uniaxial anisotropic energies; i.e., for example RbMnF_2 , MnF_2 , Cr_2O_3 , and $\text{MnBr}_2 \cdot 4\text{H}_2\text{O}$. (J. H. SCHELLENG and S. FRIEDBERG 1963, and I. TSUJIKAWA and J. KANDA 1959) These materials have been treated by many investigators experimentally and theoretically, and are known that they show various different phases according to the strength and direction of externally applied magnetic field at sufficiently low temperatures.

The ground state in various ordered states is investigated by means of two different approaches, when the magnetic field is applied in a parallel direction to

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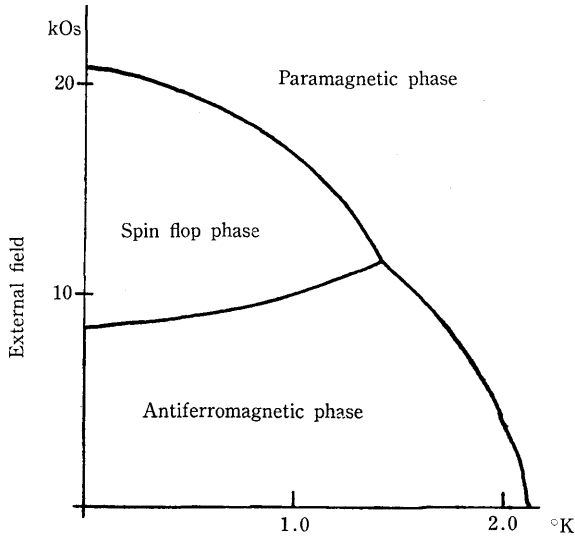


Fig. 1. Phase diagram in $\text{MnBr}_2 \cdot 4\text{H}_2\text{O}$. External field is applied along the parallel direction to the easy axis of magnetization for anisotropy.

the easy axis of magnetization for anisotropy. In this situation there have been found to be three phases; antiferromagnetic phase, spin flop phase and paramagnetic phase, which are shown schematically in Fig. 1. The spins of magnetic ions which have spin quantum number S are considered to interact each other through the negative exchange interaction between nearest neighbours. The crystal structure of magnetic ions is assumed to consist of two interpenetrating sublattices, each of which has $N/2$ magnetic ions respectively. In the first order phase transition between spin flop phase and antiferromagnetic phase, two metastable state can exist analogously as those in a gas-liquid transition. This may be investigated from the two different points of view, in one of which the system is assumed to be in the true lowest energy state and the dependence upon the magnetic field of the ground state energy is studied, and thus we can therefore obtain a value for the critical field between antiferromagnetic and spin flop phases. On the other hand, we may treat the dynamical behaviour of the system, the stability conditions of which for the ordered states give two different values for the critical field strength in the phase transition. The transition from antiferromagnetic phase to spin flop phase is denoted here by anti-flop transition and that from spin flop phase to antiferromagnetic phase by flop-anti transition.

For the former case, an analysis was carried out by F. B. ANDERSON and H. B. CALLEN at a finite temperature. Their method was a decoupling procedure for the equation of motion of the Green function. For the latter case, a standard spin-wave analysis was carried out by WANG and CALLEN. We also worked out an analysis in the harmonic approximation, and obtained the same results. But these spin wave analysis is not suited to take in an effect of kinematical interaction.

This effect is not negligible under the condition of small S . We discuss a usefulness of a coupled boson representation for spin system, and show how the

interaction affects the solution. Our calculation technique is a method of a Green function of coupled boson operators which was developed by WANG and CALLEN, and the results are restricted to zero temperature cases. Refined values for the excitation energy of a magnon and critical values of magnetic field at flop-anti and anti-flop transition are obtained.

§ 2. Model and harmonic approximation

The Hamiltonian for our antiferromagnet is described as

$$\mathcal{H} = \sum_{i,\alpha} 2J_{i\alpha} \mathbf{S}_i \cdot \mathbf{S}_\alpha - D \left\{ \sum_i (S_i^z)^2 + \sum_\alpha (S_\alpha^z)^2 \right\} - \beta H \left\{ \sum_i S_i^z + \sum_\alpha S_\alpha^z \right\}, \quad (1)$$

where $J_{i\alpha}$ is the absolute value for the exchange interaction between nearest neighbour pairs, H is the strength of applied magnetic field along the z -direction, β is a unit of magnetic dipole moment for an ion, i. e., $g\mu_B$, and D is an anisotropy constant for an ion in the crystal with the z -axis as the direction of easy magnetization. At absolute zero of temperature the ordered state in each phase can be represented in terms of directions of spins in two sublattices. In antiferromagnetic phase spins are oppositely directed each other along the z -direction, and in spin flop phase spins are nearly in an $x-y$ plane oppositely directed each other inclined from the plane to the z -axis with some amount of angle θ .

As for antiferromagnetic phase we take the z -axis as the direction for quantization, and represent spin operators in terms of Holstein-Primakoff operators A_j and B_α . Introducing the Fourier transformation into \mathbf{k} -space for these quantities,

$$A_j = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} a_{\mathbf{k}} \exp(-i\mathbf{k} \cdot \mathbf{R}_j),$$

$$B_\alpha = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} b_{\mathbf{k}} \exp(+i\mathbf{k} \cdot \mathbf{R}_\alpha),$$

and

$$J_{\mathbf{k}} = \sum_\alpha J_{j\alpha} \exp i\mathbf{k} \cdot (\mathbf{R}_j - \mathbf{R}_\alpha), \quad (2)$$

where A_j and B_α are destruction operators referring to j -sublattice site and α -sublattice site respectively, we obtain the following Hamiltonian in harmonic approximation neglecting the fourth order and higher terms for destruction and creation operators,

$$\mathcal{H} = E_{j\parallel}^0 + \sum_{\mathbf{k}} \{ (\xi_{j\parallel} + \beta H) a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + (\xi_{j\parallel} - \beta H) b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \eta_{j\parallel\mathbf{k}} (a_{\mathbf{k}}^\dagger b_{\mathbf{k}}^\dagger + a_{\mathbf{k}} b_{\mathbf{k}}) \}, \quad (3)$$

where we have used abbreviations;

$$E_{j\parallel}^0 = -NS^2(J_0 + D),$$

$$\xi_{j\parallel} = 2S(J_0 + D), \quad \text{and} \quad \eta_{j\parallel\mathbf{k}} = 2SJ_{\mathbf{k}}. \quad (4)$$

The above Hamiltonian (3) can be diagonalized by Bogolyubov transformation into the form;

$$\mathcal{H} = E_{//}^0 + \Delta E_{//}^0 + \sum_{\mathbf{k}} (\epsilon_{\mathbf{k}}^d \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} + \epsilon_{\mathbf{k}}^u \beta_{\mathbf{k}}^\dagger \beta_{\mathbf{k}}), \quad (5)$$

where $\epsilon_{\mathbf{k}}^u$ and $\epsilon_{\mathbf{k}}^d$ represent the excitation energies for spin waves with wave vector \mathbf{k} , and are obtained from positive solutions of the following equation;

$$\begin{vmatrix} \xi_{//} + \beta H - \epsilon, & \eta_{//\mathbf{k}}, & 0, & 0, \\ \eta_{//} & \xi_{//} - \beta H + \epsilon, & 0, & 0, \\ 0, & 0, & \xi_{//} - \beta H - \epsilon, & \eta_{//}, \\ 0, & 0, & \eta_{//\mathbf{k}}, & \xi_{//} + \beta H + \epsilon. \end{vmatrix} = 0. \quad (6)$$

The zero-point energy of the system is denoted as $\Delta E_{//}^0$;

$$\Delta E_{//}^0 = \frac{1}{2} \sum_{\mathbf{k}} (\epsilon_{\mathbf{k}}^u + \epsilon_{\mathbf{k}}^d - 2\xi_{//}). \quad (7)$$

The ground state energy in antiferromagnetic phase, i.e., $E_{//}^0 + \Delta E_{//}^0$ is therefore expressed as follows,

$$E_{//}^0 + \Delta E_{//}^0 = -NS(S+1)(J_0 + D) + \frac{1}{2} \sum_{\mathbf{k}} (\epsilon_{\mathbf{k}}^u + \epsilon_{\mathbf{k}}^d). \quad (8)$$

Increasing the strength of field we notice that the minimum excitation energy of a magnon, $\epsilon_{\mathbf{k}}^d$ may become zero at the critical field $H_f^{(+)}$ given by

$$\beta H_f^{(+)} = 2S\{D(2J_0 + D)\}^{\frac{1}{2}}. \quad (9)$$

This can be considered the critical field for anti-flop transition.

Next we proceed to treat spin flop phase in which the directions of quantization are set up as in Fig. 2. The axes of quantization, which we denote as ζ and

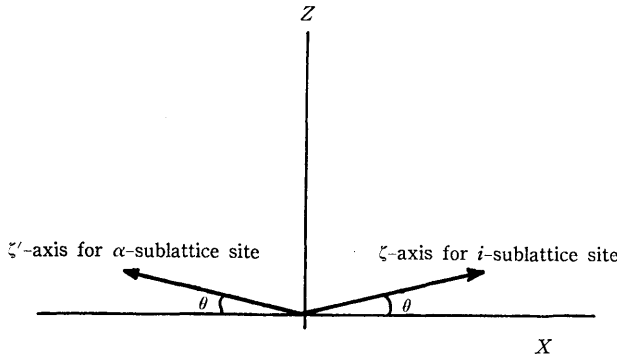


Fig. 2. The spin configuration in spin flop phase. The axis of quantization is denoted as ζ -axis (or ζ' -axis) for i -sublattice site (or α -sublattice site) respectively.

ζ' axes respectively, are defined in terms of the angle θ . Raising and lowering operators may be defined in order to get the right hand system for ζ and ζ' axes respectively. Our Hamiltonian, in terms of Holstein-Primakoff operators in coordinate space previously mentioned, may be written in harmonic approximation and in \mathbf{k} -space representation,

$$\begin{aligned} \mathcal{H} = & E_{\perp}^0 + \Delta E_{\perp}^0 + \zeta(a_0 + b_0 + a_0^+ + b_0^+) \\ & + \sum_{\mathbf{k}} \{ \xi(a_{\mathbf{k}}^+ a_{\mathbf{k}} + b_{\mathbf{k}}^+ b_{\mathbf{k}}) + \eta_{\mathbf{k}}(a_{\mathbf{k}} b_{\mathbf{k}} + a_{\mathbf{k}}^+ b_{\mathbf{k}}^+) + \lambda_{\mathbf{k}}(a_{\mathbf{k}}^+ b_{-\mathbf{k}} + a_{\mathbf{k}} b_{-\mathbf{k}}^+) \\ & + \Gamma(a_{\mathbf{k}} a_{-\mathbf{k}} + b_{\mathbf{k}} b_{-\mathbf{k}} + a_{\mathbf{k}}^+ a_{-\mathbf{k}}^+ + b_{\mathbf{k}}^+ b_{-\mathbf{k}}^+) \}, \end{aligned} \quad (10)$$

where we have used abbreviations for spin flop phase,

$$\begin{aligned} \zeta &= -\frac{1}{2} \sqrt{NS} \{ \beta H \cos \theta - S(2J_0 - D) \sin 2\theta \}, \\ \xi &= \beta H \sin \theta + 2S \left\{ J_0 \cos 2\theta + D \left(\sin^2 \theta - \frac{1}{2} \cos^2 \theta \right) \right\}, \\ \eta_{\mathbf{k}} &= SJ_{\mathbf{k}}(\cos 2\theta + 1), \quad \lambda_{\mathbf{k}} = SJ_{\mathbf{k}}(\cos 2\theta - 1), \quad \Gamma = -\frac{1}{2} SD \cos^2 \theta, \\ E_{\perp}^0 &= -NS \{ \beta H \sin \theta + S(J_0 \cos 2\theta + D \sin^2 \theta) \}, \end{aligned}$$

and

$$\Delta E_{\perp}^0 = -\frac{1}{2} NSD \cos^2 \theta. \quad (11)$$

The angle θ is determined from the requirement that classical energy E_{\perp}^0 attains the minimum value for that angle, and thus we may find that the coefficient of linear terms ζ vanishes automatically. Remaining Hamiltonian can be diagonalized by Bogolyubov transformation to get the result;

$$\mathcal{H} = E_{\perp}^0 + \Delta E_{\perp}^0 + \frac{1}{2} \sum_{\mathbf{k}} \{ \epsilon_{\mathbf{k}}^{(1)} + \epsilon_{\mathbf{k}}^{(2)} - 2\xi \} + \sum_{\mathbf{k}} \{ \epsilon_{\mathbf{k}}^{(1)} \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} + \epsilon_{\mathbf{k}}^{(2)} \beta_{\mathbf{k}}^+ \beta_{\mathbf{k}} \}, \quad (12)$$

whence $\epsilon_{\mathbf{k}}^{(1)}$ and $\epsilon_{\mathbf{k}}^{(2)}$ are obtained as the positive roots from the following equation;

$$\begin{vmatrix} \xi - \epsilon_{\mathbf{k}}, & \lambda_{\mathbf{k}}, & 2\Gamma, & \eta_{\mathbf{k}}, \\ \lambda_{\mathbf{k}}, & \xi - \epsilon_{\mathbf{k}}, & \eta_{\mathbf{k}}, & 2\Gamma, \\ 2\Gamma, & \eta_{\mathbf{k}}, & \xi + \epsilon_{\mathbf{k}}, & \lambda_{\mathbf{k}}, \\ \eta_{\mathbf{k}}, & 2\Gamma, & \lambda_{\mathbf{k}}, & \xi + \epsilon_{\mathbf{k}}, \end{vmatrix} = 0. \quad (13)$$

Here we have obtained the two kinds of magnon excitation energies;

$$\begin{aligned} \epsilon_{\mathbf{k}}^{(1)} &= 2S \left[(J_0 - J_{\mathbf{k}}) \left\{ J_0 + J_{\mathbf{k}} - D - \frac{(2J_{\mathbf{k}} - D)(\beta H)^2}{(2S)^2(2J_0 - D)^2} \right\} \right]^{\frac{1}{2}}, \\ \epsilon_{\mathbf{k}}^{(2)} &= 2S \left[(J_0 + J_{\mathbf{k}}) \left\{ J_0 - J_{\mathbf{k}} - D + \frac{(2J_{\mathbf{k}} + D)(\beta H)^2}{(2S)^2(2J_0 - D)^2} \right\} \right]^{\frac{1}{2}}. \end{aligned} \quad (13')$$

$\epsilon_k^{(1)}$ is always found to become to zero when \mathbf{k} tends to zero. This mode of excitation satisfies the Goldstone theorem. The minimum excitation energy for the mode $\epsilon_k^{(2)}$ becomes zero at the critical field $H_f^{(-)}$ when the field is weakened,

$$\beta H_f^{(-)} = 2S(2J_0 - D) \frac{D^{\frac{1}{2}}}{(2J_0 + D)^{\frac{1}{2}}}. \quad (14)$$

The above field is expected to be the critical one for flop-anti transition.

We may obtain the third value for critical field requiring that the system should be in the true lowest energy state, leading to the condition

$$E_{//}^0 + \Delta E_{//} = E_{\perp}^0 + \Delta E_{\perp} + \frac{1}{2} \sum_{\mathbf{k}} \{ \epsilon_k^{(1)} + \epsilon_k^{(2)} - 2\xi \} \quad (15)$$

at the field. Performing the \mathbf{k} -sum in the equation (15) approximately upon condition that the anisotropy constant D is much smaller than J_k , we have obtained the third value;

$$\beta H_f^{(0)} = 2\sqrt{S(S-\delta)D(2J_0-D)},$$

where

$$\delta = \frac{2}{N} \sum_{\mathbf{k}} \left(1 - \frac{J_k^2}{J_0^2} \right)^{-\frac{1}{2}} + \frac{1}{N} \sum_{\mathbf{k}} \left(1 - \frac{J_k^2}{J_0^2} \right)^{\frac{1}{2}} - 1. \quad (16)$$

The phase transition is the first order one in which metastable state may exist. The two fields, $H_f^{(+)}$ and $H_f^{(-)}$ may be interpreted as the limiting one for the meta-stable spin flop phase and antiferromagnetic phase respectively, and the following relation may be valid,

$$H_f^{(-)} < H_f^{(0)} < H_f^{(+)}, \quad (17)$$

which we may confirm approximately in weak anisotropy case, using the approximate expressions (9), (14) and (16). These expressions however have the unsatisfactory feature in some respects. In the case $S=1/2$, we may expect that the effect of anisotropy should vanish, which is not yet satisfied in the equations (9), (14) and (16). The expression (16) contains the quantity δ , the value of which is found to be approximately equal to 0.5 in a three dimensional simple cubic lattice, and therefore δ may represent a kind of quantum effect of spin contraction, we suppose. Energy of a magnon may be expanded by powers of $(zS)^{-1}$, although we have treated the first order approximation (order S^1 for the equations (9) and (14)), and the next approximation should therefore treat the contribution of order S^0 properly.

§ 3. Coupled boson treatment

In order to consider the effect of dynamical and kinematical interactions properly and proceed into the next approximation for the results obtained in § 2, we

make use of the coupled boson approach which has the advantage that the Hamiltonian can be expressed by products of finite number of boson operators. Spin operators, which are defined along the suitable ζ -axis for quantization, can be represented as follows using two kinds of independent boson operators u 's and v 's.

$$\begin{aligned} S_j^+ &= u_j^+ v_j, & S_j^- &= v_j^+ u_j, \\ S_j^z &= \frac{1}{2}(u_j^+ u_j - v_j^+ v_j) = \hat{S}_j - v_j^+ v_j, \\ [u_j, u_j^+] &= [v_j, v_j^+] = \delta_{j,j'}, \end{aligned}$$

and all other commutators vanish. (18)

Here, \hat{S}_j is an operator which defines the magnitude for each spin vector;

$$\hat{S}_j = \frac{1}{2}(u_j^+ u_j + v_j^+ v_j). \quad (19)$$

Since the magnitude of a spin for each lattice point is definite, namely is equal to S , the constancy for the equation (19) describes the effect of kinematical interaction between spin waves. The excitations for v -particles may be interpreted as the deviations from the configuration of the classical spin vectors since S_j^z is equal to $\hat{S}_j - v_j^+ v_j$. In our antiferromagnet which consists of two magnetic sublattices, we must have two kinds of axes for quantization. We use the following state vector which corresponds to the ordered spin state along ζ and ζ' axes,

$$|0\rangle = \prod_{i,\alpha} \{(2S)!\}^{-1} (u_i^+)^{2S} (u_\alpha^+)^{2S} |0\rangle, \quad (20)$$

in which the state $|0\rangle$ represents the true vacuum state for u - and v -particles, and we use subscripts i, j, k, \dots for lattice points in ζ -sublattice and $\alpha, \beta, \gamma, \dots$ for lattice points in ζ' -sublattice.

For the antiferromagnetic phase, the Hamiltonian is expressed in our coupled boson representation,

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}',$$

$$\mathcal{H}_0 = -NS^2(J_0 + D) + \omega_d \sum_i v_i^+ v_i + \omega_u \sum_\alpha v_\alpha^+ v_\alpha,$$

$$\omega_d = 2S(J_0 + D) + \beta H = \xi_{//} + \beta H, \quad \omega_u = 2S(J_0 + D) - \beta H = \xi_{//} - \beta H,$$

and

$$\begin{aligned} \mathcal{H}' &= \sum_{i,\alpha} \{ J_{i\alpha} (v_i^+ v_\alpha^+ u_i u_\alpha + v_i v_\alpha u_i^+ u_\alpha^+) - 2J_{i\alpha} v_i^+ v_i v_\alpha^+ v_\alpha \} \\ &\quad - D \{ \sum_i (v_i^+ v_i)^2 + \sum_\alpha (v_\alpha^+ v_\alpha)^2 \}. \end{aligned} \quad (21)$$

In order to investigate elementary excitations for this spin system we take up the following Green's function,

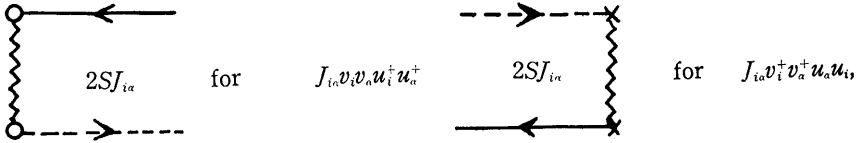
$$\begin{aligned} G(j, m) &= -i \langle 0 | P \tilde{S}_j^+(t_j) \tilde{S}_m^-(t_m) | 0 \rangle \\ &= -i \langle 0 | P v_j(t_j) v_m^+(t_m) u_j^+(t_j) u_m(t_m) S(\infty) | 0 \rangle / \langle 0 | S(\infty) | 0 \rangle. \end{aligned} \quad (22)$$

In the first expression, $\tilde{S}_j^+(t_j)$ is an operator in Heisenberg representation and the state $|0\rangle$ denotes the true ground state, and in the second expression each v and u

operator is written in the interaction representation. We have used the method of the connected diagram expansion and the lock-diagram convention developed by Wang et al.^{4),5)}

Each term in the interaction Hamiltonian \mathcal{H}' has the following order of magnitudes. The first and second terms which are called transverse interaction have the order S^1 due to the presence of products of two u operators. The third term has the order S^0 which has no u operators and is called longitudinal interaction. The last terms, i.e., the anisotropy energy parts may be renormalized into the harmonic part as we shall show later.

First we consider the interaction of order S^1 . In this approximation all u operators are replaced by $(2S)^{\frac{1}{2}}$ and only the transverse interaction is considered. For this interaction, the structure of each vertex is represented by



in which the real (or dotted) line denotes the free Green's function for i -lattice site (or for α -lattice site). The chain diagram is constructed by using these two points vertices only. In order to sum up contributions of all diagrams we may use Fourier transformation for space and time for both of the Green's function and the exchange interaction,

$$G(\mathbf{k}, \omega) = \frac{2}{N} \sum_l \int_{-\infty}^{+\infty} dt_l G(l, m) \exp i\{\omega(t_l - t_m) - \mathbf{k} \cdot (\mathbf{R}_l - \mathbf{R}_m)\},$$

and $J_{\mathbf{k}} = \sum J_{j\alpha} \exp i\mathbf{k} \cdot (\mathbf{R}_j - \mathbf{R}_\alpha).$ (23)

Thus each diagram corresponds to the simple products for these elements. For free Green's function we obtain;

$$G^0(\omega) = (\omega - \omega_d + i\delta)^{-1}; \quad \text{---} \leftarrow \text{---} \quad \delta \rightarrow +0.$$

$$G^0(\omega) = (\omega - \omega_u + i\delta)^{-1}; \quad \text{---} \leftarrow \text{---} \quad \delta \rightarrow +0. \quad (24)$$

Introducing new propagators for the sums, all diagrams in chain diagram approximation are reproduced from a Dyson equation given by Y.L. WANG et al. The poles for the propagator are determined by an equation which coincides with the equation (6) exactly. Matrix elements in the determinant are proportional to S^1 . Thus we may conclude that our next task is to evaluate each element correctly up to the order S^0 at least. The lock-diagram convention due to WANG et al. is suitable for this purpose.

The anisotropy energy part in the interaction Hamiltonian (21) must be considered in this point of view. Rewriting the interaction into the form,

$$(v_i^\dagger v_i)^2 = v_i^\dagger v_i (2S) - v_i^\dagger u_i v_i u_i^\dagger + v_i^\dagger v_i, \quad (25)$$

we may note that the first two terms cancel out in the diagram without overlapping and the remaining last term can be renormalized into the harmonic Hamiltonian. Thus the effect of renormalization is expressed,

$$\begin{aligned} \omega_d &\longrightarrow \omega'_d = 2S(J_0 + D\lambda) + \beta H, \\ \text{and} \quad \omega_u &\longrightarrow \omega'_u = 2S(J_0 + D\lambda) - \beta H, \end{aligned} \quad (26)$$

where we introduced the notation $\lambda = 1 - (2S)^{-1}$. The diagram with overlapping part must be considered in the approximation up to the order S^0 . Following the idea of WANG et al. we have to proceed to consider the effect of the diagrams with one locking for the transverse and longitudinal interaction. Some quantities are defined as follows. The spin contraction δS^c is defined;

$$\begin{aligned} \delta S^c &= S - \langle 0 | S^c | 0 \rangle \\ &= i(2S)^{-1} \lim_{t_m \rightarrow t_l + 0} 2N^{-1} \sum_k \int \frac{d\omega}{2\pi} G(k, \omega) \exp \{-i\omega(t_l - t_m)\}, \end{aligned} \quad (27)$$

Inserting the approximate chain Green's function G'^c with the effect of renormalization (26) for the definition (27), we get

$$\delta S^c = N^{-1} \sum_k \left[\frac{\omega'_d + \omega'_u}{\omega_k^{(1)} + \omega_k^{(2)}} \right] - 1, \quad (28)$$

where $\omega_k^{(1)}$ and $\omega_k^{(2)}$ are positive roots for the equation (6) with substitution (26). Another quantity is also defined;

$$J = N^{-1} \sum_k \left[1 - \frac{\omega_k^{(1)} + \omega_k^{(2)}}{\omega'_d + \omega'_u} \right]. \quad (29)$$

The new vertices in the Wang's approximation with one locking are expressed in terms of the above defined quantities.

We used the chain Green's function G'^c or g'^c instead of free Green's function G^0 or g^0 since the vertices may have any arbitrary size. Each renormalized interaction has the order S^0 and therefore corresponds just to the correction term which we wanted to attain. We have investigated the effects of locking for diagrams with many overlaps and have concluded that the effect of anisotropy Hamiltonian can be renormalized into the harmonic part with substitution (26) in the one locking approximation at least, although the procedure (26) may not be sufficient for the higher approximation of order S^{-1} and so on. Including the effects of all vertices, we can construct the Dyson equations. The poles for the renormalized Green functions are obtained from a determinant equation, that the equation (6) is similar to. Each element of the determinant has the corrected value which may be supposed to be valid up to the order S^0 . Although we have used ξ' for computation of the self-energy terms, the approximation up to order S^0 may be obtained from using ξ instead of ξ' , ξ' may be preferred rather than ξ because ξ' is able to satisfy the physical requirement that the effect of anisotropy should vanish when S is equal to $1/2$. The energies of a magnon are obtained from the positive roots of the determinant equation;

$$\omega_k = \pm \beta H + 2S\{(\tilde{J}_0 + D\lambda)^2 - \tilde{J}_k^2\}^{1/2}, \quad (30)$$

where the modified interaction \tilde{J}_k is introduced by

$$\tilde{J}_k = J_k[1 + S^{-1}\{J + J_0^{-1}D\lambda(\delta S^z + J)\}]. \quad (31)$$

The corrected value of the field for anti-flop transition becomes

$$\beta H_f^{(+)} = 2S(D\lambda)^{1/2}(2\tilde{J}_0 + D\lambda)^{1/2}. \quad (32)$$

Next we consider the spin flop phase. The Hamiltonian in coupled boson representation consists of the harmonic part, the linear part for v operators, and the interaction part;

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_0 + \delta\mathcal{H} + \mathcal{H}_I, \\ \mathcal{H}_0 &= E_{\perp}^0 + JE_{\perp}^0 + \omega_0(\sum_i v_i^{\dagger}v_i + \sum_{\alpha} v_{\alpha}^{\dagger}v_{\alpha}), \\ \omega_0 &= 2S(J_0 \cos 2\theta + D \sin^2 \theta) + \beta H \sin \theta, \\ \delta\mathcal{H} &= \cos \theta \left(2SJ_0 \sin \theta - SD\lambda \sin \theta - \frac{1}{2}\beta H \right) \left(\sum_i F_i + \sum_{\alpha} F_{\alpha} \right), \\ F_i &= v_i u_i^{\dagger} + v_i^{\dagger} u_i, \quad F_{\alpha} = v_{\alpha} u_{\alpha}^{\dagger} + v_{\alpha}^{\dagger} u_{\alpha}, \\ \mathcal{H}_I &= \sum_{i,\alpha} 2J_{i\alpha} \left[\frac{1}{2} \cos^2 \theta (v_i v_{\alpha} u_i^{\dagger} u_{\alpha}^{\dagger} + v_i^{\dagger} v_{\alpha}^{\dagger} u_i u_{\alpha}) \right. \\ &\quad - \frac{1}{2} \sin^2 \theta (v_i v_{\alpha}^{\dagger} u_i^{\dagger} u_{\alpha} + v_i^{\dagger} v_{\alpha} u_i u_{\alpha}^{\dagger}) \\ &\quad \left. - \cos 2\theta (v_i^{\dagger} v_i v_{\alpha}^{\dagger} v_{\alpha}) - \cos \theta \sin \theta (v_i^{\dagger} v_i (v_{\alpha} u_{\alpha}^{\dagger} + v_{\alpha}^{\dagger} u_{\alpha}) + v_{\alpha}^{\dagger} v_{\alpha} (v_i u_i^{\dagger} + v_i^{\dagger} u_i)) \right] \\ &\quad - \frac{1}{2} D \cos^2 \theta \left[\sum_i v_i^{\dagger} v_i u_i^{\dagger} u_i + \sum_{\alpha} v_{\alpha}^{\dagger} v_{\alpha} u_{\alpha}^{\dagger} u_{\alpha} \right] \\ &\quad - \frac{1}{4} D \cos^2 \theta \left[\sum_i \{(v_i u_i^{\dagger})^2 + (v_i^{\dagger} u_i)^2\} + \sum_{\alpha} \{(v_{\alpha} u_{\alpha}^{\dagger})^2 + (v_{\alpha}^{\dagger} u_{\alpha})^2\} \right] \\ &\quad - D \sin^2 \theta \left[\sum_i (v_i^{\dagger} v_i)^2 + \sum_{\alpha} (v_{\alpha}^{\dagger} v_{\alpha})^2 \right] \\ &\quad + D \sin \theta \cos \theta \left[\sum_i \{v_i^{\dagger} v_i^2 u_i^{\dagger} + v_i^{\dagger} v_i^{\dagger} v_i u_i\} + \sum_{\alpha} \{v_{\alpha}^{\dagger} v_{\alpha}^2 u_{\alpha}^{\dagger} + v_{\alpha}^{\dagger} v_{\alpha}^{\dagger} v_{\alpha} u_{\alpha}\} \right]. \end{aligned} \quad (33)$$

The angle θ can be determined from the requirement that $E_{\perp}^0 + JE_{\perp}^0$ should attain minimum value, from which we obtain

$$\beta H = 2S(2J_0 - D\lambda) \sin \theta', \quad (34)$$

and at this angle θ' , $\delta\mathcal{H}$ vanishes simultaneously. We note that θ' is different from θ in Holstein-Primakoff treatment, but this angle θ' satisfies the physical requirement that the effect of D should vanish when S is equal to $1/2$. We may therefore expect θ' is the next approximation for θ . In chain diagram approxima-

tion, we neglect in the interaction, the linear term for u -operator and the longitudinal part, and consider only the quadratic terms for u -operators. Taking care of the order of magnitude of u -operators for our chain diagram approximation, each u -operator should be replaced by $(2S)^{1/2}$ when the u -operators are arranged into correct order with respect to corresponding v -operators, thus we may have the following effective interaction Hamiltonian,

$$\begin{aligned} \mathcal{H}' = & -SD\lambda \cos^2 \theta' (\sum_i v_i^+ v_i + \sum_\alpha v_\alpha^+ v_\alpha) \\ & + 2S \sum_{i,\alpha} J_{i\alpha} [\cos^2 \theta' (v_i v_\alpha + v_i^+ v_\alpha^+) - \sin^2 \theta' (v_i v_\alpha^+ + v_i^+ v_\alpha)] \\ & - \frac{1}{2} SD\lambda \cos^2 \theta' [\sum_i (v_i^2 + v_i^{+2}) + \sum_\alpha (v_\alpha^2 + v_\alpha^{+2})]. \end{aligned} \quad (35)$$

The first term in (35) can be renormalized into harmonic part,

$$\begin{aligned} \omega_0 \rightarrow \omega'_0 = & \omega_0 - SD\lambda \cos^2 \theta' \\ = & 2S \left[J_0 \cos 2\theta' + D\lambda \left(\sin^2 \theta' - \frac{1}{2} \cos^2 \theta' \right) \right] + \beta H \sin \theta'. \end{aligned} \quad (36)$$

When we evaluate the last terms in (35) as the values of vertices we must multiply them by two because of two possible ways of connection for external lines, thus we may obtain the value for each vertex with the quantities introduced previously. Dyson equations can be written down as follows in symbolically,

$$\begin{aligned} G^c; \Rightarrow \Leftarrow & = \Leftarrow \Leftarrow + \begin{array}{c} \bullet \\ \leftarrow \\ \text{wavy } \lambda_k \\ \bullet \\ \Leftarrow \end{array} + \begin{array}{c} \bullet \\ \leftarrow \\ \text{double } 2\Gamma \\ \bullet \\ \Leftarrow \end{array} + \begin{array}{c} \bullet \\ \leftarrow \\ \text{wavy } \gamma_k \\ \bullet \\ \Leftarrow \end{array} \\ G^1; \Rightarrow \Leftarrow & = \begin{array}{c} \bullet \\ \leftarrow \\ \text{wavy } \lambda_k \\ \bullet \\ \Leftarrow \end{array} + \begin{array}{c} \bullet \\ \leftarrow \\ \text{wavy } \gamma_k \\ \bullet \\ \Leftarrow \end{array} + \begin{array}{c} \bullet \\ \leftarrow \\ \text{double } 2\Gamma \\ \bullet \\ \Leftarrow \end{array} \\ G^2; \Rightarrow \Rightarrow & = \Rightarrow \Rightarrow + \begin{array}{c} \bullet \\ \Rightarrow \\ \text{wavy } \gamma_k \\ \bullet \\ \Rightarrow \end{array} + \begin{array}{c} \bullet \\ \Rightarrow \\ \text{wavy } \lambda_k \\ \bullet \\ \Rightarrow \end{array} \\ G^3; \Rightarrow \Leftarrow & = \Leftarrow \Leftarrow + \begin{array}{c} \bullet \\ \leftarrow \\ \text{wavy } \gamma_k \\ \bullet \\ \Leftarrow \end{array} + \begin{array}{c} \bullet \\ \leftarrow \\ \text{double } 2\Gamma \\ \bullet \\ \Leftarrow \end{array} + \begin{array}{c} \bullet \\ \leftarrow \\ \text{wavy } \lambda_k \\ \bullet \\ \Leftarrow \end{array} \end{aligned} \quad (37)$$

They are able to be written in matrix form, noting that G^0 is equal to g^0 in spin flop phase. The poles for $G^0(\omega)$ are obtained from an equation which should be compared with the corresponding equation (13). Each element has the same term of order S^1 as in the harmonic approximation, but in that equation it has the terms of order S^0 due to the difference between θ' and θ and further due to the result of renormalization (36). The excitation energies for a magnon is obtained from the positive roots of that equation ;

$$\begin{aligned}\omega_k^{(1)} &= 2S \left[(J_0 - J_k) \left\{ (J_0 + J_k - D\lambda) - (2J_k - D\lambda) \frac{(\beta H)^2}{\{2S(2J_0 - D\lambda)\}^2} \right\} \right]^{\frac{1}{2}} \\ \omega_k^{(2)} &= 2S \left[(J_0 + J_k) \left\{ (J_0 - J_k - D\lambda) - (2J_k + D\lambda) \frac{(\beta H)^2}{\{2S(2J_0 - D\lambda)\}^2} \right\} \right]^{\frac{1}{2}}\end{aligned}\quad (38)$$

Thus we have obtained the field for flop-anti transition from $\omega_k^{(2)}$,

$$\beta H_f^{(\leftarrow)} = 2S(2J_0 - D\lambda) \left\{ \frac{D\lambda}{2J_0 + D\lambda} \right\}^{\frac{1}{2}}. \quad (39)$$

§ 4. Discussion

We have considered the phase transition for the anisotropic antiferromagnet and obtained three kinds of critical fields for the transition. Our results may be expected to satisfy the condition (17) even in higher approximation (32) and (39) if the spin quantum number S is large. When S tends to $1/2$ there remains some uncertainty, although we have obtained the desired expressions (30) and (38) for the excitation energies for a magnon. These excitation energies have no dependence upon D when S is equal to $1/2$ as it should. The main reason is expected because the effects of anisotropy can be renormalized into the harmonic part according to (26) and (36) approximately. But the approximation may not be sufficient enough when we consider the diagrams with two or more lockings. Thus we may say that the renormalization (26) and (36) represents the much higher approximation for the effect of anisotropy D , but is only the next step to the results of § 2 for the term of order S^0 . For the spin flop phase our result may contain some unsatisfactory points because we have neglected some terms in obtaining the effective interaction Hamiltonian (35). These terms seem to contribute some new corrections of order S^0 when we proceed to higher treatment.

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