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X-932-75-207
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NASA TM X- 70971

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(NASA-TM-X-70971) ON THE MAGNON INTERACTION IN HAEMATITE. 2: MAGNON ENERGY OF THE ACOUSTICAL MODE AND MAGNETIC CRITICAL FIELDS (NASA) 23 p HC $3.25 CSCL 08G

N75-31622

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AUGUST 1975

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ON THE MAGNON INTERACTION IN HAEMATITE. II. MAGNON ENERGY OF THE ACOUSTICAL MODE AND MAGNETIC CRITICAL FIELDS

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ABSTRACT

Recent findings indicate that magnetic anomalies occur in rocks in the sector in which an earthquake is recorded. The enhancement of the local geomagnetic field is believed to result from the pressure buildup beneath the ground before the quake. Also, there are certain cases in which magnetic anomalies appear to correlate with areas of tectonic activity and mineral concentration.

The antiferromagnet Haematite (α-Fe_2O_3) exhibits an anomalistic behavior about a magnetic ordering temperature called the Morin phase transition. Below this temperature the system exists entirely in the antiferromagnetic state, while above it, a ferromagnetic phase, and hence a net magnetic moment appears. In this report, a more extensive spin wave theory of Haematite is developed. The behaviour of thermodynamic quantities around the Morin transition temperature is studied, and also the latent heat of the Morin transition is calculated. The temperature dependence of the antiferromagnetic resonance frequency and the critical fields $Hc_{\parallel}$ and $Hc\perp$ is calculated. It is found that the theory agrees well with experiment.

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ON THE MAGNON INTERACTION IN HAEMATITE.
II. MAGNON ENERGY OF THE ACOUSTICAL MODE
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I. INTRODUCTION

All processes in nature including geophysical phenomena are thermodynamic in origin. Because of the magnitude of the pressures, volumes and temperatures associated with processes in the Earth's interior, most external manifestations of such events are large scale in time and space.

Magnetic field studies are a source of information on the properties, structure, and dynamics of the Earth's crust, mantle and core. Geomagnetic anomalies are known to correlate with geologic structures and with areas of mineral concentrations and tectonic activity. One of the most important mineral oxides to be found in sedimentary rocks is the magnetic compound known as Haematite. Haematite has the corundum structure and orders magnetically at the Néel temperature $T_N = 947$ K. For temperatures between T_N and the Morin temperature $T_M = 261$ K, haematite is a canted antiferromagnet with the spins perpendicular to the c axis. For temperatures below T_M, haematite is a uniaxial antiferromagnet with the spins lying along the c axis. The transition at T_M is called the Morin transition. There are a number of experimental and theoretical studies most of which are described in the paper of Jacobs et al. (1971).

The mechanism of the Morin transition in haematite was believed to be the sign change of the uniaxial anisotropy energy at T_M (Kanamori 1963, Artman et al 1965). However, several experiments including the antiferromagnetic resonance experiment of Foner and Williamson (1965) and the magnetically induced and the stress-induced Morin transition experiments (Flanders 1969, Allen 1973) were not explained by the simple anisotropy theory. In order to investigate the mechanism of the Morin transition, several people have recently studied the effect of the magnon interaction on the optical-mode magnon energy (Herbert 1970, Nagai et al. 1972). On the other hand, the temperature dependence of the acoustical-mode energy was studied by Bonavito (1970) and Watarai (1973) by using the two-sublattice model for this crystal. Very recently, the present authors (Nagai et al. 1973) precisely studied the spin-wave theory of haematite and pointed out that the critical temperature T_L at which the magnon energy in low-temperature phase becomes imaginary is slightly higher than the temperature T_H at which the magnon energy in the high-temperature phase becomes imaginary. Thus, we have two kinds of locally stable states in a narrow temperature region near T_M.

II. ACOUSTIC MODE ENERGY

We start from the following model Hamiltonian

$$H = 2J \sum_{\langle j,m \rangle} S_j . S_m - B[\sum_j S_{jz}^2 + \sum_m S_{mz}^2] - A \sum_{\langle j,m \rangle} S_{jz} S_{mz} - D \sum_{\langle j,m \rangle} (S_j \times S_m)_z , \tag{1}$$

assuming the two-sublattice body-centred lattice. Here $\mathbf{S}_j$ and $\mathbf{S}_m$ are the vector spin operators associated with the jth and mth atoms of the crystal and the sum $\Sigma_{\langle j,m \rangle}$ is extended over all nearest-neighbour pairs. The exchange interaction is given by the first term of (1). Since we assume $J > 0$, the jth and mth spins are coupled antiferromagnetically. The second term denotes the one-ion type uniaxial anisotropy energy which makes spins direct to the $\pm z$ axis. On the other hand, the dipolar type interaction which is given by the third term tends to make the spins lie in the xy plane. The rôle of the Dzyaloshinsky-Moriya (DM) interaction, the last term, is similar to that of the dipolar-type interaction except that the DM interaction makes the interacting spins orthogonal to each other when the spins lie in the xy plane.

Recently, Jacobs et al. (1972) pointed out that the crystalline anisotropy energy of the form

$$- C[\sum_j S_{jz}^4 + \sum_m S_{mz}^4] \tag{2}$$

may be important in explaining AFMR experiments. The effect of this energy will be discussed in IV.

In the low-temperature phase, the spins are parallel to the $\pm z$ axis, as described in the Introduction. By using the method described in our previous paper (1973), the magnon Hamiltonian H_L is given by

$$H_L = (2JSz)\sum_k [g_1(a_k^\dagger a_k + b_k^\dagger b_k) + g_2\gamma_k a_k b_k + g_2^*\gamma_k a_k^\dagger b_k^\dagger + g_3\gamma_k(a_k^\dagger b_{-k} + a_k b_{-k}^\dagger)$$
$$+ g_4(a_k a_{-k} + b_k b_{-k}) + g_4^*(a_k^\dagger a_{-k}^\dagger + b_k^\dagger b_{-k}^\dagger)]. \tag{3}$$

Here g_1 etc are given by

$$
\begin{aligned}
g_1 &= (1 - u_L - w_{Lr}) - \epsilon(1 - u_L) + \nu(1 - 1/2S - 2u_L) - idw_{Li},\\
g_2 &= (1 - u_L - w_L) - id(1 - u_L) + \epsilon w_L,\\
g_3 &= -x_L(1 - \epsilon) - \tfrac{1}{4}(y_L + y_L^*) - \tfrac{1}{4}id(y_L - y_L^*),\\
g_4 &= -\tfrac{1}{4}(2\nu y_L + x_L - idx_L),
\end{aligned}
\tag{4}
$$

where $\epsilon = A/2J$ and $\nu = B/Jz$. The parameters u_L, w_L, x_L and y_L are defined by $(2/NS)\Sigma_q\langle a_q^\dagger a_q\rangle$, $(2/NS)\Sigma_q\langle a_q^\dagger b_q^\dagger\rangle\gamma_q$, $(2/NS)\Sigma_q\langle a_q^\dagger b_{-q}\rangle\gamma_q$ and $(2/NS)\Sigma_q\langle a_q^\dagger a_{-q}^\dagger\rangle$ respectively. In (4), w_{Lr} and w_{Li} are defined by $(w_L + w_L^*)/2$ and $(w_L - w_L^*)/2$, respectively. The diagonalization of the Hamiltonian may be done as follows. Confining our attention to the positive half space of k, we define the new operators:

$$
\begin{aligned}
a_{+k} &= (\tfrac{1}{2})^{1/2}(A_k + C_k), \qquad & b_{-k} &= (\tfrac{1}{2})^{1/2}(A_k - C_k),\\
a_{-k} &= (\tfrac{1}{2})^{1/2}(B_k + D_k), \qquad & b_{+k} &= (\tfrac{1}{2})^{1/2}(B_k - D_k).
\end{aligned}
\tag{5}
$$

Furthermore, we introduce F_k and G_k by

$$
A_k = \frac{g_{2k}^* + 2g_4^*}{|g_{2k} + 2g_4|}F_k, \qquad C_k = -\frac{g_{2k} - 2g_4}{|g_{2k} - 2g_4|}G_k,
$$

where $g_{2k} = g_{2\gamma k}$ and $g_{3k} = g_{3\gamma k}$.

Finally, introducing the operators α_{ik} and β_{ik} by

$$
\begin{aligned}
F_k^\dagger &= \alpha_{1k}^\dagger\cosh\theta_k - \beta_{1k}\sinh\theta_k, \qquad & B_k^\dagger &= \beta_{1k}^\dagger\cosh\theta_k - \alpha_{1k}\sinh\theta_k,\\
G_k^\dagger &= \alpha_{2k}^\dagger\cosh\phi_k - \beta_{2k}\sinh\phi_k, \qquad & D_k^\dagger &= \beta_{2k}^\dagger\cosh\phi_k - \alpha_{2k}\sinh\phi_k,
\end{aligned}
$$

where

$$
\tanh 2\theta_k = \frac{|g_{2k} + 2g_4|}{g_1 + g_{3k}} \text{ and } \tanh 2\phi_k = \frac{|g_{2k} - 2g_4|}{g_1 - g_{3k}},
$$

we obtain

$$H_L = \sum_k \sum_{i=1}^{2} \hbar\omega_{ik}(\alpha_{ik}^{\dagger}\alpha_{ik} + \tfrac{1}{2}), \tag{6}$$

where

$$\hbar\omega_{ik} = 2JSz\left[(g_1 - (-1)^i g_{3k})^2 - |g_{2k} - (-1)^i 2g_4|^2\right]^{1/2}. \tag{7}$$

In our approximation, u_L etc are determined from the following equations:

$$u_L = \frac{1}{2NS}\sum_k \left[\cosh 2\theta_{1k}(1 + 2n_{1k}^L) + \cosh 2\theta_{2k}(1 + 2n_{2k}^L) - 2\right],$$

$$w_L = -\frac{1}{2NS}\sum_k \left[\frac{g_{2k}^* + 2g_4^*}{\hbar\omega_{1k}} \times (1 + 2n_{1k}^L) + \frac{g_{2k}^* - 2g_4^*}{\hbar\omega_{2k}} \times (1 + 2n_{2k}^L)\right] \times (2JSz),$$

$$x_L = \frac{1}{2NS}\sum_k \left[\frac{g_1 + g_{3k}}{\hbar\omega_{1k}} \times (1 + 2n_{1k}^L) - \frac{g_1 - g_{3k}}{\hbar\omega_{2k}} \times (1 + 2n_{2k}^L)\right] \times (2JSz), \tag{8}$$

$$y_L = -\frac{1}{2NS}\sum_k \left[\frac{g_{2k}^* + 2g_4^*}{\hbar\omega_{1k}} \times (1 + 2n_{1k}^L) - \frac{g_{2k}^* - 2g_4^*}{\hbar\omega_{2k}} \times (1 + 2n_{2k}^L)\right] \times (2JSz),$$

where $n_{ik}^L = [\exp(\beta\hbar\omega_{ik}) - 1]^{-1}$ with $\beta = 1/k_BT$. In the magnon renormalization approximation (MRA), u_L, w_L, x_L and y_L are calculated from (8) self-consistently.

Since u_L etc are of the order (1/S), the square root of (7) can be expanded in a power series with respect to (1/S). Since we retained only those terms up to order of $(1/S)^{-1}$ and $(1/S)^0$ in the Hamiltonian, the square root of (7) will be accurate up to order of (1/S), as pointed out previously (Nagai 1969).

By assuming that the anisotropy energy is very much smaller than the exchange energy, the temperature-dependent magnon energy with wavevector k can be written as

$$\hbar\omega_{ik} = 2JSz[(1-\gamma_k^2)(1-u_L-w_{L,t})^2 + (1-u_L-w_{L,t})\{2\nu'(1-2u_L) - (2\epsilon + d^2)(1-u_L+w_{L,t})\}]^{1/2}, \quad (9)$$

where $\nu' = \nu(1 - 1/2S)$. For the sake of convenience, we give the estimation of the magnitude of the anisotropy parameters in (9). According to the experiment, the critical field $H_{c\|}$ is 68 kG at T = 0 K. This corresponds to $\hbar\omega_{i0}$ = 10 K. We may assume 2JSz = 1000 K because T_N = 947 K. Hence we find that $(2\nu' - 2\epsilon - d^2)$ is of the order 10^{-4}. On the other hand, the experimental value of $H_{c\perp}$ is 160 kG at T = 0 K. The magnitude of d is estimated to be of the order of 10^{-2} from the relation $d = (g\mu_B/2JSz)(H_{c\|}^2/H_{c\perp})$, which will be explained in III-2. The calculation of dipolar energy leads to $\epsilon = 10^{-3}$, and hence ν is supposed to be of the order 10^{-3}.

If we use (9) for the magnon energy, the equation for T_L at which $\hbar\omega_{i0}(T)$ vanishes is given by

$$2\nu'(1-2u_L) - (2\epsilon + d^2)(1-u_L+w_L) = 0. \quad (10)$$

By using this equation, T_L is calculated as

$$\left(\frac{k_B T_L}{2JSz}\right) = \left(\frac{15S}{4\pi^2}\right)^{1/4}\left(\frac{2\nu' - 2\epsilon - d^2}{2\epsilon + d^2}\right)^{1/4}, \quad (11)$$

and $\hbar\omega_{i0}(T)$ is written as

$$\hbar\omega_{i0}(T) = \hbar\omega_{L0}(T) = \hbar\omega_{L0}(0)\,[1-(T/T_L)^4]^{1/2}, \quad (12)$$

where $\hbar\omega_{L0}(0) = 2JSz(2\nu' - 2\epsilon - d^2)^{1/2}$.

In the high-temperature phase, the spins are in the xy plane and they are canted slightly by the DM interaction. If we denote the angle between the jth and the mth spin vectors by $(\pi - 2\theta)$ as seen in Figure 1, the Hamiltonian is written as

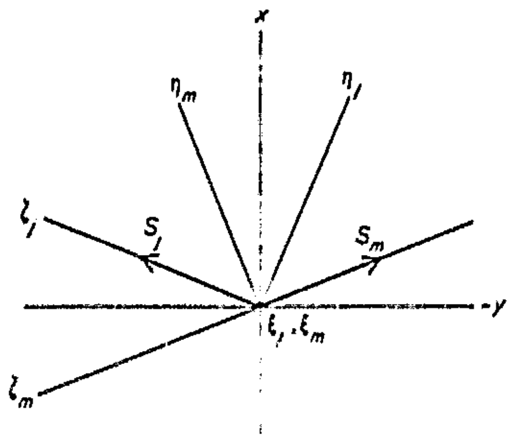

Figure 1. Coordinate Axes in the High-Temperature Phase.

$$H = \sum_{\langle j,m \rangle} [(2J\cos 2\theta + D\sin 2\theta)(S_{j\zeta}S_{m\zeta} + S_{j\eta}S_{m\eta}) + (2J - A)\, S_{j\xi}S_{m\xi} + (2J\sin 2\theta - D\cos 2\theta)(S_{j\zeta}S_{m\eta} - S_{j\eta}S_{m\zeta})] - B(\sum_j S_{j\xi}^2 + \sum_m S_{m\xi}^2). \tag{13}$$

The angle θ may be determined from the condition $2J \sin 2\theta - D \cos 2\theta = 0$. By using again the Holstein–Primakoff formalism, the magnon Hamiltonian for the high-temperature spin configuration is given in the same form with (3). In this case, g_1 etc, which are written as g_{II1} etc, are given by

$$\begin{aligned}
g_{II1} &= (1 - u_{II} - w_{IIr} - \tfrac{1}{2}x_{II}) + \tfrac{1}{2}\epsilon(w_{IIr} + x_{II}) + \tfrac{1}{2}d^2(1 - u_{II} - w_{IIr} + \tfrac{1}{2}x_{II}) \\
&\quad - \tfrac{1}{2}v'[1 - 2u_{II} - \tfrac{3}{4}(y_{II} + y_{II}^*)], \\
g_{II2} &= (1 - u_{II} - w_{II}) - \tfrac{1}{2}\epsilon(1 - u_{II} - \tfrac{1}{2}y_{II}) + \tfrac{1}{2}d^2[\tfrac{1}{2}(1 - u_{II}) - w_{II} + \tfrac{1}{4}y_{II}], \\
g_{II3} &= -x_{II} - \tfrac{1}{4}(y_{II} + y_{II}^*) - \tfrac{1}{2}\epsilon[1 - u_{II} - \tfrac{1}{4}(y_{II} + y_{II}^*)] - \tfrac{1}{2}d^2[\tfrac{1}{2}(1 - u_{II}) \\
&\quad + x_{II} + \tfrac{1}{8}(y_{II} + y_{II}^*)], \\
g_{II4} &= -\tfrac{1}{8}(2x_{II} + w_{II}) + \tfrac{1}{8}\epsilon(x_{II} + w_{II}) + \tfrac{1}{16}d^2(w_{II} - x_{II}) - \tfrac{1}{4}v'(1 + 1/4S - \tfrac{3}{2}u_{II} - \tfrac{1}{2}y_{II})
\end{aligned} \tag{14}$$

We may calculate the magnon energy $h\omega_{H\mathbf{k}}(T)$ using the formula (7), and then we obtain the critical temperature T_H where the magnon energy with infinite wavelength vanishes, $h\omega_{H0}(T_H) = 0$. We have shown in the previous paper (Nagai et al. 1973) that the critical temperature in the low-temperature phase T_L is slightly higher than the critical temperature in the high-temperature phase T_H. The difference between them is obtained as $\frac{1}{2}\nu' \times T_L$ and it is of the order 0·1 K. Using the condition that the free energies in both phases are equal, we calculate the Morin temperature T_M as $T_M = T_L \times (1 - \frac{3}{4}\nu)$. By using this T_M in (12), $h\omega_0(T_M)$ is estimated to be only a few kG. According to Foner and Williamson (1965), the magnitude of $h\omega_0(T_M)$ is 28 kG. Thus the theory does not agree with the experiment on this point. Recently, Jacobs et al. (1971) pointed out that this contradiction may be removed if we assume the S_z^4 type crystalline anisotropy energy. Even if this type of anisotropy is introduced the magnon energy spectrum with $k \neq 0$ may not be affected and accordingly the thermodynamic quantities are not affected. Hence we calculate the thermodynamic quantities without introducing the higher-order anisotropy energy in this section. The effect of this anisotropy energy on the physical quantities will be discussed in IV.

It was shown in our previous paper (1973) that the magnon energy with $k \neq 0$ at moderately high temperatures is given by

$$h\omega_{k\lambda}(T) = H_{\lambda 2}(1 - \gamma_k^2)^{1/2}, \tag{15}$$

where $H_{L2} = 2JSz(1 - u_L - w_L)$ for the low-temperature spin configuration and $H_{H2} = 2JSz(1 - u_H - w_H - \frac{1}{2}c)$ for the high-temperature spin configuration. The thermodynamic quantities around the Morin temperature can be calculated in the same way as in a previous paper (Nagai and Tanaka 1969). The difference between the sublattice magnetization M just below T_M and M just above T_M is calculated as

$$\Delta M = M(T_M - 0) - M(T_M + 0) \simeq \frac{N}{3}\left(\frac{k_B T_M}{2JSz}\right)^2 \times \nu. \tag{16}$$

The corresponding quantity in the case of the magnetic specific heat C(T) is calculated as

$$\Delta C = C(T_M - 0) - C(T_M + 0) \simeq Nk_B\left(\frac{8\pi^2}{15}\right)\left(\frac{k_B T_M}{2JSz}\right)^3 \times \nu. \quad (17)$$

Thus, we find both $(\Delta M/M)$ and $(\Delta C/C)$ are smaller than 1%. Recently, Allen (1973) analysed the experiment of the stress-induced Morin transition of this substance. He estimated the latent heat of this transition to be 5×10^5 erg/cm^{-3}. According to the present theory, the latent heat is calculated as

$$T_M \times [S(T_M + 0) - S(T_M - 0)] = Nk_B T_M\left(\frac{8\pi^2}{15}\right)\left(\frac{k_B T_M}{2JSz}\right)^3 \times \nu \quad (18)$$

where S(T) denotes the entropy. The latent heat of (18) is evaluated to be $2 \times 10^8 \times \nu$ erg cm^{-3}. This value is of the same order of magnitude as the Allen value. In the above calculations, we neglected the effect of the optical-mode magnon energy because its magnitude is of the order 10^3 K.

III. MAGNETIC CRITICAL FIELDS

1. Parallel Critical Field $H_{c\parallel}(T)$

It is known that the spin-flop critical field $H_{c\parallel}$ in the case of the uniaxial antiferromagnet is, in the molecular-field approximation (Nagamiya et al. 1955), given by

$$g\mu_B H_{c\parallel} = \hbar\omega_0(T)\left(\frac{\chi_\perp}{\chi_\perp - \chi_\parallel}\right)^{1/2}. \quad (19)$$

On the other hand, the spin-wave theory gives the same result at low temperatures (Nagai and Tanaka 1970). In the latter paper, the critical field is defined as the field at which the energy of the magnon with infinite wavelength vanishes. Exactly speaking, the critical field $H_{c\parallel}$ obtained in this way is slightly different from the critical field obtained from the condition that the free energies in both the flopped and unflopped phases are equal. Actually, according to our theory, AFMR does not vanish at T_M and its magnitude is calculated to be 2 kG at T_M. Therefore, $H_{c\parallel}(T_M)$ is also equal to 2 kG if the equation (19) is used for $H_{c\parallel}$. However, it is known that $H_{c\parallel}(T)$ must vanish at the Morin transition temperature because the Morin transition occurs without a magnetic field. If we inspect the equation for determining $H_{c\parallel}(T)$ more precisely, we will find $H_{c\parallel}(T_M) = 0$. Actually, T_L is very close to T_M and $H_{c\parallel}(T_M) = 2$ kG is very small compared with $H_{c\parallel}(0) = 68$ kG. Hence $H_{c\parallel}(T)$ is approximately given by (19) of which the main temperature behaviour is written as

$$H_{c\parallel}(T) = H_{c\parallel}(0)\,[1 - (T/T_M)^4]^{1/2} \tag{20}$$

where $H_{c\parallel}(0) = (2JSz/g\mu_B)\,(2\nu' - 2e - d^2)^{1/2}$. The theoretical result of $H_{c\parallel}(T)$ is compared with experiment of Foner and Shapira (1969) in Figure 2.

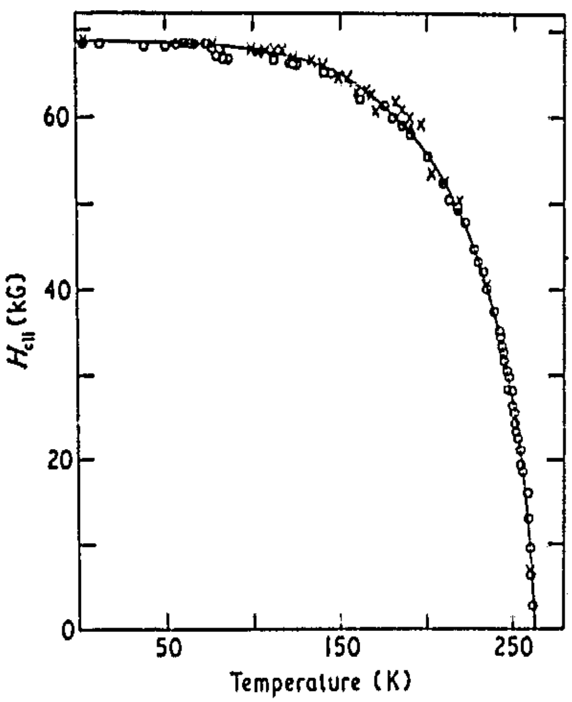

Figure 2. Temperature Dependence of $H_{c\parallel}(T)$ which is Computed from (19) is Compared with Experiment (Foner and Shapira 1969).

Next let us calculate T_M and $H_{c\parallel}(T)$ in the molecular-field approximation in order to see the correspondence of the spin-wave result to the molecular-field result. We neglect the DM interaction for simplicity.

The Weiss molecular-field Hamiltonian H_M^u in the unflopped phase is given by

$$H_M^u = (2J - A) \sum_{\langle j,m \rangle} [S_{j\zeta}\langle S_{m\zeta}\rangle + \langle S_{j\zeta}\rangle S_{m\zeta} - \langle S_{j\zeta}\rangle\langle S_{m\zeta}\rangle] - B[\sum_j S_{j\zeta}^2 + \sum_m S_{m\zeta}^2] - g\mu_B H [\sum_j S_{j\zeta} + \sum_m S_{m\zeta}]. \qquad (21)$$

Let us introduce the variables p and n by $\langle S_{j\zeta}\rangle = p + n$ and $\langle S_{m\zeta}\rangle = -p + n$. Here p denotes the average value of jth spin in the case $H = 0$ and n is approximately proportional to H. Furthermore, we define p_0 and p' by $p = p_0 + p'$. Here p' is proportional to A or B and $p_0 = \langle S_{j\zeta}\rangle_0 = \mathrm{Tr}[S_{j\zeta}\exp(-\beta H_M^0)]/Z_0$, where $H_M^0 = -2Jzp_0\Sigma_j S_{j\zeta}$ and $Z_0 = \mathrm{Tr}[\exp(-\beta H_M^0)]$. The free energy F_M^u in the unflopped phase is calculated as

$$F_M^u = -k_B T \ln Z_0 + NJzp_0^2(1 + \epsilon) - NJz\nu\langle S_{j\zeta}^2\rangle_0 - (\chi_\parallel H^2/2). \qquad (22)$$

The molecular-field Hamiltonian H_M^f in the flopped phase is given by

$$H_M^f = \sum_{\langle j,m \rangle} [(2J\cos 2\theta + A\sin^2\theta)\{S_{j\zeta}\langle S_{m\zeta}\rangle + \langle S_{j\zeta}\rangle S_{m\zeta} - \langle S_{j\zeta}\rangle\langle S_{m\zeta}\rangle\} - (B/z)\{(S_{j\zeta}^2 + S_{m\zeta}^2)\sin^2\theta + (S_{j\xi}^2 + S_{m\xi}^2)\cos^2\theta\}] - g\mu_B H\sin\theta(\sum_j S_{j\zeta} - \sum_m S_{m\zeta}), \qquad (23)$$

where the angle between H and S_j is given by $(\frac{1}{2}\pi - \theta)$.

Let us define p and n' by $\langle S_{j\zeta}\rangle = -\langle S_{m\zeta}\rangle = p + n'$ where p is a θ-independent term. Again we introduce p_0 and p'' by $p = p_0 + p''$ where p'' is proportional to A or B. Differentiating the free energy F_M^f with respect to θ, we obtain $\sin\theta = g\mu_B H/4Jzp_0$. Finally, the free energy is written as

$$F_M^f = NJzp_0^2 - k_B T \ln Z_0 - NB\langle S_{j\zeta}^2\rangle_0 - (\chi_\perp H^2/2). \qquad (24)$$

By equating F_M^u to F_M^f, we may calculate the Morin temperature T_M and also the critical field $H_{c\parallel}(T)$.

The equation for determining T_M in the case H = 0 is given by

$$2\nu\langle S_{j\zeta}^2 - S_{j\xi}^2\rangle_0 - 2\epsilon\langle S_{j\zeta}\rangle_0^2 = 0. \quad (25)$$

This equation was already used previously in order to explain the Morin transition (Kanamori 1963). The c itical field $H_{c\parallel}(T)$ is calculated from the following equation:

$$g\mu_B H_{c\parallel}(T) = 2Jz[2\nu\langle S_{j\zeta}^2 - S_{j\xi}^2\rangle_0 - 2\epsilon\langle S_{j\zeta}\rangle_0^2]^{1/2} \times [1 + (\chi_\parallel/2\chi_\perp)], \quad (26)$$

which should be compared with (19). If the DM interaction is taken into account, ϵ in (25) and (26) is replaced by $[\epsilon + (d^2/2)]$.

2. Perpendicular Critical Field $H_{c\perp}$

If a magnetic field is applied perpendicular to the c axis, say, parallel to the y axis, then the spins rotate in the xz plane and suddenly flop into the x axis at the critical field $H_{c\perp}(T)$. This situation can be seen in Figure 3.

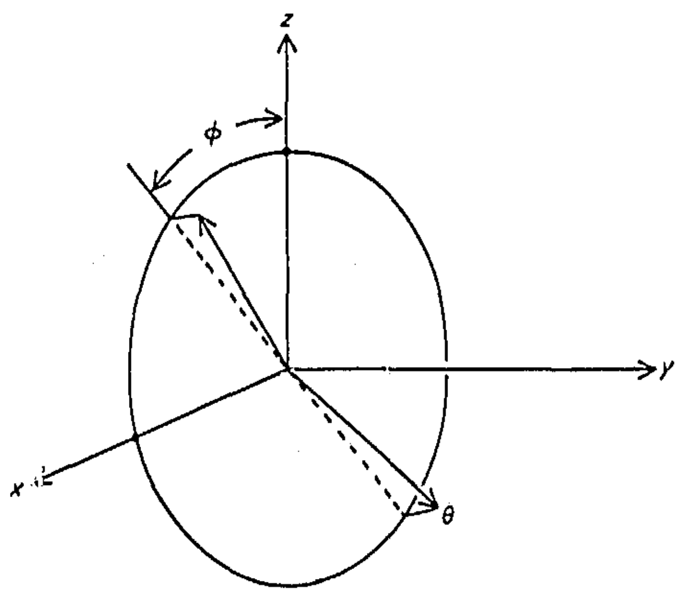

Figure 3. Spin Arrangement in the Magnetic Field (H||y Axis).

The Hamiltonian of the system is the sum of (1) and the Zeeman term

$$H_Z = -g\mu_B H(\sum_j S_{jy} + \sum_m S_{my}). \tag{27}$$

We take the coordinate axes $\{\xi_j \eta_j \zeta_j\}$ for the jth spin and the axes $\{\xi_{m} \eta_{m} \zeta_m\}$ for the mth spin. In this new system S_j and S_m are written as

$$\begin{pmatrix} S_{jx} \\ S_{jy} \\ S_{jz} \end{pmatrix} = \Phi_+(\theta, \phi) \begin{pmatrix} S_{j\xi} \\ S_{j\eta} \\ S_{j\zeta} \end{pmatrix} \qquad \begin{pmatrix} S_{mx} \\ S_{my} \\ S_{mz} \end{pmatrix} = \Phi_-(\theta, \phi) \begin{pmatrix} S_{m\xi} \\ S_{m\eta} \\ S_{m\zeta} \end{pmatrix}, \tag{28}$$

where

$$\Phi_\pm(\theta, \phi) = \begin{pmatrix} \cos\phi & \mp \sin\theta \sin\phi & \cos\theta \sin\phi \\ 0 & \cos\theta & \pm\sin\theta \\ -\sin\phi & \mp\sin\theta\cos\phi & \cos\theta\cos\phi \end{pmatrix}. \tag{29}$$

Substituting (28) into the Hamiltonian and expanding the Hamiltonian in terms of spin-wave operators, then we obtain a series

$$H = E_0 + H_1 + H_2 + H_3 + H_4, \tag{30}$$

where E_0 denotes the classical energy and is of the order S^2, and H_1, H_2, H_3 and H_4 are, respectively, of the order $S^{3/2}$, $S^{2/2}$, $S^{1/2}$ and S^0.

If we omit the last two terms in (30), we can obtain the temperature-independent solution of $H_{c\perp}$. The angles θ and ϕ are, in this approximation, given as follows: (i) $\sin\theta = h(\nu - \epsilon)/(2\nu - 2\epsilon - d^2)$ and $\sin\phi = d\tan\theta/(\nu - \epsilon)$ (unflopped phase); (ii) $\sin\theta = (h + d)/2$ and $\phi = \pi/2$ (flopped phase). Here h is defined by $h = g\mu_B H/2JSz$.

The free-magnon Hamiltonian in the unflopped phase is obtained in the same manner as that given in the previous section. After a somewhat extended manipulation, the free-magnon energies are given by

$$\hbar\omega^{u}_{1k} = 2JSz[1 - \gamma_k^2 + 2\nu - 2\epsilon - d^2 - (d^2h^2/(2\nu - 2\epsilon - d^2))]^{1/2},$$
$$\hbar\omega^{u}_{2k} = 2JSz[1 - \gamma_k^2 + 2\nu - 2\epsilon - d^2 + h^2\{4(\nu - \epsilon)^2 - 4d^2(\nu - \epsilon) - d^4\}/(2\nu - 2\epsilon - d^2)^2]^{1/2}, \quad (31)$$

where we used the relation $1 \gg d^2,\ \nu,\ \epsilon$. The critical field $h_{c\perp}$ is determined from the condition $\hbar\omega^{u}_{10} = 0$ and is given by $h_{c\perp} = (2\nu - 2\epsilon - d^2)/d$. On the other hand, $h_{c\parallel}$ is given by $h_{c\parallel} = (2\nu - 2\epsilon - d^2)^{1/2}$ and hence $h_{c\perp}$ is written as $h_{c\perp} = h^2_{c\parallel}/d$.

The magnon Hamiltonian in the flopped phase is obtained similarly. The free-magnon frequencies in the flopped phase are written as

$$\hbar\omega^{f}_{1k} = 2JSz[1 - \gamma_k^2 + hd - (2\nu - 2\epsilon - d^2)]^{1/2},$$
$$\hbar\omega^{f}_{2k} = 2JSz[1 - \gamma_k^2 + h(h + d)]^{1/2}. \quad (32)$$

The temperature dependence of the critical field $H_{c\perp}$ (T) may be obtained from the condition that the temperature-dependent magnon energy with inifinite wavelength vanishes. The calculation of $H_{c\perp}$ (T) was performed as follows: (i) the temperature dependence of θ and ϕ is taken into account by applying the RPA in the cubic terms of magnon operators in H_3; (ii) the quartic terms of magnon operators in H_4 are treated by using RPA; (iii) the non-diagonal matrix elements of the cubic terms of magnon operators in H_3 are taken into account by using the second-order perturbation approximation (Maekawa 1973). After a complicated manipulation, the temperature dependence of the perpendicular critical field was found to be $h_{c\perp}(T) = [h_{c\parallel}(T)]^2/d$. We will not write out the derivation of this result by the use of the above-mentioned method, because it is much too complicated and lengthy. Instead, we derive the same result by the use of molecular-field approximation.

The molecular-field Hamiltonian is written as

$$H_M = 2J\sum_{\langle j,m\rangle}[(\cos 2\theta - \cos^2\theta\cos^2\phi + d\sin 2\theta\sin\phi)(S_{j\zeta}p_m + p_jS_{m\zeta} - p_jp_m) - (\epsilon/2)\{\sin^2\phi(S^2_{j\xi} + S^2_{m\xi}) + \sin^2\theta\cos^2\phi(S^2_{j\eta} + S^2_{m\eta})\} - Sh\sin\theta(S_{j\zeta} - S_{m\zeta})], \quad (33)$$

where $p_j = \langle S_{j\zeta}\rangle$. By using a method similar to that given in the previous section, the free energy in the molecular-field theory can be calculated.

From the conditions $\partial F_M/\partial \theta = 0$ and $\partial F_M/\partial \phi = 0$, we obtain the following two equations:

$$\cos\theta\cos\phi[\cos\theta\sin\phi(\nu\langle S_{j\zeta}^2 - S_{j\xi}^2\rangle_0 - \epsilon\langle S_{j\zeta}\rangle_0^2) - d\sin\theta\langle S_{j\zeta}\rangle_0^2] = 0,$$

$$\sin 2\theta[2\langle S_{j\zeta}\rangle_0^2 + \cos^2\phi\{\nu\langle S_{j\zeta}^2 - S_{j\xi}^2\rangle_0 - \epsilon\langle S_{j\zeta}\rangle_0^2\}] - 2d\cos 2\theta\sin\phi\langle S_{j\zeta}\rangle_0^2 = 2S\langle S_{j\zeta}\rangle_0 h\cos\theta. \quad (34)$$

In deriving (34), we used the relation $\langle S_{j\xi}S_{j\eta} + S_{j\eta}S_{j\xi}\rangle_0 = \langle S_{m\xi}S_{m\eta} + S_{m\eta}S_{m\xi}\rangle_0 = 0$. In the unflopped phase, θ and ϕ are determined from the following equations:

$$\sin\phi = d\tan\theta/(\nu_1 - \epsilon),$$

$$h = \sin\theta(\langle S_{j\zeta}\rangle_0/S) \times \left(2 - \frac{d^2}{\nu_1 - \epsilon}\right), \quad (35)$$

where $\nu_1 = \nu\langle S_{j\zeta} - S_{j\xi}\rangle_0/\langle S_{j\zeta}\rangle_0^2$. On the other hand, we have $\phi = \pi/2$ and

$$2\langle S_{j\zeta}\rangle_0\sin\theta = Sh + d\langle S_{j\zeta}\rangle_0 \quad (36)$$

for the flopped phase. By equating F_M (flopped phase) to F_M (unflopped phase), the equation for determing the perpendicular critical field is given by

$$h' = (2\nu_1 - 2\epsilon - d^2)/d \quad (37)$$

where $h' = Sh/\langle S_{j\zeta}\rangle_0$. If we use (26) we may write

$$h_{c\perp}(T) = \frac{1}{d}\frac{S}{\langle S_{j\zeta}\rangle_0}[h_{c\parallel}(T)]^2 \quad (38)$$

of which the main temperature dependence is given by $[1-(T/T_M)^4]$. The computed result of $h_{c\perp}$ (T) is compared with experiment in Figure 4.

IV. FOUR-SUBLATTICE MODEL

Let us compare the theory based on the four-sublattice model with experiment. As can be seen in the previous paper (Nagai et al. 1972), the energy of the magnon with infinite wavelength is written as

$$(\hbar\omega_0)^2 = -S^2D^2 + 2S^2(2J_0 + 6J_2 + 12J_3)[2U - \tfrac{3}{4}(\Phi_{11} - \Phi_{12} - \Phi_{13} + \Phi_{14})]. \quad (39)$$

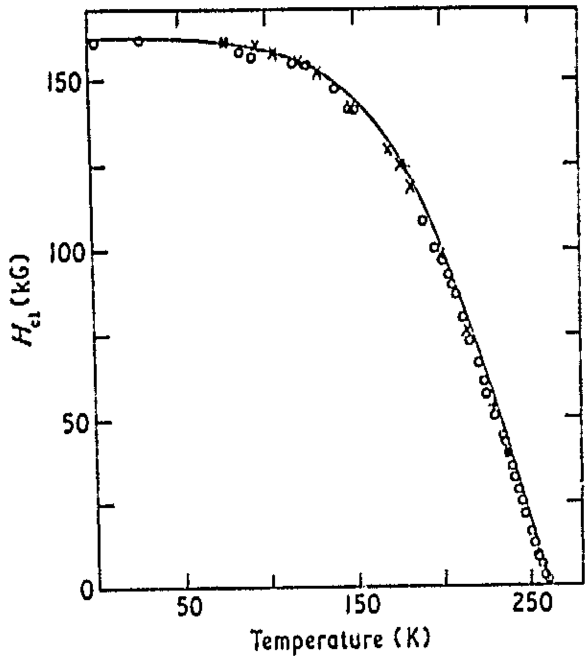

Figure 4. Temperature Dependence of $H_{c\perp}$ (T) which is Computed from (38) is Compared with Experiment (Foner and Shapira 1969).

where D denotes the DM constant and U the uniaxial anisotropy constant. The dipolar anisotropy constants ϕ_{NM} are defined by

$$\Phi_{NM} = 4\mu_B^2 \sum_{R_{NM}} (R_{NM}^2 - 3Z_{NM}^2)/(R_{NM})^5 = (4\mu_B^2/a^3)\, P_{NM}$$

where a denotes the lattice parameter 5 · 42 x 10^{-8}cm, R_N and R_M belong to the N and M sublattices, respectively, and $R_{NM} = R_N - R_M$. The numerical values are given by $P_{11} = 1 \cdot 09$, $P_{12} = -9 \cdot 15$, $P_{13} = -2 \cdot 03$ and $P_{14} = 11 \cdot 01$. By using the exchange parameters proposed by Samuelsen and Shirane (1970), we obtain $2J_0 + 6J_2 + 12J_3 = 468$ K. It is seen by inspection that we may adopt the following substitutions:

$$2Jz = 2J_0 + 6J_2 + 12J_3, \qquad h = (g\mu_B H/S)/(2J_0 + 6J_2 + 12J_3),$$

$$v = U/(J_0 + 3J_2 + 6J_3), \qquad d = D/(2J_0 + 6J_2 + 12J_3),$$

$$\epsilon = \frac{3}{4}\frac{\Phi_{11} - \Phi_{12} - \Phi_{13} + \Phi_{14}}{2J_0 + 6J_2 + 12J_3}.$$

By using the experimental values of the critical fields at T = 0 K, we obtain $U = 0 \cdot 222\ k_B$ and T_M is estimated to be 258 K.

1. Effect of S_z^4-term

In this section, we give a qualitative discussion of the effect of the higher-order anisotropy energy on the magnon frequency. If we use the Holstein-Primakoff formalism for spin operators, the higher-order anisotropy energy is written in a series

$$-CS_{jz}^4 = -CS^4[1 - (4/S)(1 - 3/2S)\, a_j^\dagger a_j + (6/S^2)\, a_j^\dagger a_j^\dagger a_j a_j + \text{higher-order terms of } (1/S)].$$

We retain up to the third term in the brackets and neglect the others. By using the method described in II, the magnon energy of (9) can be written as

$$\hbar\omega_{Lk}(T) = 2JSz[(1 - \gamma_k^2)(1 - u_L - w_L)^2 + (1 - u_L - w_L)\{2v'(1 - 2u_L) + 4c(1 - 6u_L) - (2\epsilon + d^2)(1 - u_L + w_L)\}]^{1/2}, \tag{40}$$

where $c = CS^2(1 - 3/2S)/Jz$. The critical temperature T_L' at which $\hbar\omega_{L0}(T)$ vanishes is given by

$$\left(\frac{k_B T_L'}{2JSz}\right) = \left(\frac{15S}{4\pi^2}\right)^{1/4} \left(\frac{2v' + 4c - 2\epsilon - d^2}{2\epsilon + d^2}\right)^{1/4}. \tag{41}$$

On the other hand, it is easily seen that the magnon energy in the high-temperature phase is given by

$$\hbar\omega_{Hk}(T) = 2JSz[(1 - \gamma_k^2)(1 - u_H - w_H - \tfrac{1}{2}\epsilon)^2 + (1 - u_H - w_H)\{(2\epsilon + d^2)(1 - u_H + w_H) - 2v'(1 - 2u_H) - 3c(1/2S) + u_H)\}]^{1/2}. \tag{42}$$

Thus, the magnon energy $\hbar\omega_{Hk}(T)$ is not affected by the introduction of c, and it is approximately written as

$$\hbar\omega_{H0} = 2JSz(2v' - 2\epsilon - d^2)^{1/2}[-1 + (T/T_H)^4]^{1/2}, \tag{43}$$

where

$$\left(\frac{k_B T_H}{2JSz}\right) = \left(\frac{15S}{4\pi^2}\right)^{1/4} \left(\frac{2\nu' - 2\epsilon - d^2}{2\epsilon + d^2}\right)^{1/4}.$$

The Morin temperature can be calculated in a manner similar to that used in III-1. The equation for determining T_M is written as

$$2\nu'(1 - 2u_L) - (2\epsilon + d^2)(1 - u_L + w_L) + 2c(1 - 4u_L) = 0, \qquad (44)$$

by which T_M is written as

$$\left(\frac{k_B T_M}{2JSz}\right) = \left(\frac{15S}{4\pi^2}\right)^{1/4} \left(\frac{2\nu' + 2c - 2\epsilon - d^2}{2\epsilon + d^2}\right)^{1/4}. \qquad (45)$$

If we use the above expression for T_M, AFMR is written as

$$\hbar\omega_{L0}(T) = 2JSz[2c + (2\nu' + 2c - 2\epsilon - d^2)\{1 - (T/T_M)^4\}]^{1/2}, \qquad (46)$$

and we also obtain $\hbar\omega_{L0}(T_M) = \hbar\omega_{H0}(T_M) = 2JSz(2c)^{1/2}$. By using the experimental value $\hbar\omega_0(T_M) = 28$ kG, c is estimated to be $0 \cdot 58 \times 10^{-5}$. If we write (46) in the following form

$$\hbar\omega_{L0}(T) = \hbar\omega_{L0}(0)[1 - (T/T_L')^4]^{1/2}, \qquad (46)$$

T_L' is written as $T_L' = T_M(1 + \delta)$. Here $\delta = (c/2)/(2\nu' + 4c - 2\epsilon - d^2)$ is estimated to be $0 \cdot 04$. The temperature dependence of $\hbar\omega_{L0}(T)$ which is computed from (40) is compared with experiment in Figure 5. Thus, the critical temperature T_L' and also $\hbar\omega_{H0}(T_M)$ are strongly modified by the introduction of c. However, the magnon energy spectrum does not strongly depend on the magnitude of c at moderately high temperatures, as can be seen from (40) and (42). Therefore, the temperature behaviour of thermodynamic quantities may not be affected by c. The previous result (16) ~ (18) will still hold. Here T_M in these equations should be replaced by T_M of (45).

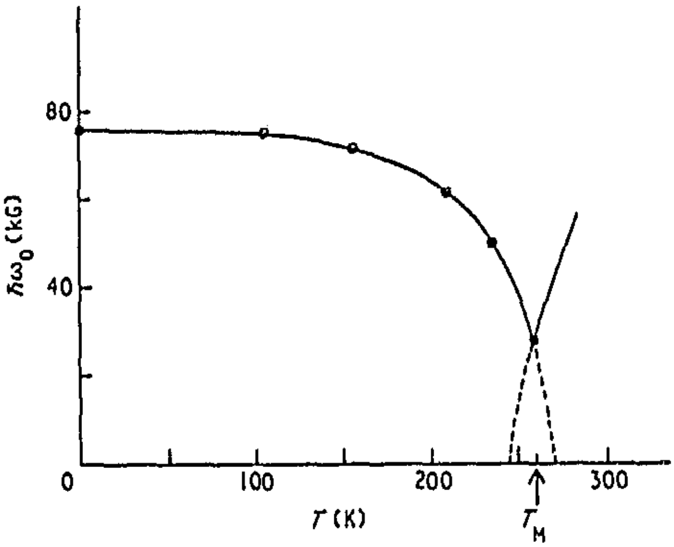

Figure 5. Temperature dependence of $h\omega_{L0}(T)$ and $h\omega_{||0}(T)$, which are computed from (40) and (42), respectively. Open circles are deduced from 70 GHz data (Foner and Williamson 1965).

Similarly, the critical fields $h_{c||}(T)$ and $h_{c\perp}(T)$ are, respectively, given by (20) and (38), where $h_{c||}(0)$ and T_M should be replaced by $(2\nu' + 2c - 2\epsilon - d^2)^{1/2}$ and (45), respectively.

V. CONCLUSION

We have seen in this paper that the calculated result of $H_{c||}$, $H_{c\perp}$ and $h\omega_0(T)$ shows a very good agreement with the experimental result. We took not $2JSz[2\nu(1 - 1/2S - 2u_L) - (2\epsilon + d^2)(1 - u_L + w_L)]^{1/2}$ but $2JSz[2\nu(1 - 1/2S)(1 - 2u_L) - (2\epsilon + d^2)(1 - u_L + w_L)]^{1/2}$ for the expression of $h\omega_{L0}(T)$. These two expressions are both accurate because the square root is accurate up to order of (1/S), as noted in II. The latter expression shows $[1 - (T/T_M)^4]^{1/2}$ dependence, while the former shows a slightly different temperature dependence. The S_z^2 term does not work as the anisotropy energy in the case $S = \frac{1}{2}$ and hence the latter expression will be more adequate. The derivation of the latter expression for $h\omega_{L0}(T)$ from the theoretical point of view may be made by using a more accurate theory than the present RPA. This remains to be done as a future problem.

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