Temperature-Dependent Magnon-Energy Theory of FeF_2 and MnF_2†

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The temperature dependence of the antiferromagnetic-resonance frequency, sublattice magnetization, and magnetic specific heat of FeF_2 and MnF_2 is calculated by use of the temperature-dependent magnon-energy theory, which was recently presented by the authors. The calculations were done in both the conventional random-phase approximation (RPA) and the magnon-renormalization approximation (MRA). The experimental data are mostly found between these two approximate calculations. Thus, it seems that the magnon interaction effects are overestimated in the MRA. The calculation of the magnon energy at the first Brillouin-zone edge in the case of MnF_2 also supports this conclusion.

I. INTRODUCTION

SINCE Anderson[1] has shown that the spin-wave theory is a good approximate method for the ground state of antiferromagnets, many authors[2–8] have applied this method to the calculation of the thermodynamic properties of these substances. It has been known that the spin-wave theory reproduces well the experiments on MnF_2 at low temperatures.[9,10] Recent calculations,[11,12] in which the dynamical spin-wave interaction is taken into account, have proved the capability of the spin-wave theory in calculating the thermodynamic quantities at moderately high temperatures such as $0.6\ T_N$. Here T_N denotes the Néel temperature. Low[11] showed that the spin-wave theory can well explain the experiments on the temperature dependence of the sublattice magnetization of MnF_2 up to $0.9\ T_N$, if we use the neutron inelastic scattering data on the temperature dependence of the magnon energy. In his calculation, he also used the experimental data of the antiferromagnetic-resonance frequency for the spin-wave energy with $\mathbf{k}=0$. Here $\mathbf{k}$ denotes the wave vector of a magnon. To obtain the temperature-dependent magnon energy theoretically, he used the magnon-renormalization approximation (MRA).[13] This MRA or the quasiparticle model for magnons may be accurate if magnon energies are obtained accurately as functions of the magnon energies themselves. However, the Hamiltonian in this approximation is expanded only up to the quartic terms with respect to the magnon operators, and, consequently, the MRA may be inappropriate at high temperatures. Actually, it has been shown that the magnon energy with $\mathbf{k}=0$ obtained in this approximation is real only below some temperature which is lower than T_N.[14] Kanamori and Itoh[12] reported a spin-wave calculation of the magnetic susceptibility in which a higher-order contribution of the spin-wave interaction terms is taken into account. They found that the theory was in agreement with the experiments on MnF_2 up to $0.6\ T_N$.

In this paper, we report on a comparison with the experiment of the spin-wave theory which was developed recently by one of the present authors (O.N.).[14] In that theory, which is based on the Holstein-Primakoff formulation for the Hamiltonian, the spin-wave interaction terms are treated by means of a random-phase approximation (RPA). Then our Hamiltonian is quadratic with respect to the magnon operators, and the obtained magnon energy includes some parameters which are statistical averages of the magnon operator functions. The approximation in which these parameters are determined self-consistently is called MRA. On the other hand, the method in which the free spin-wave theory is used for the calculation of these parameters may be called the RPA. If we use this RPA in the case of ferromagnetism, it is shown that the ferromagnetic magnon energy decreases with increasing temperature. The decrease is proportional to $T^{5/2}$ at low temperatures. Here T is the temperature in °K. If we take into account this temperature dependence of magnon

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energy, we obtain the correct low-temperature dependence of the magnetization, except for the additional factor $0.2/S$ to the T^4 term,[15] S denoting the magnitude of spin. This was first pointed out by Keffer and Loudon.[16] Similarly, if we use this RPA in the case of antiferromagnetism, we will obtain the *correct* low-temperature dependence of the sublattice magnetization, within the framework of the Holstein-Primakoff approach.

The spin-wave theory will be rather precisely compared with the experiments on FeF_2 and MnF_2 in both cases of RPA and MRA in this paper. According to the present result, the experimental data exists mostly between these two approximate calculations. Thus, it seems to be suggested that the MRA overestimates the spin-wave interaction effects. We calculate the antiferromagnetic-resonance frequency (AFMR), sublattice magnetization, and magnetic specific heat. As mentioned in the previous paper,[14] the temperature dependence of AFMR depends on both the temperature dependence of the exchange stiffness and that of the anisotropy energy. On the other hand, in the case of MnF_2, the sublattice magnetization and specific heat are not strongly affected by the anisotropy energy at high temperatures, and they are affected by the temperature dependence of the exchange stiffness. Thus, it may be interesting to examine our theory by comparing the calculation of the above-mentioned quantities with experiments. The method of calculation will be recapitulated in Sec. II. A numerical computation and its comparison with experiments will be shown in Sec. III.

II. MATHEMATICAL METHOD

In this section, we will outline our theory briefly and define the several terminologies which describe the method of numerical computation.

For the sake of convenience, we develop our theory by assuming that the Hamiltonian is given by the sum of the exchange and the uniaxial one-ion-type anisotropy energies:

$$H=2J\sum_{\langle j,m\rangle}\mathbf{S}_j\cdot\mathbf{S}_m-D[\sum_j(S_j^z)^2+\sum_m(S_m^z)^2], \quad (2.1)$$

where J denotes the exchange coupling between nearest-neighboring spins S_j and S_m, which belong to different sublattices $+$ and $-$, and D the constant proportional to the anisotropy energy. A sum $\sum_{\langle j,m\rangle}$ is extended over all those neighboring pairs.

Thus, the theory in this section may be applicable to FeF_2. The theory of MnF_2, whose magnetic anisotropy is mainly originated from the interionic dipolar interaction, will be given in Sec. III.

Following Kubo,[2] we define

$$\begin{aligned} &S_j^+=(2S)^{1/2}f_ja_j, && S_m^+=(2S)^{1/2}b_m^\dagger f_m,\\ &S_j^-=(2S)^{1/2}a_j^\dagger f_j, && S_m^-=(2S)^{1/2}f_mb_m, \quad (2.2)\\ &S_j^z=S-a_j^\dagger a_j, && S_m^z=-S+b_m^\dagger b_m,\\ &f_j=(1-a_j^\dagger a_j/2S)^{1/2}, && f_m=(1-b_m^\dagger b_m/2S)^{1/2}, \end{aligned}$$

where $a_j^\dagger$ and $b_m^\dagger$ are creation operators, and a_j and b_m are destruction operators of spin deviations. These operators satisfy the boson commutation relation. Substituting (2.2) into (2.1) and expanding f_j and f_m in a power series of $1/S$, we have a series

$$H=H_1+H_2+\cdots, \quad (2.3)$$

apart from a leading term which equals the classical energy to be obtained in the case of complete alignment of spins. Here H_1 and H_2 are terms of the order $(1/S)^{-1}$ and $(1/S)^0$, respectively. Other higher-order terms will be neglected. We introduce the Fourier transforms of a_j and b_m:

$$a_{\mathbf{k}}=\left(\frac{2}{N}\right)^{1/2}\sum_j a_je^{i\mathbf{k}\cdot\mathbf{R}_j}, \quad b_{\mathbf{k}}=\left(\frac{2}{N}\right)^{1/2}\sum_m b_me^{-i\mathbf{k}\cdot\mathbf{R}_m}, \quad (2.4)$$

where $\frac{1}{2}N$ is the number of spins belonging to a sublattice. Then H_1 and H_2 can be written

$$H_1=2JSz\sum_{\mathbf{k}}\{[1+\theta(1-1/2S)](a_{\mathbf{k}}^\dagger a_{\mathbf{k}}+b_{\mathbf{k}}^\dagger b_{\mathbf{k}})+\gamma_{\mathbf{k}}(a_{\mathbf{k}}b_{\mathbf{k}}+a_{\mathbf{k}}^\dagger b_{\mathbf{k}}^\dagger)\}, \quad (2.5)$$

$$\begin{aligned} H_2=&-2Jz\frac{2}{N}\sum_{\mathbf{k}_1\cdots\mathbf{k}_4}\gamma_{\mathbf{k}_3-\mathbf{k}_4}a_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2}b_{\mathbf{k}_3}^\dagger b_{\mathbf{k}_4}\delta(\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3+\mathbf{k}_4)\\ &-2Jz\frac{1}{2N}\sum_{\mathbf{k}_1\cdots\mathbf{k}_4}\{(\gamma_{\mathbf{k}_4}a_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2}a_{\mathbf{k}_3}b_{\mathbf{k}_4}+\gamma_{\mathbf{k}_1}a_{\mathbf{k}_1}^\dagger b_{\mathbf{k}_2}^\dagger b_{\mathbf{k}_3}^\dagger b_{\mathbf{k}_4})\delta(\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3+\mathbf{k}_4)\\ &\qquad+(\gamma_{\mathbf{k}_1}a_{\mathbf{k}_1}b_{\mathbf{k}_2}^\dagger b_{\mathbf{k}_3}b_{\mathbf{k}_4}+\gamma_{\mathbf{k}_4}a_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2}^\dagger a_{\mathbf{k}_3}b_{\mathbf{k}_4}^\dagger)\delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\}\\ &-D\frac{2}{N}\sum_{\mathbf{k}_1\cdots\mathbf{k}_4}(a_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2}^\dagger a_{\mathbf{k}_3}a_{\mathbf{k}_4}+b_{\mathbf{k}_1}^\dagger b_{\mathbf{k}_2}^\dagger b_{\mathbf{k}_3}b_{\mathbf{k}_4})\delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4). \end{aligned} \quad (2.6)$$

Here z denotes the number of nearest neighbors $\theta=D/Jz$ and $\gamma_{\mathbf{k}}=(1/z)\sum_\rho e^{i\mathbf{k}\cdot\boldsymbol{\rho}}$, $\boldsymbol{\rho}$ being a vector directed between nearest neighbors. In keeping with the previous paper,[14] we use a RPA for the quartic terms of the magnon oper-

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ators in (2.6). For instance,

$$\sum_{\mathbf{k}_1\cdots\mathbf{k}_4}\gamma_{\mathbf{k}_3-\mathbf{k}_4}a_{\mathbf{k}_1}{}^{\dagger}a_{\mathbf{k}_2}b_{\mathbf{k}_3}{}^{\dagger}b_{\mathbf{k}_4}\delta(\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3+\mathbf{k}_4)=\sum_{\mathbf{k},\mathbf{q}}[a_{\mathbf{k}}{}^{\dagger}a_{\mathbf{k}}\langle b_{\mathbf{q}}{}^{\dagger}b_{\mathbf{q}}\rangle+b_{\mathbf{k}}{}^{\dagger}b_{\mathbf{k}}\langle a_{\mathbf{q}}{}^{\dagger}a_{\mathbf{q}}\rangle-\langle a_{\mathbf{k}}{}^{\dagger}a_{\mathbf{k}}\rangle\langle b_{\mathbf{q}}{}^{\dagger}b_{\mathbf{q}}\rangle$$
$$+\gamma_{\mathbf{k}-\mathbf{q}}(a_{\mathbf{k}}b_{\mathbf{k}}\langle a_{\mathbf{q}}{}^{\dagger}b_{\mathbf{q}}{}^{\dagger}\rangle+a_{\mathbf{k}}{}^{\dagger}b_{\mathbf{k}}{}^{\dagger}\langle a_{\mathbf{q}}b_{\mathbf{q}}\rangle-\langle a_{\mathbf{k}}{}^{\dagger}b_{\mathbf{k}}{}^{\dagger}\rangle\langle a_{\mathbf{q}}b_{\mathbf{q}}\rangle+a_{\mathbf{k}}b_{-\mathbf{k}}\langle a_{\mathbf{q}}{}^{\dagger}b_{-\mathbf{q}}{}^{\dagger}\rangle+a_{\mathbf{k}}{}^{\dagger}b_{-\mathbf{k}}{}^{\dagger}\langle a_{\mathbf{q}}b_{-\mathbf{q}}\rangle-\langle a_{\mathbf{k}}{}^{\dagger}b_{-\mathbf{k}}{}^{\dagger}\rangle\langle a_{\mathbf{q}}b_{-\mathbf{q}}\rangle)]. \quad (2.7)$$

By $\langle\ \rangle$ we denote a statistical average. For a while, let us leave them undetermined. The c-number terms in this equation are useful in the calculation of the magnetic specific heat. Our Hamiltonian is now quadratic with respect to $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ and their complex conjugates:

$$H=\sum_{\mathbf{k}}[H_{1\mathbf{k}}(a_{\mathbf{k}}{}^{\dagger}a_{\mathbf{k}}+b_{\mathbf{k}}{}^{\dagger}b_{\mathbf{k}})+H_{2\mathbf{k}}(a_{\mathbf{k}}b_{\mathbf{k}}+a_{\mathbf{k}}{}^{\dagger}b_{\mathbf{k}}{}^{\dagger})]-(\tfrac{1}{2}NS)\times\sum_{\mathbf{k}}[(H_{1\mathbf{k}}-H_{1\mathbf{k}}{}^{F})u_{\mathbf{k}}{}'+(H_{2\mathbf{k}}-H_{2\mathbf{k}}{}^{F})w_{\mathbf{k}}{}'], \quad (2.8)$$

where

$$H_{1\mathbf{k}}=2JSz[1+\theta(1-1/2S)-(u'+w')-2\theta u'], \quad (2.9a)$$

$$H_{2\mathbf{k}}=2JSz(1-u'-w')\gamma_{\mathbf{k}}, \quad (2.9b)$$

and u' and w' are defined by

$$u'=\sum_{\mathbf{q}}u_{\mathbf{q}}{}'=\frac{2}{NS}\sum_{\mathbf{q}}\langle a_{\mathbf{q}}{}^{\dagger}a_{\mathbf{q}}\rangle, \quad (2.10a)$$

$$w'=\sum_{\mathbf{q}}w_{\mathbf{q}}{}'\gamma_{\mathbf{q}}=\frac{1}{NS}\sum_{\mathbf{q}}\gamma_{\mathbf{q}}(\langle a_{\mathbf{q}}b_{\mathbf{q}}\rangle+\langle a_{\mathbf{q}}{}^{\dagger}b_{\mathbf{q}}{}^{\dagger}\rangle). \quad (2.10b)$$

In the second term of (2.8), $H_{1\mathbf{k}}{}^{F}$ and $H_{2\mathbf{k}}{}^{F}$ are defined by $H_{1\mathbf{k}}$ and $H_{2\mathbf{k}}$, respectively, in which $u_{\mathbf{q}}{}'$ and $w_{\mathbf{q}}{}'$ are replaced by zero,

$$H_{1\mathbf{k}}{}^{F}=2JSz[1+\theta(1-1/2S)] \quad (2.11a)$$

$$H_{2\mathbf{k}}{}^{F}=2JSz\gamma_{\mathbf{k}}. \quad (2.11b)$$

The diagonal Hamiltonian is written

$$H=\sum_{\mathbf{k}}[\hbar\omega_{\mathbf{k}}(\alpha_{\mathbf{k}}{}^{\dagger}\alpha_{\mathbf{k}}+\beta_{\mathbf{k}}{}^{\dagger}\beta_{\mathbf{k}}+1)-H_{1\mathbf{k}}]-(\tfrac{1}{2}NS)\times\sum_{\mathbf{k}}[(H_{1\mathbf{k}}-H_{1\mathbf{k}}{}^{F})u_{\mathbf{k}}{}'+(H_{2\mathbf{k}}-H_{2\mathbf{k}}{}^{F})w_{\mathbf{k}}{}'], \quad (2.12)$$

where $\alpha_{\mathbf{k}}$ and $\beta_{\mathbf{k}}$ and their complex conjugates are again boson operators. The magnon energy $\hbar\omega_{\mathbf{k}}$ is given by

$$\hbar\omega_{\mathbf{k}}=2JSz[(H_{1\mathbf{k}})^2-(H_{2\mathbf{k}})^2]^{1/2}, \quad (2.13)$$

which is temperature-dependent.

Now we consider the following possible approximations.

A. Free Spin-Wave Approximation (FSW)

In this approximation, the spin-wave interaction is neglected. Hence, we have

$$\langle a_{\mathbf{q}}{}^{\dagger}a_{\mathbf{q}}\rangle=\langle b_{\mathbf{q}}{}^{\dagger}b_{\mathbf{q}}\rangle=\langle a_{\mathbf{q}}b_{\mathbf{q}}\rangle=\langle a_{\mathbf{q}}{}^{\dagger}b_{\mathbf{q}}{}^{\dagger}\rangle=0, \quad (2.14)$$
$$\langle a_{\mathbf{q}}a_{\mathbf{q}}{}^{\dagger}\rangle=1.$$

Then, $u_{\mathbf{q}}{}'$, $w_{\mathbf{q}}{}'$, and $v_{\mathbf{q}}{}'$, which will be written $u_{F\mathbf{q}}$, $w_{F\mathbf{q}}$, and $v_{F\mathbf{q}}$, respectively, are given by $u_{F\mathbf{q}}=w_{F\mathbf{q}}=0$ and $v_{F\mathbf{q}}=1/NS$, where

$$v'=\sum_{\mathbf{q}}v_{\mathbf{q}}{}'=\frac{1}{NS}\sum_{\mathbf{q}}(\langle a_{\mathbf{q}}{}^{\dagger}a_{\mathbf{q}}\rangle+\langle a_{\mathbf{q}}a_{\mathbf{q}}{}^{\dagger}\rangle). \quad (2.10')$$

The magnon Hamiltonian (2.12) is now definitely defined, and we can calculate thermodynamic quantities. The magnon energy in FSW, $\hbar\omega_{F\mathbf{k}}$, is given by

$$\hbar\omega_{F\mathbf{k}}=[(H_{1\mathbf{k}}{}^{F})^2-(H_{2\mathbf{k}}{}^{F})^2]^{1/2}. \quad (2.15)$$

The FSW is described well in Kubo's paper.[2]

B. Random-Phase Approximation (RPA)

In the conventional RPA, the average $\langle\ \rangle$ in (2.7) is taken on the basis of the unperturbed Hamiltonian H_1. Thus,

$$\langle\cdots\rangle=\mathrm{Tr}\cdots e^{-\beta H_1}/\mathrm{Tr}\ e^{-\beta H_1}, \quad (2.16)$$

where $\beta=1/k_BT$, k_B being the Boltzman constant. In this paper, we call the approximation of (2.16) the RPA. Then, the magnon Hamiltonian (2.8) or (2.12) is easily defined. $H_{1\mathbf{k}}$ and $H_{2\mathbf{k}}$ in these equations, which will be written as $H_{1\mathbf{k}}{}^{R}$ and $H_{2\mathbf{k}}{}^{R}$, respectively, are

$$H_{1\mathbf{k}}{}^{R}=2JSz[(1-u_R-w_R)+\theta(1-u_R-v_R)], \quad (2.17a)$$

$$H_{2\mathbf{k}}{}^{R}=2JSz(1-u_R-w_R)\gamma_{\mathbf{k}}, \quad (2.17b)$$

where $v_R(=u_R+1/2S)$, and w_R are given by

$$v_R=\sum_{\mathbf{k}}v_{R\mathbf{k}}=\frac{1}{NS}\sum_{\mathbf{k}}\frac{H_{1\mathbf{k}}{}^{F}}{\hbar\omega_{F\mathbf{k}}}\left(1+\frac{2}{(e^{\beta\hbar\omega\mathbf{k}}-1)}\right), \quad (2.18a)$$

$$w_R=\sum_{\mathbf{k}}w_{R\mathbf{k}}\gamma_{\mathbf{k}}=-\frac{1}{NS}\sum_{\mathbf{k}}\frac{H_{2\mathbf{k}}{}^{F}\gamma_{\mathbf{k}}}{\hbar\omega_{F\mathbf{k}}}\left(1+\frac{2}{(e^{\beta\hbar\omega\mathbf{k}}-1)}\right). \quad (2.18b)$$

The magnon energy in RPA, $\hbar\omega_{R\mathbf{k}}$, is given by

$$\hbar\omega_{R\mathbf{k}}=[(H_{1\mathbf{k}}{}^{R})^2-(H_{2\mathbf{k}}{}^{R})^2]^{1/2}. \quad (2.19)$$

AFMR is given by $\hbar\omega_{R\mathbf{k}=0}$, which can be calculated from (2.19).

C. Magnon-Renormalization Approximation (MRA)

If the unknown parameters $v_{\mathbf{q}}{}'$ and $w_{\mathbf{q}}{}'$ in the magnon Hamiltonian of (2.8) or (2.12) are determined self-consistently, this approximation is called the MRA, which was discussed rather precisely in the previous paper.[14] In the MRA, $v_{\mathbf{q}}{}'$ and $w_{\mathbf{q}}{}'$, which will be written

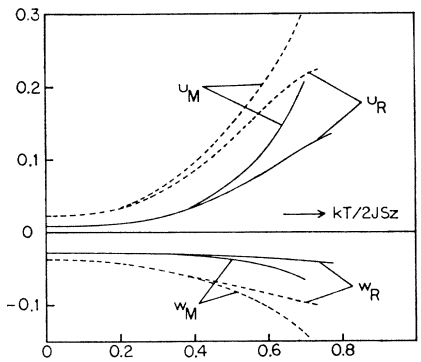

FIG. 1. Dependence of v_R, w_R, v_M, and w_M upon the reduced temperature $k_BT/2JSz$. Solid line, $S=2.0$ and $\theta=0.44$. Dashed line, $S=2.5$ and $\theta=0.02$.

as v_{Mq} and w_{Mq}, respectively, are given by

$$v_{Mk}=\frac{1}{NS}\frac{{H_{1k}}^M}{\hbar\omega_{Mk}}\left(1+\frac{2}{(e^{\beta\hbar\omega_{Mk}}-1)}\right), \quad (2.20a)$$

$$w_{Mk}=-\frac{1}{NS}\frac{{H_{2k}}^M}{\hbar\omega_{Mk}}\left(1+\frac{2}{(e^{\beta\hbar\omega_{Mk}}-1)}\right), \quad (2.20b)$$

where

$$\hbar\omega_{Mk}=[({H_{1k}}^M)^2-({H_{2k}}^M)^2]^{1/2}, \quad (2.21)$$

$${H_{1k}}^M=2JSz[(1-u_M-w_M)+\theta(1-u_M-v_M)], \quad (2.22a)$$

$${H_{2k}}^M=2JSz(1-u_M-w_M)\gamma_k, \quad (2.22b)$$

with $u_M=\sum_q u_{Mq}$, $v_M=\sum_q v_{Mq}$, and $w_M=\sum_q\gamma_q w_{Mq}$. In the case of uniaxial anisotropy, ${H_{1k}}^M$ and ${H_{2k}}^M$ are functions of $u_M(=v_M-\frac{1}{2}S)$ and w_M. Hence, we have only two equations to be solved self-consistently,

$$v_M=\frac{1}{NS}\sum_k\frac{{H_{1k}}^M}{\hbar\omega_{Mk}}\left(1+\frac{2}{(e^{\beta\hbar\omega_{Mk}}-1)}\right), \quad (2.23a)$$

$$w_M=-\frac{1}{NS}\sum_k\frac{\gamma_k{H_{2k}}^M}{\hbar\omega_{Mk}}\left(1+\frac{2}{(e^{\beta\hbar\omega_{Mk}}-1)}\right). \quad (2.23b)$$

We show the temperature dependences of v_M and w_M together with those of v_R and w_R for the cases of $\theta=0.44$ and $S=2.0$ and $\theta=0.02$ and $S=2.5$ in Fig. 1.[17]

III. NUMERICAL COMPUTATION AND COMPARISON WITH EXPERIMENT

In this section, first we calculate the AFMR, sublattice magnetization, and magnetic specific heat of FeF_2 in the various approximations, and we compare them with experiments. Secondly, we consider the spin-wave theory of MnF_2, whose anisotropy is mainly of the interionic dipolar interaction. The mathematical treatment in this case is almost parallel to that in the case of FeF_2, although it is somewhat complicated. We also calculate the above-mentioned physical quantities in the case of this substance and compare them with experiments.

A. FeF_2

FeF_2 is a typical antiferromagnetic substance with a Néel temperature of 78.4°K. The crystal structure of this substance is of the rutile type: The unit cell of Fe^{2+} ions may be conveniently pictured as a body-centered cube compressed along the z axis. In the antiferromagnetic state, the spins on body-centered sites are antiparallel to those on corner sites, the spin direction being along the tetragonal axis.

According to the experiment by Owen *et al.*,[18] the exchange interaction within the same sublattice for Mn^{2+} in ZnF_2 is much weaker than that between the corner and body-centered sites. If this is also the case for FeF_2, we may neglect the exchange interaction within the same sublattice. The spin energy of Fe^{2+} in ZnF_2 was determined by Tinkham[19] to be

$$H_{an}=-DS_z^2+E(S_x^2-S_y^2),$$

with $D=7.3$ cm^{-1} and $E=0.70$ cm^{-1}. The sign of E is opposite for different sublattices. Because of both its small size and alternating sign, we may neglect the E term in the present calculation. The contribution of the interionic magnetic dipolar interaction to the anisotropy energy is smaller than 1 cm^{-1}, which amounts to 5% of the observed anisotropy energy, and so may be also neglected. Thus, the total Hamiltonian for FeF_2 may be given by (2.1), in which we have $S=2$, $z=8$, and

$$\gamma_k=\cos(\tfrac{1}{2}k_xa)\cos(\tfrac{1}{2}k_ya)\cos(\tfrac{1}{2}k_zc). \quad (3.1)$$

The calculation of the thermodynamic quantities is a rather simple matter. Therefore, we shall make a few remarks prior to performing numerical computations.

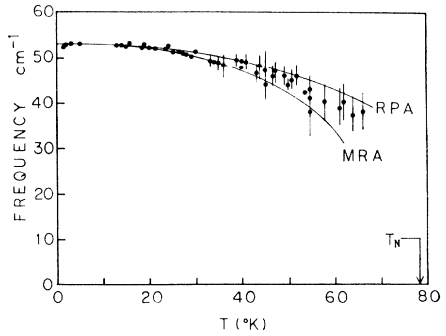

FIG. 2. AFMR of FeF_2. The solid lines represent the theoretical results in various approximations. The black circles denote the experimental data.

[17] For the numerical calculation of v_M and w_M, see Ref. 14.

[18] J. Owen, M. R. Brown, and R. Stevenson, in *Proceedings of the International Conference on Magnetism and Crystallography* (The Physical Society of Japan, Kyoto, Bun Kyo-Ru-Tokyo, 1962), p. 428.

[19] M. Tinkham, Proc. Roy. Soc. (London) **A236**, 535 (1956).

TABLE I. Calculated values of AFMR in RPA and in MRA. The J values are obtained by equating the calculated AFMR in RPA, $\hbar\omega_{R0}$, to the experimental value $\hbar\omega_{\text{expt}}=75.9\ k_B$. $S=2.0$ and $z=8$.

θ	0.40	0.44	0.48
$\hbar\omega_{R0}/2JSz$	0.8231	0.8694	0.9143
$\hbar\omega_{M0}/2JSz$	0.8229	0.8692	0.9141
J/k_B	2.88	2.73	2.60
D/k_B	9.22	9.66	10.0

The expression of the sublattice magnetization is given by

$$M(T)=(\tfrac{1}{2}NS)(1+1/2S-\sum_{\mathbf{k}} v_{\mathbf{k}}), \tag{3.2}$$

where $v_{\mathbf{k}}$ is the thermal average of $(NS)^{-1}\ (a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+a_{\mathbf{k}}a_{\mathbf{k}}^{\dagger})$ on the basis of the total Hamiltonian $\mathbf{H}$, and it is calculated as

$$v_{\mathbf{k}}=\frac{1}{NS}\frac{H_{1\mathbf{k}}}{\hbar\omega_k}\left(1+\frac{2}{(e^{\beta\hbar\omega k}-1)}\right). \tag{3.3}$$

In FSW, $\hbar\omega_{\mathbf{k}}$ and $H_{1\mathbf{k}}$ are, respectively, replaced by $\hbar\omega_{F\mathbf{k}}$ and $H_{1\mathbf{k}}{}^{F}$ defined in (2.15) and (2.11a). Similarly, $\hbar\omega_{\mathbf{k}}$ and $H_{1\mathbf{k}}$ are respectively replaced by $\hbar\omega_{R\mathbf{k}}$ and $H_{1\mathbf{k}}{}^{F}$ defined in (2.19) and (2.17a) in RPA and by $\hbar\omega_{M\mathbf{k}}$ and $H_{1\mathbf{k}}{}^{M}$ defined in (2.21) and (2.22a) in MRA.

The magnetic specific heat is calculated by differentiating the internal energy E with respect to the temperature. E is calculated from (2.8) or (2.12), and it is given by

$$E=NS\sum_{\mathbf{k}}[H_{1\mathbf{k}}u_{\mathbf{k}}+H_{2\mathbf{k}}w_{\mathbf{k}}-\tfrac{1}{2}(H_{1\mathbf{k}}-H_{1\mathbf{k}}{}^{F})u_{\mathbf{k}}'-\tfrac{1}{2}(H_{2\mathbf{k}}-H_{2\mathbf{k}}{}^{F})w_{\mathbf{k}}']. \tag{3.4}$$

Here $u_{\mathbf{k}}=v_{\mathbf{k}}-NS^{-1}$ is given by (3.3), and $w_{\mathbf{k}}$, which is the thermal average of $(NS)^{-1}\ (a_{\mathbf{k}}b_{\mathbf{k}}+a_{\mathbf{k}}{}^{\dagger}b_{\mathbf{k}}{}^{\dagger})$ on the basis of the total Hamiltonian, is calculated as

$$w_{\mathbf{k}}=-\frac{1}{NS}\frac{H_{2k}}{\hbar\omega_{\mathbf{k}}}\left(1+\frac{2}{(e^{\beta\hbar\omega k}-1)}\right). \tag{3.5}$$

Thus, the magnetic specific heat is calculated by the following equation:

$$C=dE/dT=(NS)(2JSz)(d/dT)\times[(u+w)-(u+w)(u'+w')+\tfrac{1}{2}(u'+w')^2+\theta(u-uu'-uv'+u'^2)], \tag{3.6}$$

where $u=\sum_{\mathbf{k}} u_{\mathbf{k}}$ and $w=\sum_{\mathbf{k}}\gamma_{\mathbf{k}}w_{\mathbf{k}}$.

In order to determine the magnitudes of J and D, we calculated the AFMR in RPA at 0°K for several values of θ. Some of the results are shown in Table I. By equating the theoretical values of AFMR, $\hbar\omega_{R0}$, with the experimental value $\hbar\omega_{\text{expt}}=75.9\ k_B$,[20] we calculated the magnitudes of J, which are tabulated in this Table.

[20] R. C. Ohlman and M. Tinkham, Phys. Rev. **123**, 425 (1961).

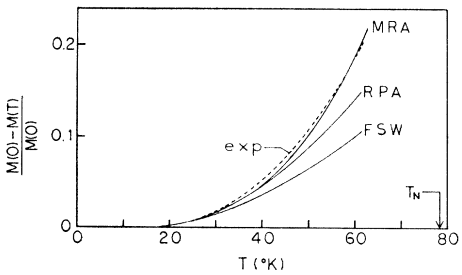

FIG. 3. Deviation of the sublattice magnetization of FeF_2. The dashed line denotes the experimental result.

We also calculated AFMR in MRA, $\hbar\omega_{M0}$, at 0°K, which is very close to $\hbar\omega_{R0}$ value. The J values obtained in MRA are also close to those in RPA, and they are not tabulated. Referring to the previous data $J=2.77\ k_B$[21] and $D=9.7\ k_B$,[22] we assume in this paper that the magnitudes of J and D are given by

$$J=2.73\ k_B \quad \text{and} \quad D=9.66\ \text{k}_B, \tag{3.7}$$

which leads to $\theta=0.44$. The AFMR in RPA and that in MRA at finite temperatures are calculated, and they are shown in Fig. 2, together with the experimental data by Ohlman and Tinkham.[20] It may be interesting to note that the experimental data exist between RPA and MRA calculations.

In Fig. 3, we show the calculation of the temperature dependence of the sublattice magnetization $M(T)$ in various approximations. The agreement between theory and experiment is rather poor.

The magnetic specific heat C is also calculated.[23] The MRA calculation seems to agree with the experiment[24] satisfactorily, as can be seen in Fig. 4.

B. MnF_2

MnF_2 is crystallographically and magnetically isomorphous with FeF_2. The lattice parameters, as determined from x-ray diffraction,[25] are $c=3.3103$ A and $a=4.8734$ A. According to Keffer,[26] the major part of the measured anisotropy in the susceptibility above the Néel point (68°K) in MnF_2 can be accounted for by magnetic dipole interactions, and the remainder is possibly because of the interactions of individual paramagnetic ions with their surrounding crystalline fields. Keffer estimated the anisotropy field at 0°K to be 8800 Oe, which consists of a dipolar anisotropy field of

[21] S. Foner, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (The Institute of Physics and The Physical Society, London, 1965), p. 438; also see M. E. Lines, Phys. Rev. **156**, 543 (1967).

[22] J. Kanamori and H. Minatono, J. Phys. Soc. Japan **17**, 1759 (1962).

[23] The numerical calculation of C was done as follows: $dE/dT=(k_B/2JSz)(\delta E/\delta T^*)$, where $T^*=k_BT/2JSz$ and $\delta T^*=0.01$ was used.

[24] J. W. Stout and E. Catalano, J. Chem. Phys. **23**, 2013 (1955).

[25] M. Griffel and J. W. Stout, J. Am. Chem. Soc. **72**, 4351 (1950).

[26] F. Keffer, Phys. Rev. **87**, 608 (1952).

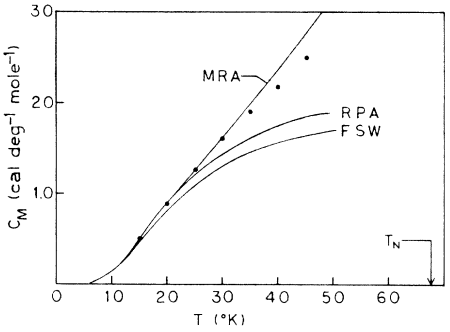

FIG. 4. Magnetic specific heat of FeF_2. The black circles denote the experimental data.

8300 Oe and a crystalline anisotropy field of 500 Oe. Thus, the crystalline anisotropy field amounts to 6% of the total anisotropy field.

Following Low *et al.*,[27] we take into account two kinds of exchange interactions. One is the antiferromagnetic exchange interaction, with the coupling constant J, between the nearest-neighboring intersublattice spins along the ⟨111⟩ directions, and the other is the ferromagnetic exchange interaction, with the coupling constant J', between the nearest-neighboring intrasublattice spins along the c axis.

The Heisenberg spin Hamiltonian of MnF_2 may be written as follows:

$$H=2J\sum_{\langle j,m\rangle}\mathbf{S}_j\cdot\mathbf{S}_m-2J'[\sum_{\langle j,j'\rangle}\mathbf{S}_j\cdot\mathbf{S}_{j'}+\sum_{\langle m,m'\rangle}\mathbf{S}_m\cdot\mathbf{S}_{m'}]$$
$$-D[\sum_j(S_j{}^z)^2+\sum_m(S_m{}^z)^2]+H_{\rm dip}, \quad (3.8)$$

where $\sum_{\langle j,j'\rangle}$ (or $\sum_{\langle m,m'\rangle}$) denotes a summation over pairs j and j' (or m and m') on the same sublattice and $H_{\rm dip}$ the dipole-dipole interaction, which is given by

$$H_{\rm dip}=(2\mu_B)^2\sum_{i,n}R_{in}{}^{-5}[R_{in}{}^2(\mathbf{S}_i\cdot\mathbf{S}_n)$$
$$-3(\mathbf{S}_i\cdot\mathbf{R}_{in})(\mathbf{S}_n\cdot\mathbf{R}_{in})]. \quad (3.9)$$

Here μ_B denotes the Bohr magneton, R_{in} is the distance between spins i and n, and $\sum_{i,n}$ are taken over pairs i and n.[28] Other notations are defined in Sec. II.

Using the method described in Sec. II and carrying out some manipulation which will be given in the Appendix, we obtain the following expression for the simplified magnon Hamiltonian:

$$H=\sum_{\mathbf{k}}[H_{1\mathbf{k}}(a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}+b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}})+H_{2\mathbf{k}}(a_{\mathbf{k}}b_{\mathbf{k}}+a_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}{}^\dagger)], \quad (3.10)$$

where

$$H_{1\mathbf{k}}=2JSz\{(1-u'-w')+\theta(1-u'-v')$$
$$+\epsilon(1-\gamma_{1\mathbf{k}})[1-\sum_{\mathbf{q}}u_{\mathbf{q}}'(1-\gamma_{1\mathbf{q}})]$$
$$+(G_0{}^z-F_0{}^z+\tfrac{1}{2}G_{\mathbf{k}}{}^z)(1-u')$$
$$-\sum_{\mathbf{q}}[u_{\mathbf{q}}'(\tfrac{1}{2}G_{\mathbf{q}}{}^z+G_{\mathbf{k}-\mathbf{q}}{}^z)+\tfrac{1}{2}w_{\mathbf{q}}'F_{\mathbf{q}}{}^z]\}, \quad (3.11a)$$

$$H_{2\mathbf{k}}=2JSz[\gamma_{\mathbf{k}}(1-u'-w')+\tfrac{1}{2}F_{\mathbf{k}}{}^z(1-u')$$
$$+\gamma_{\mathbf{k}}\sum_{\mathbf{q}}w_{\mathbf{q}}'F_{\mathbf{q}}{}^z]. \quad (3.11b)$$

We omitted the c-number terms in (3.10) for simplicity. Here $u_{\mathbf{q}}'$, etc., are defined by (2.10a), etc., and other notations are defined as follows:

$$\epsilon=(J'z'/Jz)=(J'/4J), \quad (3.12a)$$

$$G_{\mathbf{k}}{}^z=-(2\mu_B{}^2/zJ)\sum_{j-j'}R_{jj'}{}^{-5}(R_{jj'}{}^2-3Z_{jj'}{}^2)$$
$$\times e^{i\mathbf{k}\cdot R_{jj'}}, \quad (3.12b)$$

$$F_{\mathbf{k}}{}^z=-(2\mu_B{}^2/zJ)\sum_{m-j}R_{mj}{}^{-5}(R_{mj}{}^2-3Z_{mj}{}^2)$$
$$\times e^{i\mathbf{k}\cdot R_{mj}}, \quad (3.12c)$$

$$z'=2, \quad \text{and} \quad \gamma_{1\mathbf{k}}=\cos(k_zc). \quad (3.12d)$$

The magnon Hamiltonian (3.10) is written in the same form with that of (2.8) in Sec. III. Henceforth, we shall often refer to the formulas derived in Sec. II.

The magnon energy is simply given by

$$\hbar\omega_{\mathbf{k}}=[(H_{1\mathbf{k}}+H_{2\mathbf{k}})(H_{1\mathbf{k}}-H_{2\mathbf{k}})]^{1/2}. \quad (3.13)$$

Before going to the numerical computation, we will give a remark on the calculation of $\sum_{\mathbf{q}}u_{\mathbf{q}}'G_{\mathbf{q}}{}^z$ and $\sum_{\mathbf{q}}w_{\mathbf{q}}'F_{\mathbf{q}}{}^z$. We write $G_{\mathbf{q}}{}^z$ and $F_{\mathbf{q}}{}^z$ as

$$G_{\mathbf{q}}{}^z=-(2\mu_B{}^2/zJa^2c)G(\mathbf{q}), \quad (3.14a)$$

$$F_{\mathbf{q}}{}^z=-(2\mu_B{}^2/zJa^2c)F(\mathbf{q}), \quad (3.14b)$$

TABLE II. Numerical values of $G_n(0)$ and expressions for $g_n(\mathbf{q})$. In the numerical calculation which is described in the text, the summation over n was taken within the sphere with the radius of 9.2 A, which is denoted by an arrow.

n	Number of neighbors	$G_n(0)$	$\sum_n G_n(0)$[a]	$g_n(\mathbf{q})$[b]
1	2	−8.6694	−8.6694	g_{001}
2	4	2.7170	−5.9524	$\frac{1}{2}(g_{100}+g_{010})$
3	8	0.1624	−5.7900	$\frac{1}{2}(g_{101}+g_{011})$
4	2	−1.0836	−6.8736	g_{002}
5	4	0.9606	−5.9130	g_{110}
6	8	0.6160	−5.2970	g_{111}
7	8	−1.0704	−6.3674	$\frac{1}{2}(g_{102}+g_{012})$
→				
8	8	−0.3168	−6.6842	g_{112}
9	4	0.3396	−6.3446	$\frac{1}{2}(g_{200}+g_{020})$
10	4	−0.3211	−6.6657	g_{003}
11	8	0.4000	−6.2657	$\frac{1}{2}(g_{201}+g_{021})$

[a] Keffer value = −6.16 [F. Keffer, Phys. Rev. **87**, 608 (1952)].
[b] $g_{lmn}=\cos lq_xa\cos mq_ya\cos nq_zc$.

[27] G. G. Low, A. Okazaki, R. W. H. Stevenson, and K. C. Turberfield, J. Appl. Phys. **35**, 998 (1964); also, A. Okazaki, K. C. Turberfield, and R. W. H. Stevenson, Phys. Letters **8**, 9 (1964).

[28] For the spin-wave calculation of the dipole-dipole interaction in antiferromagnets, see Ref. 9. Also, see J. Van Kranendonk and J. H. Van Vleck, Rev. Mod. Phys. **30**, 1 (1958).

respectively, where

$$G(\mathbf{q})=a^2c\sum_{j-j'}R_{jj'}{}^{-5}(R_{jj'}{}^2-3Z_{jj'}{}^2)e^{i\mathbf{q}\cdot\mathbf{R}_{jj'}}, \quad (3.15a)$$

$$F(\mathbf{q})=a^2c\sum_{m-j}R_{mj}{}^{-5}(R_{mj}{}^2-3Z_{mj}{}^2)e^{i\mathbf{q}\cdot\mathbf{R}_{mj}}. \quad (3.15b)$$

Then, let us write $G(\mathbf{q})$ and $F(\mathbf{q})$ as

$$G(\mathbf{q})=\sum_n G_n(0)g_n(\mathbf{q}) \quad \text{and} \quad F(\mathbf{q})=\sum_n F_n(0)f_n(\mathbf{q}), \quad (3.16)$$

where $G_n(0)g_n(\mathbf{q})$ denotes the contribution from the nth-neighbor spins in the same sublattice and $F_n(0)f_n(\mathbf{q})$ from tke nth-neighbor spins in the different sublattice. Here $g_n(0)=f_n(0)=1$. For several neighbors, $G_n(0)$ and $g_n(\mathbf{q})$ are tabulated in Table II and $F_n(0)$ and $f_n(\mathbf{q})$ in Table III. We remark that $\sum_\mathbf{q} u_\mathbf{q}{}'g_n(\mathbf{q})$ and $\sum_\mathbf{q} w_\mathbf{q}{}'f_n(\mathbf{q})$ are written as follows:

$$U_n=\sum_\mathbf{q} u_\mathbf{q}{}'g_n(\mathbf{q})=(1/2S^2)\langle S_j{}^xS_{j'}{}^x\rangle, \quad (3.17a)$$

$$W_n=\sum_\mathbf{q} w_\mathbf{q}{}'f_n(\mathbf{q})=(1/2S^2)\langle S_j{}^xS_m{}^x\rangle, \quad (3.17b)$$

where $\langle S_j{}^xS_{j'}{}^x\rangle$ denotes the correlation function of the two spins belonging to the same sublattice and $\langle S_j{}^xS_m{}^x\rangle$ the correlation function of the two spins belonging to the different sublattices. The subscript n denotes $j-j'$ or $j-m$ symbolically. Finally, we can write

$$\sum_\mathbf{q} u_\mathbf{q}{}'G_\mathbf{q}{}^z=-(2\mu_B{}^2/zJa^2c)\sum_n G_n(0)U_n, \quad (3.18a)$$

$$\sum_\mathbf{q} w_\mathbf{q}{}'F_\mathbf{q}{}^z=-(2\mu_B{}^2/zJa^2c)\sum_n F_n(0)W_n. \quad (3.18b)$$

1. FSW

Inserting zero into $u_\mathbf{q}{}'$ and $w_\mathbf{q}{}'$ in $H_{1\mathbf{k}}$ and $H_{2\mathbf{k}}$, we obtain the well-known expressions.[28] The dipolar anisotropy term in the magnon energy depends on the wave vector $\mathbf{k}$. However, for finite $\mathbf{k}$ values, the exchange energy term is much larger than the dipolar term in the case of MnF_2. Then, we can neglect the $\mathbf{k}$ dependence of the anisotropy term, and thus the dipolar anisotropy energy can be replaced by the uniaxial anisotropy energy in the Hamiltonian.

TABLE III. Numerical values of $F_n(0)$ and expressions for $f_n(\mathbf{q})$.

n	Number of neighbors	$F_n(0)$	$\sum_n F_n(0)$ [a]	$f_n(\mathbf{q})$ [b]
1	8	4.9268	4.9268	f_{111}
2	8	−2.9192	2.0076	f_{113}
3	16	2.2296	4.2372	$1(f_{311}+f_{131})$
4	8	−1.3600	2.8772	f_{115}
5	16	0.1956	3.0728	$\frac{1}{2}(f_{133}+f_{313})$
→				
6	8	0.5096	3.5824	f_{331}
7	16	−0.5280	3.0544	$\frac{1}{2}(f_{135}+f_{315})$
8	8	0.1825	3.2369	f_{333}

[a] Keffer value = 3.23 [F. Keffer, Phys. Rev. **87**, 608 (1952)].
[b] $f_{lmn}=\cos\frac{1}{2}lq_xa\cos\frac{1}{2}mq_ya\cos\frac{1}{2}nq_zc$.

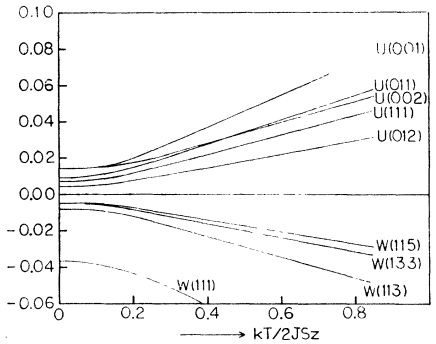

FIG. 5. Temperature dependence of U_n and W_n, which are defined by (3.17a) and (3.17b), respectively, in RPA for $S=2.5$ and $\theta=0.02$. $U(la,ma,nc)$ and $W(\frac{1}{2}la,\frac{1}{2}ma,\frac{1}{2}nc)$ are denoted by $U(lmn)$ and $W(lmn)$, respectively, in the figure. The temperature dependence of U_n and W_n in MRA is also calculated, and a similar temperature dependence to that shown in Fig. 1 is found, which is not shown in this figure.

The value for J used in the present calculation is 1.76 k_B, which corresponds to the figure derived by Trapp and Stout on the basis of an accurate measurement of the perpendicular susceptibility of MnF_2.[29] Low *et al.*[27] deduced the magnitude of J' to be (0.3 ± 0.1) k_B, of which the effect on the thermodynamic properties is supposed to be rather small. The above value of J taken together with the low-temperature AFMR determined by Johnson and Nethercot[30] leads to the θ value of 0.020. These values for J and J' will be also used in the following calculations.

2. RPA

In this approximation, $u_\mathbf{q}{}'$ and $w_\mathbf{q}{}'$, which will be denoted by $u_{R\mathbf{q}}$ and $w_{R\mathbf{q}}$, are calculated by using the FSW, as defined in Sec. II. Since we can assume crystalline anisotropy instead of dipolar anisotropy, $u_{R\mathbf{q}}$ and $w_{R\mathbf{q}}$ can then be calculated from (2.18a) and (2.18b), respectively. Using the θ value of 0.020, we calculated U_n and W_n, which are shown in Fig. 5.

AFMR is given by the following expression:

$$\begin{aligned}\hbar\omega_{R0}&=[(H_{10}+H_{20})(H_{10}-H_{20})]^{1/2}\\&\cong(2JSz)\{2(1-u_R-w_R)\\&\quad\times[\theta(1-u_R-v_R)+\tfrac{3}{2}(G_0{}^z-F_0{}^z)(1-u_R)\\&\qquad-\tfrac{3}{2}\sum_\mathbf{q}(u_{R\mathbf{q}}G_\mathbf{q}{}^z+w_{R\mathbf{q}}F_\mathbf{q}{}^z)]\}^{1/2}.\end{aligned}$$

The calculated result of $\hbar\omega_{R0}$ is shown in Fig. 6. In this figure, RPA(dipolar) stands for the calculation in which only the dipolar anisotropy energy is taken into account, and the uniaxial anisotropy energy is neglected. On the other hand, RPA(uniaxial) denotes the calculation in

[29] C. Trapp and J. W. Stout, Phys. Rev. Letters **10**, 157 (1963).
[30] F. M. Johnson and H. Nethercot, Phys. Rev. **114**, 705 (1959).

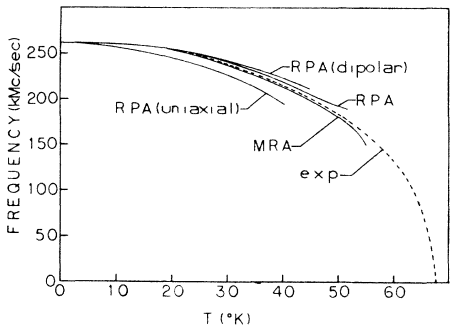

FIG. 6. AFMR of MnF_2. The solid lines represent the theoretical results in various approximations. See the text for definitions of these lines. The dashed line denotes the experimental result.

which only the uniaxial crystalline anisotropy energy is considered. By properly accounting for both the dipolar and uniaxial anisotropy energies, the experimental data can be fitted with the theory up to 50°K. If we assume 6% of the uniaxial anisotropy field and 94% of the dipolar anisotropy field in the total amount of the anisotropy field, which is in accordance with Keffer's estimation,[26] the calculation denoted by RPA is very close to the experiment,[30] although still we find a slight discrepancy.

The sublattice magnetization is defined by (3.2) with (3.3). Here H_{1k} is given by (3.11a) in which u_q' and w_q' are calculated from (2.18a) and (2.18b), respectively. In the calculation of (2.18a) and (2.18b), the crystalline anisotropy was assumed, and the constant θ was assumed to be 0.020. The calculated result is shown in Fig. 7 together with the experimental data.[31,32]

The magnetic specific heat was calculated by use of the formula for E, (3.4), with (3.11a) and (3.11b). The calculated result and the experimental data are shown in Fig. 8.

3. MRA

The self-consistent equations for v_{Mq} and w_{Mq} are given by (2.20a) and (2.20b), where $H_{1k}{}^M$ and $H_{2k}{}^M$ are given by (3.11a) and (3.11b), respectively. In the expression of H_{1k} and H_{2k}, u_q', etc., are replaced by u_{Mq}, etc. Thus, it is tedious to carry out the self-consistent calculation for these unknown parameters in the case of MnF_2. Therefore, we proceed with our calculation on the magnon energy by use of the following approximation. We calculate $v_q'[=u_q'+(NS)^{-1}]$ and w_q', which will be written v_{Mq} and w_{Mq}, assuming the crystalline anisotropy and neglecting the intra-sublattice exchange interaction. That is, we calculate v_M and w_M from (2.23a) and (2.23b) self-consistently. Here θ value is assumed to 0.020. Using these v_M and w_M, we calculate v_{Mq} and w_{Mq} from (2.20a) and (2.20b). We substitute v', w', v_q' and w_q' in (3.11a) and (3.11b) by v_M, w_M, v_{Mq}, and w_{Mq}, respectively.

[31] V. Jaccarino and L. R. Walker, J. Phys. Radium **20**, 341 (1959).

[32] P. Heller and G. B. Benedek, Phys. Rev. Letters **8**, 428 (1962).

Assuming 6% of the crystalline field and 94% of the dipolar field to the total anisotropy field, we computed the temperature dependence of AFMR, which is denoted by MRA in Fig. 6. The experimental values reasonably agree with both RPA and MRA calculations, though the experiment seems to exist between these two approximate calculations.

The sublattice magnetization and magnetic specific heat are calculated by use of the magnon energy which was obtained in the above-mentioned approximation. They are shown in Figs. 7 and 8, respectively.

Finally, we computed the magnon energy at the first Brillouin-zone edge[33] $k_xa=k_y=k_zc=\pi$ by assuming the crystalline anisotropy energy. We also assumed $J'=0$. According to the present calculation, the experimental data seems to exist between both RPA and MRA calculations, as can be seen from Fig. 9.

IV. SUMMARY AND DISCUSSION

We have computed the temperature dependence of the AFMR, sublattice magnetization, and magnetic specific heat of FeF_2 and MnF_2 at moderately high temperatures. We applied the temperature-dependent magnon-energy theory to the calculation of these physical quantities in the cases of RPA and MRA.

By the present calculations, we found the following: The experimental values of AFMR are found between two approximate calculations, RPA and MRA, in both cases of FeF_2 and MnF_2. The temperature dependence of the magnon energy at the first Brillouin-zone edge is calculated in the case of MnF_2. The experimental values are again found between RPA and MRA calculations. The RPA calculation of the sublattice magnetization of MnF_2 fits well with the experiment at high temperature of 0.7 T_N, although we find that the experimental values exist between these two approximate calculations. On the other hand, the sublattice magnetization data on

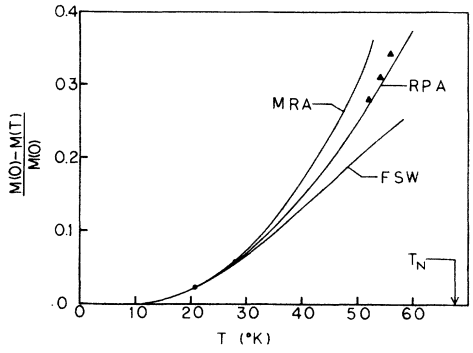

FIG. 7. Deviation of the sublattice magnetization of MnF_2. The black circles denote the experimental data by Jaccarino and Walker (Ref. 31) and the black triangles those by Heller and Benedek (Ref. 32).

[33] R. L. Greene, D. D. Sell, W. M. Yen, A. L. Schawlow, and R. M. White, Phys. Rev. Letters **15**, 656 (1965); also, R. M. White, Phys. Letters **19**, 453 (1965).

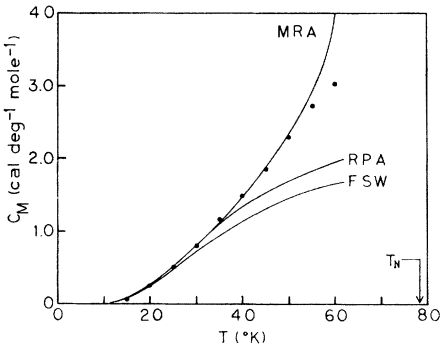

FIG. 8. Magnetic specific heat of MnF_2. The black circles denote the experimental data.

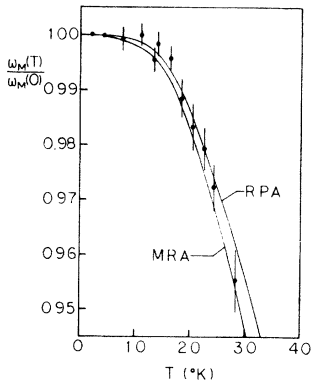

FIG. 9. Comparison of the optical absorption data of Green *et. al* (Ref. 33) with the present theory for MnF_2. $\omega_M(T)$ denotes the magnon energy at the Brillouin-zone edge.

FeF_2 is not found between these two calculations. The MRA calculation of the magnetic specific heat does fit well with the experiment in both cases of FeF_2 and MnF_2. However, the present Hamiltonian is truncated at the quartic terms with respect to the magnon operators, and there may appear some discrepancies between the theory and experiment, if we take into account the higher-order terms which are neglected in this theory. Thus, the present calculation, particularly in the cases of the AFMR and the magnon energy at the zone edge, seems to suggest that the MRA over estimates the magnon interaction effects.

It is known that the anisotropy energy of FeF_2 is very much larger than that of MnF_2. Actually, we assumed that $\theta=0.44$ for FeF_2 and $\theta=0.02$ for MnF_2 in this paper. In the case of infinitely large anisotropy energy, the spin system may be considered as the Ising spin system in which the spin-wave model may not be valid. However, according to the recent two-spin cluster calculation on FeF_2,[34] the Néel temperature is calculated as $T_N=77°K$, which is very close to the Néel temperature of the system with $\theta=0$,[35]

$$T_N(D=0)=76°K.$$

On the other hand, we obtain[36]

$$T_N(D=\infty)=156°K$$

from the equation for T_N of the Ising spin system. Thus, the spins in FeF_2 are well described by the vector spins rather than the Ising spins used in the description of the Néel temperature.

Recently, Murao and Matsubara[37] developed a Green's-function theory of ferromagnetism in which the two types of the excitation spectrum are introduced. One corresponds to the usual collective excitation of the spin-wave type, and the other corresponds to the single-ion excitation of the Weiss molecular-field-theory type. The latter may not be taken into account in the present theory. The effects of the single-ion excitations on the thermodynamic quantities may become appreciable at high temperatures. This remains to be done as a future problem.

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APPENDIX

In this Appendix, we derive the Hamiltonian of (3.10). By use of the procedure described in Sec. II, the spin Hamiltonian of (3.8) can be written[38]

$$H=2JSz\sum_{\mathbf{k}}[H_{1\mathbf{k}}(a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}})+H_{2\mathbf{k}}(a_{\mathbf{k}}b_{\mathbf{k}}+a_{\mathbf{k}}^{\dagger}b_{\mathbf{k}}^{\dagger})+H_{3\mathbf{k}}(a_{\mathbf{k}}^{\dagger}b_{-\mathbf{k}}+a_{\mathbf{k}}b_{-\mathbf{k}}^{\dagger})+H_{4\mathbf{k}}(a_{\mathbf{k}}a_{-\mathbf{k}}+b_{\mathbf{k}}b_{-\mathbf{k}}+a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}+b_{\mathbf{k}}^{\dagger}b_{-\mathbf{k}}^{\dagger})],\quad\text{(A1)}$$

where $H_{1\mathbf{k}}$, etc., are given by

$$H_{1\mathbf{k}}=2JSz\{1+\theta+G_0{}^z-F_0{}^z+\tfrac{1}{2}G_{\mathbf{k}}{}^z+\epsilon(1-\gamma_{1\mathbf{k}})[1-\sum_{\mathbf{q}}u_{\mathbf{q}}'(1-\gamma_{1\mathbf{q}})]-\sum_{\mathbf{q}}[u_{\mathbf{q}}'(1+\theta+G_0{}^z-F_0{}^z+G_{\mathbf{k}-\mathbf{q}}{}^z+\tfrac{1}{2}(G_{\mathbf{k}}{}^z+G_{\mathbf{q}}{}^z))+\theta v_{\mathbf{q}}'+w_{\mathbf{q}}'(\gamma_{\mathbf{q}}+\tfrac{1}{2}F_{\mathbf{q}}{}^z)+\tfrac{1}{2}x_{\mathbf{q}}'F_{\mathbf{q}}{}^{-}+\tfrac{1}{4}y_{\mathbf{q}}'(2G_{\mathbf{q}}{}^{-}+G_{\mathbf{k}}{}^{-})]\},\quad\text{(A2a)}$$

[34] T. Tanaka, L. Libelo, and R. Kligman, Phys. Rev. **171**, 531 (1968).
[35] P. W. Kasteleijn and J. Van Kranendonk, Physica **22**, 367 (1956).
[36] P. W. Kasteleijn, Physica **22**, 387 (1956).
[37] T. Murao and T. Matsubara, J. Phys. Soc. Japan **25**, 352 (1968).
[38] We omitted the c-number terms in (A1) for simplicity. The conclusion in this Appendix does not change in the calculation of the magnetic specific heat.

$$H_{2k}=2JSz\{\gamma_k+\tfrac{1}{2}F_k{}^z-\sum_q[u_q'(\gamma_k+\tfrac{1}{2}F_k{}^z)+w_q'(\gamma_{q-k}-F_{q-k}{}^z)+\tfrac{1}{4}y_q'F_k{}^-]\}, \quad \text{(A2b)}$$

$$H_{3k}=2JSz\{\tfrac{1}{2}F_k{}^--\sum_q[\tfrac{1}{2}u_q'F_k{}^-+x_q'(\gamma_{q-k}-F_{q-k}{}^z)+\tfrac{1}{2}y_q'(\gamma_k+\tfrac{1}{2}F_k{}^z)]\}, \quad \text{(A2c)}$$

$$H_{4k}=2JSz\{\tfrac{1}{4}G_k{}^--\tfrac{1}{2}\epsilon\sum_q y_q'\times[\gamma_{1q-k}-\tfrac{1}{2}(\gamma_{1k}+\gamma_{1q})]-\tfrac{1}{4}\sum_q[u_q'(G_k{}^-+\tfrac{1}{2}G_q{}^-)+\tfrac{1}{2}w_q'F_q{}^-+x_q'(\gamma_q+\tfrac{1}{2}F_q{}^z)+2y_q'(\theta+G_{k-q}{}^z+\tfrac{1}{4}(G_k{}^z+G_q{}^z))]\}. \quad \text{(A2d)}$$

Here the following definitions are used:

$$x_q'=(NS)^{-1}\langle a_qb_{-q}{}^\dagger+a_q{}^\dagger b_{-q}\rangle, \quad \text{(A3a)}$$

$$y_q'=(NS)^{-1}\langle a_qa_{-q}+a_q{}^\dagger a_{-q}{}^\dagger\rangle=(NS)^{-1}\langle b_qb_{-q}+b_q{}^\dagger b_{-q}{}^\dagger\rangle, \quad \text{(A3b)}$$

$$G_k{}^-=(6\mu_B{}^2/zJ)\sum_{j-j'}R_{jj'}{}^{-5}(Y_{jj'}{}^2-X_{jj'}{}^2)e^{i\mathbf{k}\cdot\mathbf{R}_{jj'}}, \quad \text{(A4a)}$$

$$F_k{}^-=(6\mu_B{}^2/zJ)\sum_{m-j}R_{mj}{}^{-5}(Y_{mj}{}^2-X_{mj}{}^2)e^{i\mathbf{k}\cdot\mathbf{R}_{mj}}. \quad \text{(A4b)}$$

Other notations are defined in Sec. II.

Now we diagonalize the magnon Hamiltonian of (A1). Confining our attention to the positive half-space of $\mathbf{k}$, we define the new operators as follows:

$$a_{\pm k}=(1/\sqrt{2})(A_{1k}\pm A_{2k}), \quad b_{\pm k}=(1/\sqrt{2})(B_{1k}\pm B_{2k}). \quad \text{(A5)}$$

Furthermore, we introduce α_{1k}, α_{2k}, α_{3k}, and α_{4k} as follows:

$$\begin{pmatrix}A_{1k}\\B_{1k}\end{pmatrix}=(1/\sqrt{2})(\alpha_{1k}\pm\alpha_{3k}), \quad \begin{pmatrix}A_{2k}\\B_{2k}\end{pmatrix}=(1/\sqrt{2})(\alpha_{2k}\pm\alpha_{4k}). \quad \text{(A6)}$$

Then, we obtain

$$H=\tfrac{1}{2}\sum_{k\geq 0}\sum_{i=1}^{4}[D_{ik}(\alpha_{ik}{}^\dagger\alpha_{ik}+\alpha_{ik}\alpha_{ik}{}^\dagger)+E_{ik}(\alpha_{ik}{}^2+\alpha_{ik}{}^{\dagger 2})], \quad \text{(A7)}$$

where

$$D_{1k}=D_{4k}=H_{1k}+H_{3k}, \quad D_{2k}=D_{3k}=H_{1k}-H_{3k}, \quad E_{1k}=-E_{4k}=H_{2k}+2H_{4k}, \quad E_{2k}=-E_{3k}=H_{2k}-2H_{4k}. \quad \text{(A8)}$$

Finally, introducing the operator c_{ik} and $c_{ik}{}^\dagger$ defined by

$$\alpha_{ik}=\cosh\theta_{ik}c_{ik}-\sinh\theta_{ik}c_{ik}{}^\dagger, \quad \text{(A9)}$$

where

$$\tanh 2\theta_{ik}=E_{ik}/D_{ik}, \quad \text{(A10)}$$

we obtain

$$H=\sum_k\sum_{i=1}^{2}\hbar\omega_{ik}(c_{ik}{}^\dagger c_{ik}+\tfrac{1}{2}), \quad \text{(A11)}$$

where

$$\hbar\omega_{ik}=(D_{ik}{}^2-E_{ik}{}^2)^{1/2}. \quad \text{(A12)}$$

The statistical averages of $(1/NS)(a_k{}^\dagger a_k+a_ka_k{}^\dagger)$, $(1/NS)(a_kb_k+a_k{}^\dagger b_k{}^\dagger)$, $(1/NS)(a_kb_{-k}{}^\dagger+a_k{}^\dagger b_{-k})$, and $(1/NS)(a_ka_{-k}+a_k{}^\dagger a_{-k}{}^\dagger)$ with respect to the magnon Hamiltonian (A11), which are denoted by v_k, w_k, x_k, and y_k, respectively, are calculated as follows:

$$v_k=\left(\frac{1}{2SN}\right)\sum_{i=1}^{2}\cosh 2\theta_{ik}(1+2n_{ik}), \quad \text{(A13a)}$$

$$w_k=\left(-\frac{1}{2SN}\right)\sum_{i=1}^{2}\sinh 2\theta_{ik}(1+2n_{ik}), \quad \text{(A13b)}$$

$$x_k=\left(\frac{1}{2SN}\right)\sum_{i=1}^{2}(-1)^{i+1}\cosh 2\theta_{ik}(1+2n_{ik}), \quad \text{(A13c)}$$

$$y_k=\left(-\frac{1}{2SN}\right)\sum_{i=1}^{2}(-1)^{i+1}\sinh 2\theta_{ik}(1+2n_{ik}), \quad \text{(A13d)}$$

where $n_{ik}=(e^{\beta\hbar\omega_{ik}}-1)^{-1}$. If we assume that the exchange energy is considerably larger than the anisotropy energy, one obtains the following relations:

$$x_k\cong\tfrac{1}{2}F_k{}^-v_k \quad \text{and} \quad y_k\cong-\tfrac{1}{2}G_k{}^-v_k. \quad \text{(A14)}$$

From the inspection of (A4) and (A13), we find that

$$\sum_q G_q{}^-v_q\cong 0 \quad \text{and} \quad \sum_q F_q{}^-v_q\gamma_q\cong 0, \quad \text{(A15)}$$

in both RPA and MRA. Furthermore, we have $G_0{}^-=F_0{}^-=0$. Thus, we obtain

$$H_{30}\cong 0 \quad \text{and} \quad H_{40}\cong 0.$$

If we consider the magnon energy for the finite $\mathbf{k}$ values, the exchange interaction term is larger than the dipolar interaction term, Consequently, we can neglect H_{3k} and H_{4k} terms in the magnon Hamiltonian of (A1), and we have a simplified form of the Hamiltonian (3.10).