sequence during cooling. The I_{AF} state is not found in the $\mathrm{Mn}_{2-x}\mathrm{Cr}_x\mathrm{Sb}$ system. An explanation of why the I_{AF} state is found in one system and not in the other would require a detailed knowledge of how the exchange integrals vary with interatomic distances in the $\mathrm{Mn_2Sb}$ structure. It is relevant to note that in the course of the present studies, negative uniaxial anisotropy was found to exist up to 245°K in $\mathrm{Mn_2Sb}_{1-y}\mathrm{As}_s$, but previous studies[1] have shown that it exists only below 200°K in $\mathrm{Mn}_{2-x}\mathrm{Cr}_x\mathrm{Sb}$. Both the I_F and I_{AF} states are energetically favored only if negative unaxial anisotropy confines the moments to the basal plane. The wider range of negative anisotropy in $\mathrm{Mn_2Sb}_{1-y}\mathrm{As}_y$ is undoubtedly interrelated with the existance of the I_{AF} state.

ACKNOWLEDGMENTS

The crystals used in these experiments were prepared by T. A. Bither and P. H. L. Walter. The x-ray measurements were performed by E. P. Moore.

PHYSICAL REVIEW VOLUME 168, NUMBER 2 10 APRIL 1968

Low-Temperature Behavior of the Anisotropic Heisenberg Antiferromagnet in the Neighborhood of the Magnetic Phase Boundaries

J. Feder and E. Pytte
IBM Zurich Research Laboratory, 8803 Rüschlikon, Switzerland

(Received 16 October 1967)

Using spin-wave theory, including spin-wave interactions to leading order in $1/2S$, the temperature dependence of the critical magnetic field curves between the antiferromagnetic, the flop, and the paramagnetic phases have been calculated for a Heisenberg antiferromagnet with both uniaxial single-ion anisotropy and anisotropic exchange interaction. The free energy has been obtained for all three phases, and the behavior of the specific heat and the susceptibility in the neighborhood of these phase boundaries as a function of magnetic field for a fixed temperature is discussed in detail.

I. INTRODUCTION

In this paper we discuss the low-temperature properties of a uniaxial Heisenberg antiferromagnet. In particular we determine the temperature dependence of the several phase transition boundaries, and discuss the behavior of thermodynamic properties such as the specific heat, the magnetization, and the susceptibility in the neighborhood of these phase boundaries.

The model we consider consists of a simple cubic array of magnetic ions of spin **S**, interacting by a negative nearest-neighbor exchange interaction. The exchange interaction is assumed to be anisotropic favoring alignment along the crystalline z axis. In addition we assume the presence of a uniaxial single-ion anisotropy and an external magnetic field, both parallel to the crystalline z axis. In this model calculation we consider only a simple cubic structure but the results are trivially extended to any other crystal structure which can also be resolved into two sublattices such that the nearest neighbors of an ion on one sublattice lie only on the other sublattice.

In the low-temperature region $T \ll T_N$, where T_N is the Néel temperature, the spins will be antiferromagnetically ordered for sufficiently small magnetic fields. As the field is increased, a phase transition occurs to the flop phase with the spins in a generally transverse direction to the field. As the field is increased further, the average direction of the individual spins will eventually become parallel to the external field. The particular value of the field for which this occurs defines a second phase transition to the paramagnetic (ferromagnetic) phase. About these average directions there will be thermal fluctuations in the form of spin waves.

In the transition from the spin-flop to the paramagnetic phase, the two phases are indistinguishable at the transition and it is therefore of the second order. At the antiferromagnetic spin-flop phase boundary, the phases are clearly distinct and the transition is of first order. Near a first-order transition, metastable superheated and supercooled states will usually be possible.

The general properties of the system described by the Hamiltonian [Eq. (1)] are most simply appreciated by performing a simple molecular-field calculation at zero temperature. We assume that all the spins on the same sublattice α point in the same direction. Then if we call the angles that $\mathbf{S}_\alpha$ and $\mathbf{S}_\beta$ make with the z axis, θ_α and θ_β, respectively, and ϕ the angle between the projections of $\mathbf{S}_\alpha$ and $\mathbf{S}_\beta$ onto the xy plane, the energy in molecular-field theory takes the form

$$\begin{aligned}E(\theta_\alpha,\theta_\beta,\phi)=\tfrac{1}{2}NS^2zJ[&\sin\theta_\alpha\sin\theta_\beta\cos\phi+\cos\theta_\alpha\cos\theta_\beta\\&+(K/J)\cos\theta_\alpha\cos\theta_\beta-(L/zJ)(\cos^2\theta_\alpha+\cos^2\theta_\beta)\\&-(\mu H/SzJ)(\cos\theta_\alpha+\cos\theta_\beta)].\end{aligned}$$

Here, N is the number of lattice sites, S the magnitude of the spin, and z the number of nearest neighbors. J, K, and L are the exchange constant and the anisotropy constants defined in the discussion following Eq. (1).

The equilibrium values for the angles are obtained by minimizing the energy with respect to the angles θ_α, θ_β, and ϕ. We find that E is always minimized by $\phi=\pi$ independent of the magnetic field such that the magnetic field and the magnetization vectors for the two sublattices are coplanar. For magnetic fields H smaller than a particular value H_{c1} the antiferromagnetic state with $\theta_\alpha=0$ and $\theta_\beta=\pi$ minimizes E. For fields satisfying the inequality $H_{c1}<H<H_{c2}$, where H_{c2} is the upper critical field, the spin-flop state with $\theta_\alpha=\theta_\beta$ and $\theta_\alpha\neq 0$ minimizes the energy, while for fields $H>H_{c2}$ the paramagnetic state with $\theta_\alpha=0$ and $\theta_\beta=0$ is the stable configuration.

However, when the field H_{c1} is reached, the energy surface described by $E(\theta_\alpha, \theta_\beta)$ still has a local minimum at the point $\theta_\alpha=0$, $\theta_\beta=\pi$ corresponding to an antiferromagnetic alignment, and this minimum persists up to a somewhat higher superheating field $H_{c1}{}^a$ at which point the local minimum has developed into a saddle point. Similarly for the flop phase when the field is decreased below H_{c1} a local minimum at $\theta_\alpha=\theta_\beta$ and $\theta_\alpha\neq 0$ persists down to a lower supercooling field $H_{c1}{}^f$. At the thermodynamic critical field H_{c1} the local minima at the points $\theta_\alpha=0$, $\theta_\beta=\pi$ and $\theta_\alpha=\theta_\beta$, $\theta_\alpha\neq 0$ have the same value, and H_{c1} is obtained by equating the free energies of the two phases. At $H=H_{c2}$ the energy surface will have a saddle point at $\theta_\alpha=0$ and $\theta_\beta=0$ corresponding to the paramagnetic state which then for higher fields develops into a true minimum.

When the energy surface is locally flat this means that some generalized coordinate finds a vanishing restoring force and the natural frequency of the corresponding dynamical mode vanishes. Thus at the critical field $H_{c1}{}^a$, one of the spin-wave frequencies of the antiferromagnetic phase vanishes. Similarly at $H_{c1}{}^f$, one of the spin-wave frequencies in the flop phase vanishes. For $H<H_{c1}{}^f$, this mode becomes purely imaginary. At the upper critical field H_{c2}, the spin-wave frequency in the paramagnetic phase and one of the spin-wave modes in the flop phase both vanish (although for different k values) and become negative, respectively, for fields smaller and larger than H_{c2}. The behavior of the spin-wave frequencies as a function of magnetic field is shown in Fig. 1.

In the simple molecular-field calculation described above, it turns out that if we consider anisotropic exchange only, then $H_{c1}{}^a=H_{c1}{}^f$. This would mean that there would be a first-order phase transition at this field but no superheating or supercooling. However, in the spin-wave calculation a splitting of H_{c1} results, when spin-wave interactions are taken into account due to zero-point motion of the spin waves such that $H_{c1}{}^a>H_{c1}{}^f$. In this case the hysteresis is entirely due to quantum-mechanical effects.

The present calculation is restricted to the low-temperature region. We use the Holstein-Primakoff[1] transformation to rewrite the Hamiltonian in terms of spin-deviation operators. Spin-wave interactions will be included to the lowest order in an expansion in $1/2S$. The critical-field boundaries will be calculated from the vanishing of the renormalized spin-wave frequencies, and the partition function will be obtained to the order indicated for all three phases. From the partition function all the thermodynamic properties can then be obtained by the standard rules.

In proceeding to the spin-wave picture, we replace the $2S$-dimensional spin space by an infinite-dimensional vector space for the spin-deviation operators. For the ferromagnet it has been shown that any error due to this approximation is exponentially small in the low-temperature region.[2] Unfortunately, no similar proof exists for the antiferromagnet. In fact, recent calculations[3] show that the so-called kinematical interactions for the antiferromagnet yield a small but nonvanishing contribution to the thermodynamic variables even at $T=0$. In particular it was found that for $S=\frac{1}{2}$ the reduction of the sublattice magnetization in the antiferromagnetic phase ($H=0$) due to zero-point motion of the interacting spin waves is substantially reduced by the kinematical interactions. The corrections due to the kinematical interactions will not be considered here. In the paramagnetic (ferromagnetic) phase, these should again be exponentially small in the low-temperature region.

The spin-wave theory has been applied to the antiferromagnet (in the antiferromagnetic phase) by Anderson[4] and by Kubo.[5,6] This work was later extended by Oguchi[7] to include spin-wave interactions. However, in these papers, interest was primarily concentrated on the properties of the system for vanishing external field. No attempt was made to study the behavior of the system near the flop phase boundary. More recently, Wang and Callen[8] applied spin-wave theory (not including spin-wave interactions) to the flop phase and obtained expressions for the critical fields $H_{c1}{}^f$ and H_{c2} at zero temperature in the case of uniaxial single-ion anisotropy. In the paramagnetic phase the spin-wave theory is formally identical to the spin-wave theory in a ferromagnet.

Of other work not based on the spin-wave theory,

---

[1] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1908 (1940).

[2] F. J. Dyson, Phys. Rev. **102**, 1217 (1956); **102**, 1230 (1956).

[3] R. P. Kenan, Phys. Rev. **159**, 430 (1967).

[4] P. W. Anderson, Phys. Rev. **86**, 694 (1952).

[5] R. Kubo, Phys. Rev. **87**, 568 (1952).

[6] R. Kubo, Rev. Mod. Phys. **25**, 344 (1953).

[7] T. Oguchi, Phys. Rev. **117**, 117 (1960).

[8] Y. L. Wang and H. B. Callen, J. Phys. Chem. Solids **25**, 1459 (1964).

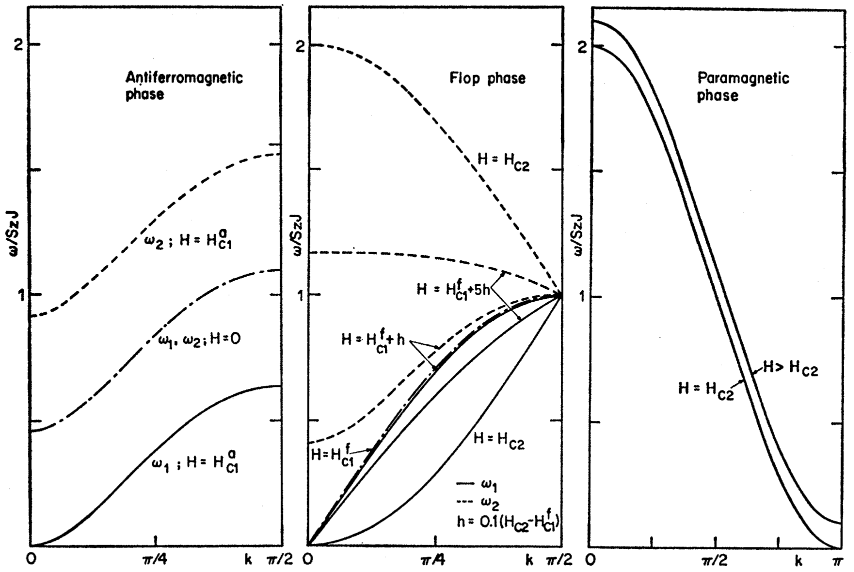

FIG. 1. Zero-order spin-wave spectra with the wave vector **k** along the [111] direction. $K=0.1J$, $L=0$. Note the change in the k scale for the paramagnetic phase.

two calculations are particularly relevant. Falk[9] studied the phase transitions using a variational method valid at low temperature for the case of zero anisotropy but for general spin, and calculated the temperature dependence $H_{c2}(T)$ of the flop-para transition curve. Anderson and Callen[10] used the random-phase approximation and the Callen decoupling method to study the antiferromagnetic and the paramagnetic phases. The flop-para transition curve was calculated for arbitrary temperatures, while the antiferromagnetic-flop critical field H_{c1}^a was calculated explicitly only at zero temperature. Calculations of thermodynamic properties such as the sublattice magnetization and susceptibility were restricted to the case of zero external magnetic field.

When the Callen decoupling method was used the result for the flop-para transition curve agreed in the low-temperature region with the result obtained by Falk.[9] The present spin-wave calculation gives the same result for the leading temperature correction. In higher-order terms differences would, however, appear. In the random-phase approximation (RPA) the coefficient for the leading temperature correction is too small by a factor of 2.

In the case of the zero-temperature value of H_{c1}^a the result obtained using spin-wave theory disagrees both with RPA and the result of the Callen decoupling method.[10] For H_{c1}^f the expression obtained in the present calculation agrees at zero temperature with the calculation of Wang and Callen,[8] apart from zero-point motion corrections.

II. HAMILTONIAN

We shall study the following Hamiltonian:

$$\begin{aligned}\mathcal{H}=J\sum_{\langle\boldsymbol{\alpha},\boldsymbol{\beta}\rangle}\mathbf{S}(\boldsymbol{\alpha})\cdot\mathbf{S}(\boldsymbol{\beta})+K\sum_{\langle\boldsymbol{\alpha},\boldsymbol{\beta}\rangle}S_z(\boldsymbol{\alpha})S_z(\boldsymbol{\beta})\\ -L\sum_{\alpha}S_z^2(\boldsymbol{\alpha})-L\sum_{\beta}S_z^2(\boldsymbol{\beta})\\ -\mu H\sum_{\alpha}S_z(\boldsymbol{\alpha})-\mu H\sum_{\beta}S_z(\boldsymbol{\beta}),\quad(1)\end{aligned}$$

where the sum $\langle\boldsymbol{\alpha},\boldsymbol{\beta}\rangle$ is taken over all nearest-neighbor pairs, and $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are the lattice sites in the α and the β sublattices, respectively. The K term represents the anisotropic exchange interaction and the L term the single-ion uniaxial anisotropy. The external field H is assumed to be applied along the axis of easy magnetization. The form this Hamiltonian takes when rewritten in terms of the spin-deviation operators using the Holstein-Primakoff transformation[1] will be discussed separately for the three phases.

In the antiferromagnetic phase, we introduce two different sets of spin-deviation operators.

These are

$$S^+(\boldsymbol{\alpha})=(2S)^{1/2}f_\alpha(S)a_\alpha,\qquad S^-(\boldsymbol{\alpha})=(2S)^{1/2}a_\alpha^\dagger f_\alpha(S),$$
$$S_z(\boldsymbol{\alpha})=S-a_\alpha^\dagger a_\alpha,$$

for an up-spin on the α sublattice, and

$$S^+(\boldsymbol{\beta})=(2S)^{1/2}b_\beta^\dagger f_\beta(S),\qquad S^-(\boldsymbol{\beta})=(2S)^{1/2}f_\beta(S)b_\beta,$$
$$S_z(\boldsymbol{\beta})=-S+b_\beta^\dagger b_\beta,$$

for a down-spin on the β sublattice, where

$$f_{\alpha,\beta}(S)=(1-n_{\alpha,\beta}/2S)^{1/2},$$

and where the choice of the up and the down sublattice is arbitrary.

---

[9] H. Falk, Phys. Rev. **133**, A1382 (1964).

[10] F. B. Anderson and H. B. Callen, Phys. Rev. **136**, A1068 (1964).

In the flop phase, we first rotate the coordinate system about the x axis an angle θ for sublattice α and an angle $-\theta$ for sublattice β,

$$\begin{aligned}
&S_x(\boldsymbol{\alpha})=\tilde{S}_x(\boldsymbol{\alpha}),\\
&S_y(\boldsymbol{\alpha})=\cos\theta\,\tilde{S}_y(\boldsymbol{\alpha})-\sin\theta\,\tilde{S}_z(\boldsymbol{\alpha}),\\
&S_z(\boldsymbol{\alpha})=\sin\theta\,\tilde{S}_y(\boldsymbol{\alpha})+\cos\theta\,\tilde{S}_z(\boldsymbol{\alpha}),\\
&S_x(\boldsymbol{\beta})=\tilde{S}_x(\boldsymbol{\beta}),\\
&S_y(\boldsymbol{\beta})=\cos\theta\,\tilde{S}_y(\boldsymbol{\beta})+\sin\,\tilde{S}_z(\boldsymbol{\beta}),\\
&S_z(\boldsymbol{\beta})=-\sin\theta\,\tilde{S}_y(\boldsymbol{\beta})+\cos\theta\,\tilde{S}_z(\boldsymbol{\beta}).
\end{aligned}$$

The choice of the x axis is arbitrary. Any axis in the x-y plane could be used, as the whole system is free to rotate about the z axis (but only in such a way that the z axis and the two sublattice magnetizations are coplanar).

For the rotated spin components $\tilde{S}_i$ we now introduce spin-deviation operators for each of the two sublattices,

$$\begin{aligned}
&\tilde{S}^+(\boldsymbol{\alpha})=(2S)^{1/2}f_\alpha(S)a_\alpha, \qquad \tilde{S}^-(\boldsymbol{\alpha})=(2S)^{1/2}a_\alpha{}^\dagger f_\alpha(S),\\
&\tilde{S}_z(\boldsymbol{\alpha})=S-a_\alpha{}^\dagger a_\alpha,\\
&\tilde{S}^+(\boldsymbol{\beta})=(2S)^{1/2}f_\beta(S)b_\beta, \qquad \tilde{S}^-(\boldsymbol{\beta})=(2S)^{1/2}b_\beta{}^\dagger f_\beta(S),\\
&\tilde{S}_z(\boldsymbol{\beta})=S-b_\beta{}^\dagger b_\beta.
\end{aligned}$$

The procedure used for the antiferromagnetic phase is, of course, equivalent to rotating one of the sublattices by π and then introducing the spin-deviation operators.

In the paramagnetic phase there is no need to introduce two sublattices and we simply write

$$\begin{aligned}
&S^+(\boldsymbol{\gamma})=(2S)^{1/2}f_\gamma(S)a_\gamma, \qquad S^-(\boldsymbol{\gamma})=(2S)^{1/2}a_\gamma{}^\dagger f_\gamma(S),\\
&S_z(\boldsymbol{\gamma})=S-a_\gamma{}^\dagger a_\gamma,
\end{aligned}$$

where $\boldsymbol{\gamma}$ refers to any site on the α or β sublattice. The operator $f_\gamma(S)$ will be approximated by the first few terms of the binomial expansion

$$f_\gamma(S)=1-(n_\gamma/4S)-\tfrac{1}{32}(n_\gamma{}^2/S^2)+\cdots.$$

The Hamiltonian will be written in terms of the Fourier-transformed spin-deviation operators defined by

$$a_{\mathbf{k}}=\left(\frac{2}{N}\right)^{1/2}\sum_\alpha a_\alpha e^{-i\mathbf{k}\cdot\boldsymbol{\alpha}}, \qquad b_{\mathbf{k}}=\left(\frac{2}{N}\right)^{1/2}\sum_\beta b_\beta e^{-i\mathbf{k}\cdot\boldsymbol{\beta}},$$

and the adjoint relations

$$a_{\mathbf{k}}{}^\dagger=\left(\frac{2}{N}\right)^{1/2}\sum_\alpha a_\alpha{}^\dagger e^{i\mathbf{k}\cdot\boldsymbol{\alpha}}, \qquad b_{\mathbf{k}}{}^\dagger=\left(\frac{2}{N}\right)^{1/2}\sum_\beta b_\beta{}^\dagger e^{i\mathbf{k}\cdot\boldsymbol{\beta}}.$$

These operators satisfy Bose commutation relations, such that

$$[a_{\mathbf{k}},a_{\mathbf{k}'}{}^\dagger]=\delta_{\mathbf{k},\mathbf{k}'}, \qquad [b_{\mathbf{k}},b_{\mathbf{k}'}{}^\dagger]=\delta_{\mathbf{k},\mathbf{k}'}, \tag{2}$$

while the commutator of all other combinations of operators is zero.

It will be convenient to introduce the dimensionless variables

$$\tilde{K}=K/J, \qquad \tilde{L}=L/zJ, \qquad h=\mu H/SzJ, \qquad \xi^2=1-1/2S,$$

where z is the number of nearest neighbors.

To further simplify the expressions we also define a set of numerical constants,

$$\eta_i=1+a_i\tilde{K}+b_i\tilde{L},$$

where a_i and b_i are constants of order unity. That is, all the η_i's are equal to 1 except for different small corrections from the anisotropy constants $\tilde{K}$ and $\tilde{L}$. The explicit expressions for all the η_i's are tabulated in Appendix B. In terms of these variables the Hamiltonian for the antiferromagnetic phase can be written

$$\mathcal{H}=E_0+\mathcal{H}_0+\mathcal{H}_{\mathrm{I}}, \tag{3}$$

where

$$E_0=-\tfrac{1}{2}NS^2zJ\eta_3, \tag{4}$$

$$\begin{aligned}
\mathcal{H}_0={}&SzJ\sum_{\mathbf{k}}\gamma(\mathbf{k})(a_{\mathbf{k}}b_{-\mathbf{k}}+a_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger)\\
&+SzJ\eta_7\sum_{\mathbf{k}}(a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}+b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}})+\mu H\sum_{\mathbf{k}}(a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}-b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}),
\end{aligned}\tag{5}$$

$$\begin{aligned}
\mathcal{H}_{\mathrm{I}}={}&-\tfrac{1}{4}(zJ)(2/N)\sum_{1,2,3}\gamma(1)(a_2{}^\dagger a_3a_{2-3-1}b_1+b_2{}^\dagger b_3b_{2-3-1}a_1\\
&+a_2{}^\dagger a_3{}^\dagger a_{2+3+1}b_1{}^\dagger+a_1{}^\dagger b_2{}^\dagger b_3{}^\dagger b_{1+2+3})\\
&-zJ\eta_1(2/N)\sum_{1,2,3}\gamma(1)a_{1+2}{}^\dagger b_3{}^\dagger a_2b_{1+3}\\
&-L(2/N)\sum_{1,2,3}(a_1{}^\dagger a_2{}^\dagger a_3a_{1+2-3}+b_1{}^\dagger b_2{}^\dagger b_3b_{1+2-3}).
\end{aligned}\tag{6}$$

Here we have introduced

$$\gamma(k)=z^{-1}\sum_\rho e^{i\mathbf{k}\cdot\boldsymbol{\rho}}, \tag{7}$$

where $\boldsymbol{\rho}$ denotes the vectors to the z nearest neighbors of a given atom. In $\mathcal{H}_{\mathrm{I}}$, $\mathbf{k}_1$, $\mathbf{k}_2$, $\mathbf{k}_3$ have been replaced by the numerical indices 1, 2, 3.

For the flop phase we write similarly,

$$\mathcal{H}=E_0+\mathcal{H}_0+\mathcal{H}_{\mathrm{I}}+\mathcal{H}_{\mathrm{II}}, \tag{8}$$

where

$$E_0=\tfrac{1}{2}NS^2zJ[\eta_4-2\eta_{10}u^2-2h(1-u^2)^{1/2}], \tag{9}$$

$$\begin{aligned}
\mathcal{H}_0=SzJ\{&A\sum_{\mathbf{k}}(a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}+b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}})\\
&+\tfrac{1}{2}B\sum_{\mathbf{k}}(a_{\mathbf{k}}a_{-\mathbf{k}}+a_{\mathbf{k}}{}^\dagger a_{-\mathbf{k}}{}^\dagger+b_{\mathbf{k}}b_{-\mathbf{k}}+b_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger)\\
&+\sum_{\mathbf{k}}d_{\mathbf{k}}(a_{\mathbf{k}}b_{-\mathbf{k}}+a_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger)+\sum_{\mathbf{k}}e_{\mathbf{k}}(a_{\mathbf{k}}b_{\mathbf{k}}{}^\dagger+a_{\mathbf{k}}{}^\dagger b_{\mathbf{k}})\},
\end{aligned}\tag{10}$$

and where in turn,

$$\begin{aligned}
&A=-\eta_8+2\eta_9u^2+h(1-u^2)^{1/2},\\
&B=\tilde{L}\xi u^2,\\
&d_{\mathbf{k}}=\eta_2u^2\gamma(\mathbf{k}),\\
&e_{\mathbf{k}}=(1-\eta_2u^2)\gamma(\mathbf{k}),
\end{aligned}\tag{11}$$

and $u\equiv\sin\theta$.

Furthermore,

$$\begin{aligned}\mathcal{H}_{\mathrm{I}}=&-\tfrac{1}{4}zJ(1-\eta_2u^2)(2/N)\sum_{1,2,3}\gamma(1)(a_2{}^\dagger a_3{}^\dagger a_{2+3-1}b_1\\&+a_1{}^\dagger b_2{}^\dagger b_3b_{1+2-3}+b_1{}^\dagger a_2{}^\dagger a_3a_{1+2-3}+a_1b_2{}^\dagger b_3{}^\dagger b_{-1+2+3})\\&+zJ(\eta_1-2\eta_2u^2)(2/N)\sum_{1,2,3}\gamma(1)a_{1+2}{}^\dagger b_3{}^\dagger a_2b_{1+3}\\&-\tfrac{1}{4}(zJ)\eta_2u^2(2/N)\sum_{1,2,3}\gamma(1)(a_1b_2{}^\dagger b_3b_{-1+2-3}\\&+b_1a_2{}^\dagger a_3a_{-1+2-3}+a_1{}^\dagger b_2{}^\dagger b_3{}^\dagger b_{1+2+3}+b_1{}^\dagger a_2{}^\dagger a_3{}^\dagger a_{1+2+3})\\&-\tfrac{1}{4}Lu^2(2/N)\sum_{1,2,3}(a_1{}^\dagger a_2{}^\dagger a_3{}^\dagger a_{1+2+3}+a_1{}^\dagger a_2a_3a_{1-2-3}\\&+b_1{}^\dagger b_2{}^\dagger b_3{}^\dagger b_{1+2+3}+b_1{}^\dagger b_2b_3b_{1-2-3})-L(1-\tfrac{3}{2}u^2)(2/N)\\&\times\sum_{1,2,3}(a_1{}^\dagger a_2{}^\dagger a_3a_{1+2-3}+b_1{}^\dagger b_2{}^\dagger b_3b_{1+2-3}),\end{aligned}\tag{12}$$

and

$$\begin{aligned}\mathcal{H}_{\mathrm{II}}=&\tfrac{1}{2}i(NS)^{1/2}SzJu\{[h-2\eta_{10}(1-u^2)^{1/2}]\\&\times(a_0-a_0{}^\dagger-b_0+b_0{}^\dagger)+(1/4S)[(h-2\eta_{11}(1-u^2)^{1/2})\\&\times(2/N)\sum_{1,2}(a_1{}^\dagger a_2{}^\dagger a_{1+2}-a_1{}^\dagger a_2a_{1-2}\\&-b_1{}^\dagger b_2{}^\dagger b_{1+2}+b_1{}^\dagger b_2b_{1-2})-8\eta_2(1-u^2)^{1/2}\\&\times(2/N)\sum_{1,2}\gamma(1)(a_1{}^\dagger b_2{}^\dagger b_{1+2}-a_1b_2{}^\dagger b_{2-1}+a_2{}^\dagger a_{2-1}b_1\\&-a_2{}^\dagger a_{1+2}b_1{}^\dagger)]\}.\end{aligned}\tag{13}$$

The terms linear and cubic in the spin-deviation operators are introduced in the flop phase as a consequence of the rotation of the coordinate system. Finally for the paramagnetic phase we write

$$\mathcal{H}=E_0+\mathcal{H}_0+\mathcal{H}_1,\tag{14}$$

where

$$E_0=\tfrac{1}{2}NS^2zJ[\eta_4-2h],\tag{15}$$

$$\mathcal{H}_0=\sum_{\mathbf{k}}\omega(\mathbf{k})a_{\mathrm{k}}{}^\dagger a_{\mathrm{k}},\tag{16}$$

$$\mathcal{H}_1=(1/N)\sum_{1,2,3}f(1,2,3)a_1{}^\dagger a_2{}^\dagger a_3a_{1+2-3},\tag{17}$$

and where

$$\omega(\mathbf{k})=\mu H-SzJ[\eta_8-\gamma(\mathbf{k})],\tag{18}$$

while

$$f(1,2,3)=-\tfrac{1}{4}(zJ)[\gamma(1)+\gamma(3)-2\eta_1\gamma(2-3)+4\tilde{L}].\tag{19}$$

For all three phases only the leading terms in the $1/2S$ expansion have been kept.

For the paramagnetic phase the $\mathbf{k}$ summation runs over the whole crystallographic Brillouin zone, while for the flop and the antiferromagnetic phases the sum is over the smaller Brillouin zone corresponding to the magnetic sublattices. We should also remark that in the case of the L term we have somewhat arbitrarily included in $\mathcal{H}_0$, through the factor ξ, part of the next-order correction in the $1/2S$ expansion. We have done this in order to be able to compare our results directly with those obtained by Wang and Callen.[8] These authors replaced the spin operators by Holstein-Primakoff boson operators in such a way that the matrix elements among the lowest three states of each spin were correctly reproduced. This method introduced the factor ξ, while the usual lowest-order spin-wave theory, which has the matrix elements among the lowest two states correct, would have $\xi=1$.

III. CALCULATION OF THE CRITICAL FIELDS

As already discussed in the Introduction, the phase boundaries will be obtained by determining the magnetic field values for which appropriate spin-wave frequencies go to zero and then become complex or negative. The spin-wave frequencies are obtained by calculating the equation of motion for the spin-deviation operators. The quadratic terms in the Hamiltonian $\mathcal{H}_0$ will give the temperature-independent spin-wave modes from which (apart from small zero-point motion corrections) the critical fields at zero temperature can be determined. The temperature dependence of the phase boundaries, as well as the zero-point motion correction, is obtained by including the interaction Hamiltonian $\mathcal{H}_{\mathrm{I}}$ in calculating the equations of motion. When these equations are linearized by replacing pairs of spin-deviation operators by their thermal expectation values, we obtain the correctly renormalized spin-wave spectra in the low-temperature region.

In the flop phase we have, in addition, terms which are linear and cubic in the spin-deviation operators. The linear terms will contribute a static part to the equations of motion for the spin-deviation operators. Requiring that the static part be zero determines the equilibrium position of the spins, that is, setting the static parts equal to zero determines the value of the angle θ for a given external magnetic field. When the cubic terms are included, the static part will be renormalized, and the angle θ will be temperature-dependent. In the flop phase, therefore, the $\mathcal{H}_{\mathrm{II}}$ part of the Hamiltonian determines the average spin orientation as a function of temperature and magnetic field, while $\mathcal{H}_0$ and $\mathcal{H}_{\mathrm{I}}$ describe the behavior of the spin waves about these equilibrium positions.

A. Antiferromagnetic Phase

In the antiferromagnetic phase the renormalized spin-wave modes are obtained from the coupled set of equations

$$\begin{aligned}i(\partial/\partial t)a_{\mathrm{k}}=&SzJ(\eta_7+h-\Delta_1{}^a)a_{\mathrm{k}}\\&+SzJ(\gamma(\mathbf{k})-\Delta_2{}^a(\mathbf{k}))b_{-\mathrm{k}}{}^\dagger,\end{aligned}\tag{20}$$

$$\begin{aligned}i(\partial/\partial t)b_{-\mathrm{k}}{}^\dagger=&-SzJ(\eta_7-h-\Delta_3{}^a)b_{-\mathrm{k}}{}^\dagger\\&-SzJ(\gamma(\mathbf{k})-\Delta_2{}^a(\mathbf{k}))a_{\mathrm{k}},\end{aligned}\tag{21}$$

where

$$\Delta_1^a=(1/2S)(2/N)\sum_1\gamma(1)(\langle a_1b_{-1}\rangle+\langle a_1^\dagger b_{-1}^\dagger\rangle)$$
$$+S^{-1}\eta_1(2/N)\sum_1\langle b_1^\dagger b_1\rangle$$
$$+(4/S)\tilde{L}(2/N)\sum_1\langle a_1^\dagger a_1\rangle, \quad (22)$$

$$\Delta_2^a(\mathbf{k})=(1/2S)\gamma(\mathbf{k})(2/N)\sum_1(\langle a_1^\dagger a_1\rangle+\langle b_1^\dagger b_1\rangle)$$
$$+S^{-1}\eta_1(2/N)\sum_1\gamma(\mathbf{k}-1)\langle a_1b_{-1}\rangle, \quad (23)$$

and where $\Delta_3^a(H)=\Delta_1^a(-H)$.

These equations follow directly from the Hamiltonian [Eq. (3)] using the Bose commutation relations (2) when the interaction term is linearized by replacing pairs of spin-deviation operators, in all possible combinations, by their thermal expectation values. Equating the secular determinant to zero, we then obtain the two spin-wave branches

$$\omega_{1,2}(\mathbf{k})$$
$$=\{\omega^2(\mathbf{k})-(SzJ)^2[\eta_3(\Delta_1^a+\Delta_3^a)-2\gamma(\mathbf{k})\Delta_2^a(\mathbf{k})]\}^{1/2}$$
$$\mp[\mu H-\tfrac{1}{2}SzJ(\Delta_1^a-\Delta_3^a)], \quad (24)$$

where

$$\omega(\mathbf{k})=SzJ[\eta_7^2-\gamma^2(\mathbf{k})]^{1/2}. \quad (25)$$

Neglecting the contribution of $\mathcal{H}_I$, we obtain the familiar result

$$\omega_{1,2}^0(\mathbf{k})=\omega(\mathbf{k})\mp\mu H. \quad (26)$$

This expression could, of course, also be obtained by diagonalizing the Hamiltonian $\mathcal{H}_0$ by means of a canonical transformation.[7]

The transition from the antiferromagnetic phase to the flop phase occurs at that field for which the frequency of the lower spin-wave branch $\omega_1(\mathbf{k})$ becomes zero. This first occurs for $k=0$, and the temperature-dependent critical field is given by

$$\mu H_{c1}^a=SzJ\{[(\eta_7^2-1)-\eta_3(\Delta_1^a+\Delta_3^a)-2\Delta_2^a(0)]^{1/2}$$
$$+\tfrac{1}{2}(\Delta_1^a-\Delta_3^a)\}. \quad (27)$$

When we neglect the contribution from $\mathcal{H}_I$, the critical fields are given by

$$\mu H_{c1,0}^a=2SzJ[\tfrac{1}{2}\tilde{K}(1+\tfrac{1}{4}\tilde{K})]^{1/2}, \quad (28)$$

and

$$\mu H_{c1,0}^a=2SzJ[\tilde{L}\xi^2(1+\tilde{L}\xi^2)]^{1/2}, \quad (29)$$

for the K- and L-type anisotropy, respectively.

Equation (29) agrees with the result obtained in Ref. 8. The correct leading order in the $1/2S$ expansion is obtained by setting $\xi=1$. It should be emphasized, however, that by setting $\xi^2=1-1/2S$ in Eq. (29) only part of the next-order correction is obtained. There will be additional contributions to this order in $1/2S$ even at zero temperature.

The effect of the interaction Hamiltonian $\mathcal{H}_I$ is contained in the functions Δ_i^a. They are given in terms of the expectation values of pairs of spin-deviation operators. Because we are performing a perturbation expansion in $1/2S$ these expectation values will be taken with respect to $\mathcal{H}_0$. Instead of diagonalizing $\mathcal{H}_0$ by means of a canonical transformation and then rewriting $\mathcal{H}$, in terms of the new spin-wave variables, we have chosen to determine the expectation values occurring in the Δ's directly by calculating the equations of motion for time-ordered products of pairs of spin-deviation operators. From these Green's functions we then obtain the required equal-time expectation values. The details of the calculation are contained in Appendix A. We find

$$\langle a_{\mathbf{k}}^\dagger a_{\mathbf{k}}\rangle=-[1/2\omega(\mathbf{k})]\{[\omega(\mathbf{k})-SzJ\eta_7][1+n_1(\mathbf{k})]$$
$$-[\omega(\mathbf{k})+SzJ\eta_7]n_2(\mathbf{k})\}, \quad (30)$$

$$\langle a_{\mathbf{k}}b_{-\mathbf{k}}\rangle=-[SzJ\gamma(\mathbf{k})/2\omega(\mathbf{k})][1+n_1(\mathbf{k})+n_2(\mathbf{k})]. \quad (31)$$

The corresponding expressions for $\langle b_{\mathbf{k}}^\dagger b_{\mathbf{k}}\rangle$ and $\langle a_{\mathbf{k}}^\dagger b_{-\mathbf{k}}^\dagger\rangle$ are obtained by interchanging n_1 and n_2 in $\langle a_{\mathbf{k}}^\dagger a_{\mathbf{k}}\rangle$ and $\langle a_{\mathbf{k}}b_{-\mathbf{k}}\rangle$, respectively (replacing H by $-H$). Here $n(\mathbf{k})$ is the Bose occupation number factor

$$n_{1,2}=[\exp(\beta\omega_{1,2}^0)-1]^{-1}.$$

From Eqs. (27), (30), and (31) the expression for the critical field can now be written

$$\mu H_{c1}^a=SzJ\Bigg\{(\eta_7^2-1)+S^{-1}(\eta_3\eta_5-1)$$
$$\times\left[1-(2/N)\sum_{\mathbf{k}}\frac{\eta_3}{(\eta_3^2-\gamma_{\mathbf{k}}^2)^{1/2}}(1+n_1(\mathbf{k})+n_2(\mathbf{k}))\right]$$
$$-2\tilde{L}^2S^{-1}(2/N)\sum_{\mathbf{k}}\frac{\gamma_{\mathbf{k}}^2}{(\eta_3^2-\gamma_{\mathbf{k}}^2)^{1/2}}[1+n_1(\mathbf{k})+n_2(\mathbf{k})]\Bigg\}^{1/2}$$
$$+\tfrac{1}{2}zJ\eta_6(2/N)\sum_{\mathbf{k}}[n_1(\mathbf{k})-n_2(\mathbf{k})]. \quad (32)$$

To the order in $1/2S$ to which we are working, the critical field can be written

$$\mu H_{c1}^a(T)=\mu H_{c1,0}^a\{1+(1/2S)\delta_0+(1/2S)\delta_T\}, \quad (33)$$

where δ_0 represents the contribution of the zero-point motion. Explicit calculations for $(kT/\mu H_{c1,0}^a)\ll 1$ give

$$\delta_0=-0.156+c_1, \quad (34)$$

$$\delta_T=(z^{3/2}/\pi^2)\tilde{K}\eta_1\eta_2(kT/\mu H_{c1,0}^a)^{5/2}\zeta(5/2)\Gamma(5/2), \quad (35)$$

for the K-type anisotropy, while

$$\delta_0=-(1+\tilde{L})^{-1}[(1+2\tilde{L})(0.156-c_1)-\tfrac{1}{2}(0.097-c_2)], \quad (36)$$

$$\delta_T=-(8z^{3/2}/\pi^2)\tilde{L}^2(1+\tilde{L})\Gamma(3/2)\zeta(3/2)(kT/\mu H_{c1,0}{}^a)^{3/2}$$
$$+4(z^{3/2}/\pi^2)\tilde{L}\Gamma(5/2)\zeta(5/2)(kT/\mu H_{c1,0}{}^a)^{5/2}, \quad (37)$$

for L-type anisotropy. Here ζ and Γ are, respectively, the Riemann-zeta function and the gamma function. The constants c_1 and c_2 are zero in the absence of anisotropy and are given by

$$c_1=(2/N)\sum_{\mathbf{k}}\{1/[1-\gamma^2(\mathbf{k})]^{1/2}-\eta_3{}^2/(\eta_3{}^2-\gamma_{\mathbf{k}}{}^2)^{1/2}\}, \quad (38)$$

$$c_2=(2/N)\sum_{\mathbf{k}}\{[\eta_3{}^2-\gamma^2(\mathbf{k})]^{1/2}-[1-\gamma^2(\mathbf{k})]^{1/2}\}. \quad (39)$$

The summations[4]

$$(2/N)\sum_{\mathbf{k}}[1-\gamma^2(\mathbf{k})]^{1/2}=1-0.097, \quad (40)$$

$$(2/N)\sum_{\mathbf{k}}\{1/[1-\gamma^2(\mathbf{k})]^{1/2}\}=1.156 \quad (41)$$

define the numerical factors occurring in Eqs. (34) and (36). We recall that for the L-type anisotropy, part of the $1/2S$ correction is contained in $\mu H_{c1,0}{}^a$ through its dependence on ξ.

B. Flop Phase

Similarly, from the Hamiltonian (8) we calculate the equations of motion for the spin-deviation operators in the flop phase. In linearized form we obtain

$$i(\partial/\partial t)a_{\mathbf{k}}=-\tfrac{1}{2}i(NS)^{1/2}u\{\mu H(1-\Delta_0{}^L)$$
$$-2zJS(1-u^2)^{1/2}[\eta_{10}-\eta_{11}\Delta_0{}^L+\eta_2\Delta_0{}^K]\}\delta_{\mathbf{k},0}$$
$$+SzJ\{(A+\Delta_1)a_{\mathbf{k}}+(e_{\mathbf{k}}+\Delta_2(\mathbf{k}))b_{\mathbf{k}}$$
$$+(B+\Delta_3)a_{-\mathbf{k}}{}^{+}+(d_{\mathbf{k}}+\Delta_4(k))b_{-\mathbf{k}}{}^{\dagger}\}, \quad (42)$$

where

$$\Delta_0{}^L=(4S)^{-1}(2/N)\sum_{1}(2\langle a_1{}^\dagger a_1\rangle-\langle a_1a_{-1}\rangle), \quad (43)$$

$$\Delta_0{}^K=S^{-1}(2/N)\sum_{1}[\gamma(1)(\langle a_1{}^\dagger b_1\rangle-\langle a_1b_{-1}\rangle)-\langle a_1{}^\dagger a_1\rangle], \quad (44)$$

$$\Delta_1=C_1(2/N)\sum_{1}\gamma(1)\langle a_1{}^\dagger b_1\rangle$$
$$+(C_3+C_5)(2/N)\sum_{1}\langle a_1{}^\dagger a_1\rangle$$
$$+C_2(2/N)\sum_{1}\gamma(1)\langle a_1b_{-1}\rangle+C_4(2/N)\sum_{1}\langle a_1a_{-1}\rangle, \quad (45)$$

$$\Delta_2(\mathbf{k})=C_1\gamma(\mathbf{k})(2/N)\sum_{1}\langle a_1{}^\dagger a_1\rangle$$
$$+\tfrac{1}{2}C_2\gamma(\mathbf{k})(2/N)\sum_{1}\langle a_1a_{-1}\rangle$$
$$+C_3(2/N)\sum_{1}\gamma(\mathbf{k}-1)\langle a_1{}^\dagger b_1\rangle, \quad (46)$$

$$\Delta_3=\tfrac{1}{2}C_1(2/N)\sum_{1}\gamma(1)\langle a_1b_{-1}\rangle$$
$$+\tfrac{1}{2}C_2(2/N)\sum_{1}\gamma(1)\langle a_1{}^\dagger b_1\rangle$$
$$+C_4(2/N)\sum_{1}\langle a_1{}^\dagger a_1\rangle+\tfrac{1}{2}C_5(2/N)\sum_{1}\langle a_1a_{-1}\rangle, \quad (47)$$

$$\Delta_4(\mathbf{k})=\tfrac{1}{2}C_1\gamma(\mathbf{k})(2/N)\sum_{1}\langle a_1a_{-1}\rangle$$
$$+C_2\gamma(\mathbf{k})(2/N)\sum_{1}\langle a_1{}^\dagger a_1\rangle$$
$$+C_3(2/N)\sum_{1}\gamma(\mathbf{k}-1)\langle a_1b_{-1}\rangle, \quad (48)$$

and where

$$C_1=-(1/S)(1-\eta_2u^2),$$
$$C_2=-(1/S)\eta_2u^2,$$
$$C_3=(1/S)(\eta_1-2\eta_2u^2),$$
$$C_4=-(1/S)\tfrac{3}{2}\tilde{L}u^2,$$
$$C_5=-(4/S)\tilde{L}(1-\tfrac{3}{2}u^2). \quad (49)$$

From the static part of Eq. (42) the relationship between the angle θ (or u) and the field H is determined,

$$\mu H=2SzJ(1-u^2)^{1/2}[\eta_{10}+\eta_2\Delta_0{}^K+4\tilde{L}\Delta_0{}^L]. \quad (50)$$

With $\Delta_0{}^K=\Delta_0{}^L=0$ this agrees with the result obtained by Wang and Callen for the L-type anisotropy they considered.

In the equation of motion for the other spin-deviation operators the vanishing of the static parts leads to the identical condition as given above, and we need consider only the dynamic parts of these equations. If we write the dynamic part of the equation for $a_{\mathbf{k}}$ in the abbreviated form

$$i(\partial/\partial t)a_{\mathbf{k}}$$
$$=\alpha_1(\mathbf{k})a_{\mathbf{k}}+\alpha_2(\mathbf{k})b_{\mathbf{k}}+\alpha_3(\mathbf{k})a_{-\mathbf{k}}{}^\dagger+\alpha_4(\mathbf{k})b_{-\mathbf{k}}{}^\dagger, \quad (51)$$

then the remaining equations required to form a complete set of coupled equations take the form

$$i(\partial/\partial t)b_{\mathbf{k}}$$
$$=\alpha_2(\mathbf{k})a_{\mathbf{k}}+\alpha_1(\mathbf{k})b_{\mathbf{k}}+\alpha_4(\mathbf{k})a_{-\mathbf{k}}{}^\dagger+\alpha_3(\mathbf{k})b_{-\mathbf{k}}{}^\dagger, \quad (52)$$

$$-i(\partial/\partial t)a_{-\mathbf{k}}{}^\dagger$$
$$=\alpha_3(\mathbf{k})a_{\mathbf{k}}+\alpha_4(\mathbf{k})b_{\mathbf{k}}+\alpha_1(\mathbf{k})a_{-\mathbf{k}}{}^\dagger+\alpha_2(\mathbf{k})b_{-\mathbf{k}}{}^\dagger, \quad (53)$$

$$-i(\partial/\partial t)b_{-\mathbf{k}}{}^\dagger$$
$$=\alpha_4(\mathbf{k})a_{\mathbf{k}}+\alpha_3(\mathbf{k})b_{\mathbf{k}}+\alpha_2(\mathbf{k})a_{-\mathbf{k}}{}^\dagger+\alpha_1(\mathbf{k})b_{-\mathbf{k}}{}^\dagger, \quad (54)$$

when we note that $\alpha_i(\mathbf{k})=\alpha_i{}^*(\mathbf{k})$ and $\alpha_i(\mathbf{k})=\alpha_i(-\mathbf{k})$.

The secular determinant can be written in the form

$$D=(\omega^2-\omega_1{}^2)(\omega^2-\omega_2{}^2), \quad (55)$$

where the two spin-wave branches ω_1 and ω_2 are given by

$$\omega_1=[(\alpha_1-\alpha_2+\alpha_3-\alpha_4)(\alpha_1-\alpha_2-\alpha_3+\alpha_4)]^{1/2}, \quad (56)$$

$$\omega_2=[\alpha_1+\alpha_2+\alpha_3+\alpha_4)(\alpha_1+\alpha_2-\alpha_3-\alpha_4)]^{1/2}, \quad (57)$$

with

$$\alpha_1=SzJ[A+\Delta_1],$$
$$\alpha_2(\mathbf{k})=SzJ[e_{\mathbf{k}}+\Delta_2(\mathbf{k})],$$
$$\alpha_3=SzJ[B+\Delta_3],$$
$$\alpha_4(\mathbf{k})=SzJ[d_{\mathbf{k}}+\Delta_4(\mathbf{k})]. \quad (58)$$

Substituting for A, B, $e_{\mathbf{k}}$, $d_{\mathbf{k}}$ from Eqs. (11) and making use of the relation between the angle and the magnetic field Eq. (50), these spin-wave spectra can be rewritten

$$\begin{aligned}\omega_{1,2}=SzJ[&1\mp\gamma(\mathbf{k})+\tilde{L}u^2\xi(1-\xi)\\&+\Delta_0+\Delta_1\mp\Delta_2+\Delta_3\mp\Delta_4]^{1/2}\\&\times[1\mp\gamma(\mathbf{k})(1-2\eta_2u^2)\\&-\tilde{L}u^2\xi(1+\xi)+\Delta_0+\Delta_1\pm\Delta_2-\Delta_3\pm\Delta_4]^{1/2},\end{aligned}\tag{59}$$

where

$$\Delta_0=2(1-u^2)[\eta_2\Delta_0{}^K+4\tilde{L}\Delta_0{}^L].\tag{60}$$

The flop-antiferromagnetic phase boundary is determined by setting $\omega_2(k=0)$ equal to zero. This gives the following condition on the angle θ:

$$u^2=(1+\Delta)/[\eta_2+\tfrac{1}{2}\tilde{L}\xi(1+\xi)],\tag{61}$$

where

$$\Delta=\tfrac{1}{2}[\Delta_0+\Delta_1+\Delta_2(0)-\Delta_3-\Delta_4(0)],\tag{62}$$

which, when substituted in the relation between the angle and the magnetic field Eq. (50), yields the expression for the critical field

$$\begin{aligned}H_{c1}{}^f=2SzJ[&\eta_{10}+\eta_2\Delta_0{}^K+4\tilde{L}\Delta_0{}^L]\\&\times\left\{\frac{2(\eta_2-1)+\tilde{L}\xi(1+\xi)-2\Delta}{2\eta_2+\tilde{L}\xi(1+\xi)}\right\}^{1/2},\end{aligned}\tag{63}$$

where $\Delta_0{}^K(u)$, $\Delta_0{}^L(u)$, and $\Delta(u)$ are to be evaluated for

$$u^2=[\eta_2+\tfrac{1}{2}\tilde{L}\xi(1+\xi)]^{-1}\quad\text{and}\quad\xi=1.$$

To leading order in $1/2S$ (apart from the correction contained in ξ), we obtain

$$\mu H_{c1,0}{}^f=2SzJ[\tfrac{1}{2}\tilde{K}(1+\tfrac{1}{2}\tilde{K})]^{1/2},\tag{64}$$

$$\mu H_{c1,0}{}^f=2SzJ(1-\tilde{L}\xi^2)\{\tilde{L}\xi(1+\xi)/[2+\tilde{L}\xi(1+\xi)]\}^{1/2},\tag{65}$$

respectively, for the K- and L-type anisotropy. Equation (65) agrees with the result obtained in Ref. 8.

When we compare these expressions with the corresponding values for $\mu H_{c1,0}{}^a$ given by Eqs. (28) and (29), we note that in the case of anisotropic exchange $\mu H_{c1,0}{}^a=\mu H_{c1,0}{}^f$. This would mean that the transition between the antiferromagnetic phase and the flop phase, although of first order, would have no superheating or supercooling associated with it. This is identical with the result obtained in the simple molecular-field calculation discussed in the Introduction.

For the L-type anisotropy, on the other hand, we find that $\mu H_{c1,0}{}^a>\mu H_{c1,0}{}^f$ which will give the kind of hysteresis effects we expect at a first-order phase transition. For a discussion of the typical size of this hysteresis and its experimental observation we refer to Ref. 10. The upper critical field $H_{c2}{}^f$ is most simply obtained by determining the field for which the angle θ becomes equal to zero. Setting $u=0$ in the relation between the angle and the magnetic field Eq. (50), we obtain

$$\mu H_{c2}{}^f=2SzJ[\eta_{10}+\eta_2\Delta_0{}^K+4\tilde{L}\Delta_0{}^L],\tag{66}$$

where $\Delta_0{}^K(u)$ and $\Delta_0{}^L(u)$ are now to be evaluated for $u=0$. Alternatively the upper critical field can be obtained by determining the field at which the spin-wave mode $\omega_1(k=0)$ becomes negative. From Eq. (59), we then obtain the same result as that just given.

The expectation values of the pairs of spin-deviation operators which occur in the Δ's are calculated in Appendix A. We obtain

$$\begin{aligned}\langle a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}\rangle=-\tfrac{1}{2}+\tfrac{1}{2}SzJ\{&[(A-e_{\mathbf{k}})/\omega_1{}^0(\mathbf{k})][n_1(\mathbf{k})+\tfrac{1}{2}]\\&+[(A+e_{\mathbf{k}})/\omega_2{}^0(\mathbf{k})][n_2(\mathbf{k})+\tfrac{1}{2}]\},\end{aligned}\tag{67}$$

$$\begin{aligned}\langle a_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}\rangle=-\tfrac{1}{2}SzJ\{&[(A-e_{\mathbf{k}})/\omega_1{}^0(\mathbf{k})][n_1(\mathbf{k})+\tfrac{1}{2}]\\&-[(A+e_{\mathbf{k}})/\omega_2{}^0(\mathbf{k})][n_2(\mathbf{k})+\tfrac{1}{2}]\},\end{aligned}\tag{68}$$

$$\begin{aligned}\langle a_{\mathbf{k}}a_{-\mathbf{k}}\rangle=-\tfrac{1}{2}SzJ\{&[(B-d_{\mathbf{k}})/\omega_1{}^0(\mathbf{k})][n_1(\mathbf{k})+\tfrac{1}{2}]\\&+[(B+d_{\mathbf{k}})/\omega_2{}^0(\mathbf{k})][n_2(\mathbf{k})+\tfrac{1}{2}]\},\end{aligned}\tag{69}$$

$$\begin{aligned}\langle a_{\mathbf{k}}b_{-\mathbf{k}}\rangle=\tfrac{1}{2}SzJ\{&[(B-d_{\mathbf{k}})/\omega_1{}^0(\mathbf{k})][n_1(\mathbf{k})+\tfrac{1}{2}]\\&-[(B+d_{\mathbf{k}})/\omega_2{}^0(\mathbf{k})][n_2(\mathbf{k})+\tfrac{1}{2}]\},\end{aligned}\tag{70}$$

where

$$\begin{aligned}\omega_{1,2}{}^0(\mathbf{k})=SzJ&[1\mp\gamma(\mathbf{k})+\tilde{L}u^2\xi(1-\xi)]^{1/2}\\&\times[1\mp\gamma(\mathbf{k})(1-2\eta_2u^2)-\tilde{L}u^2\xi(1+\xi)]^{1/2},\end{aligned}\tag{71}$$

$$A\pm e_{\mathbf{k}}=(1-\tilde{L}\xi^2u^2)\pm(1-\eta_2u^2)\gamma(\mathbf{k}),\tag{72}$$

$$B\pm d_{\mathbf{k}}=[\tilde{L}\xi\pm\eta_2\gamma(\mathbf{k})]u^2.\tag{73}$$

By means of these relations the expressions for the critical fields can be written in rather simple forms. Consider first the lower critical field given by Eq. (63) for K-type anisotropy,

$$\begin{aligned}\mu H_{c1}{}^f=\mu H_{c1,0}{}^f\{&1+(2S)^{-1}\\&\times[1-(2/N)\sum_{\mathbf{k}}2/[1-\gamma^2(\mathbf{k})]^{1/2}[n(\omega_1{}^0(\mathbf{k}))+\tfrac{1}{2}]]\},\end{aligned}\tag{74}$$

where

$$\omega_1{}^0(\mathbf{k})=SzJ[1-\gamma^2(\mathbf{k})]^{1/2}.\tag{75}$$

For L-type anisotropy a similar but somewhat more complicated expression obtains. The upper critical field including both types of anisotropies may be written

$$\begin{aligned}H_{c2}{}^f=2SzJ&\\&\times\{\eta_{10}-(2S)^{-1}(2/N)\sum_{\mathbf{k}}[(\eta_2(1+\gamma_{\mathbf{k}})-2\tilde{L})n_1(\mathbf{k})\\&+(\eta_2(1-\gamma_{\mathbf{k}})-2\tilde{L})n_2(\mathbf{k})]\}.\end{aligned}\tag{76}$$

Explicit evaluation for the lower critical field yields the following expressions:

$$H_{c1}{}^f=H_{c1,0}{}^f[1+\delta_0/2S+\delta_T/2S],\tag{77}$$

with

$$\delta_0=-0.156\tag{78}$$

and

$$\delta_T=-(3^{3/2}/2\pi^2)\Gamma(2)\zeta(2)(2kT/SzJ)^2\tag{79}$$

for the K anisotropy, while

$$\delta_0=\left(\frac{1-2\tilde{L}}{1-\tilde{L}}+\frac{3}{8}\frac{1}{1+\tilde{L}}\right)-\frac{1}{1-\tilde{L}^2}\left(\frac{1+\tilde{L}}{1-\tilde{L}}\right)^{1/2}\times\left[\frac{0.903}{2}(1+3\tilde{L})+1.156(1-4\tilde{L}-\tilde{L}^2)\right], \quad (80)$$

$$\delta_T=-\frac{3^{3/2}}{2\pi^2}\left(\frac{1+\tilde{L}}{1-\tilde{L}}\right)^{3/2}\left(1-\frac{4\tilde{L}}{1-\tilde{L}^2}\right)\Gamma(2)\zeta(2)\left(\frac{2kT}{SzJ}\right)^2 \quad (81)$$

for the L-type anisotropy. The numerical factors in δ_0 are defined by Eqs. (40) and (41).

Comparing Eqs. (34) and (78) we see that for the K anisotropy, the zero-temperature values of the critical fields H_{c1}^a and H_{c1}^f will differ due to the zero-point motion contributions. Because the constant c_1 in Eq. (34) is positive, H_{c1}^f will be lower than H_{c1}^a. At finite temperature this splitting is further increased as μH_{c1}^a increases as $(kT/\mu H_{c1}^a)^{5/2}$ while μH_{c1}^f decreases as $(kT/SzJ)^2$.

For the L-type anisotropy H_{c1}^a will, in the extremely low-temperature region $[(kT/\mu H_{c1}^a)\ll 1]$, decrease more rapidly than H_{c1}^f and thus decrease the splitting between the superheating and supercooling fields.

For the upper critical field we find, including both types of anisotropy,

$$\mu H_{c2}^f=\mu H_{c2,0}\times[1-(2S)^{-1}(3^{3/2}/\pi^2)\Gamma(3/2)\zeta(3/2)(2kT/SzJ)^{3/2}], \quad (82)$$

where

$$\mu H_{c2,0}=2SzJ\eta_{10}. \quad (83)$$

C. Paramagnetic Phase

In the paramagnetic phase we need consider only a single equation of motion. When linearized this has the form

$$i(\partial/\partial t)a_{\mathbf{k}}=\{\mu H-SzJ[\eta_8-\gamma(\mathbf{k})-\Delta^p(\mathbf{k})]\}a_{\mathbf{k}}, \quad (84)$$

where

$$\Delta^p(\mathbf{k})=S^{-1}N^{-1}\sum_{\mathbf{k}'}n(\mathbf{k}')\times[\eta_1(1+\gamma(\mathbf{k}'-\mathbf{k}))-\gamma(\mathbf{k}')-\gamma(\mathbf{k})-4\tilde{L}]. \quad (85)$$

As the magnetic field is lowered the spin-wave mode given by Eq. (84) first becomes zero at the point $K_0=(\pi/a)(1, 1, 1)$ in reciprocal space. Setting $\omega(K_0)=0$, we obtain

$$\mu H_{c2}^p=2SzJ\{\eta_{10}-(2S)^{-1}(2/N)\times\sum_{\mathbf{k}}[\eta_2(1-\gamma(\mathbf{k}))-2\tilde{L}]n(\mathbf{k})\} \quad (86)$$

for the critical field. Explicit evaluation gives the result given by Eq. (82). As expected for a phase transition of second order, the critical fields as calculated from the flop side and the paramagnetic side of the phase boundary are identical. The expression for the critical field Eq. (82) agrees with the results previously obtained by Falk[9] and by Anderson and Callen[10] (for zero anisotropy).

IV. FREE ENERGY

The free energy is defined by

$$F=-kT\ln Z, \quad (87)$$

where Z is the partition function

$$Z=\operatorname{Tr}\exp(-\beta\mathcal{H}). \quad (88)$$

To the order in $1/2S$ that we are working, the free energy can conveniently be expressed in terms of the renormalized spin-wave frequencies discussed in the previous section.

For the antiferromagnetic phase we obtain

$$F=E_0+SzJ\sum_{\mathbf{k}}\{[\eta_7^2-\gamma^2(\mathbf{k})]^{1/2}-\eta_3\}-\tfrac{1}{4}zJ(2/N)\sum_{\mathbf{k},\mathbf{k}'}\{A(\mathbf{k},\mathbf{k}')+\eta_5-2B(\mathbf{k})\}+\beta^{-1}\sum_{\mathbf{k}}\{\ln[1-e^{-\beta\omega_1(\mathbf{k})}]+\ln[1-e^{-\beta\omega_2(\mathbf{k})}]\}, \quad (89)$$

where

$$A(\mathbf{k},\mathbf{k}')=\frac{\eta_3^2\eta_5-\eta_3[\gamma^2(\mathbf{k})+\gamma^2(\mathbf{k}')]+\eta_1\gamma^2(\mathbf{k})\gamma^2(\mathbf{k}')}{[\eta_3^2-\gamma^2(\mathbf{k})]^{1/2}[\eta_3^2-\gamma^2(\mathbf{k}')]^{1/2}} \quad (90)$$

and

$$B(\mathbf{k})=[\eta_3\eta_5-\gamma^2(\mathbf{k})]/[\eta_3^2-\gamma^2(\mathbf{k})]^{1/2}. \quad (91)$$

E_0 is given by Eq. (4) and $\omega_{1,2}$ by Eq. (24).

The free energy as given by Eq. (89) agrees with the result previously obtained by Oguchi[7] in the limit of zero anisotropy, $\eta_i=1$, when the last term is expanded about the temperature-independent part of the renormalized spin-wave frequencies. Both expressions are correct to the same order in $1/2S$.

In the flop phase we obtain similarly,

$$F=E_0-\tfrac{1}{2}NSzJA+\tfrac{1}{2}\sum_{\mathbf{k}}[\omega_1^0(\mathbf{k})+\omega_2^0(\mathbf{k})]+G+\beta^{-1}\sum_{\mathbf{k}}\{\ln[1-e^{-\beta\omega_1(\mathbf{k})}]+\ln[1-e^{-\beta\omega_2(\mathbf{k})}]\}. \quad (92)$$

E_0 and A are defined in Eqs. (9) and (11), respectively, and $\omega_{1,2}$ by Eq. (59). The G term has its origin in $\mathcal{H}_{\mathrm{I}}$ and is of one order higher in $1/2S$ as compared to the leading contribution from $\mathcal{H}_0$ in the two preceding terms. Because the expression for G is rather lengthy, we have chosen to give the explicit expression in Appendix B.

In the flop phase there is an additional term $\mathcal{H}_{\mathrm{II}}$ in the Hamiltonian as compared to the other phases. From this term the angle of the magnetization with the anisotropy axis could be determined. When the angle satisfies the equilibrium condition, Eq. (50), the

$\mathcal{H}_{\mathrm{II}}$ term will not contribute directly to the partition function to the order in $1/2S$ to which we are working. This term will, however, contribute to F through the dependence of u on the magnetic field and the temperature. From Eq. (50) we have

$$\mu H = 2SzJ(1-u^2)^{1/2}[\eta_{10}+\eta_2\Delta_0{}^K+4\tilde{L}\Delta_0{}^L],$$

where the terms involving $\Delta_0{}^K$, $\Delta_0{}^L$ are of one order higher in $1/2S$ compared to the leading term in η_{10}. For the terms in F due to $\mathcal{H}_{\mathrm{I}}$ we can eliminate u in terms of the magnetic field simply using

$$\mu H = 2SzJ\eta_{10}(1-u^2)^{1/2}; \qquad \xi = 1.$$

However, in the remaining terms we need to include the correction terms $\Delta_0{}^K$ and $\Delta_0{}^L$. For instance, the terms

$$E_0(u) - \tfrac{1}{2}NSzJA(u) + \tfrac{1}{2}\sum_{\mathbf{k}}[\omega_1{}^0(\mathbf{k}) + \omega_2{}^0(\mathbf{k})] \tag{93}$$

are functions of both temperature and magnetic field and will contribute both to the specific heat and the susceptibility.

For the paramagnetic phase we obtain

$$F = E_0 + \beta^{-1}\sum_{\mathbf{k}} \ln[1-e^{-\beta\omega(\mathbf{k})}], \tag{94}$$

where from Eq. (84),

$$\omega(\mathbf{k}) = \mu H - SzJ[\eta_8 - \gamma(\mathbf{k}) - \Delta^p(\mathbf{k})], \tag{95}$$

and where E_0 is given by Eq. (15).

V. SPECIFIC HEAT

From the free energy the specific heat at constant H is obtained in the usual way

$$C_H = -T(\partial^2 F/\partial T^2)\,|_H.$$

We consider first the leading term in the $1/2S$ expansion. The results are given in Table I.

For a fixed temperature as a function of the magnetic field, the specific heat peaks at each of the three critical fields. For $\beta\mu\,|H-H_c|\ll 1$ it falls off linearly as a function of this parameter. In the opposite limit $\beta\mu\,|H-H_c|\gg 1$ the specific heat falls off exponentially on the paramagnetic side of H_{c2} and on the antiferromagnetic side of $H_{c1}{}^a$. The exponential behavior is a consequence of the gaps that develop in the low-lying spin-wave modes for $H\neq H_c$ in these phases. In the flop phase one of the modes remains fixed at zero frequency for $k=0$. This results in a more complicated behavior of the specific heat in the limit $\beta\mu\,|H-H_c|\gg 1$ as shown in the table. At the critical fields H_{c2} and $H_{c1}{}^a$ the specific heat is proportional to $(kT/SzJ)^{3/2}$ as a consequence of quadratic dispersion laws for these values of the magnetic field. In the flop phase both spin-wave modes become linear at $H=H_{c1}{}^f$ giving a specific heat proportional to $(kT/SzJ)^3$. However, the coefficient at $H_{c1}{}^a$ is smaller than that at $H_{c1}{}^f$ by the factor $(H_{c1}/H_{c2})^{3/2}$ and which of the two peaks is the larger will depend on the particular values of the anisotropy constants and the temperature being considered. In either case the peak at H_{c2} will in general be considerably larger than the peaks at H_{c1}.

In the absence of spin-wave interactions, the specific heat at the upper phase boundary is continuous. However, when the spin-wave interactions are included, a finite discontinuity is obtained as one would expect at a second-order phase transition. The reason for this is simple. The expression for the free energy in the flop phase goes continuously into the expression for the free energy in the paramagnetic phase when the angle θ goes to zero. However, when spin-wave interactions are included, this angle is temperature-dependent and will give an additional contribution to the specific heat. More specifically, the discontinuity is obtained because

$$\lim_{u\to 0}[\partial u/\partial T]\neq 0.$$

The specific heat for constant u on the other hand, that is along the phase boundary, is continuous.

From the paramagnetic side of the phase boundary the contribution of the spin-wave interactions to the specific heat at $H=H_{c2}$ is given by

$$C_{(1)}{}^p = -(2S)^{-1}Nk3\eta_{14} \times[(3^{3/2}/\pi^2)\Gamma(3/2)\zeta(3/2)]^2(2kT/SzJ)^2. \tag{96}$$

From the flop side, we obtain

$$C_{(1)}{}^f = C_{(1)}{}^p + \Delta C,$$

where the discontinuity ΔC is given by

$$\Delta C = (2S)^{-1}Nk\tfrac{9}{2}\eta_{15} \times[(3^{3/2}/\pi^2)\Gamma(3/2)\zeta(3/2)]^2(2kT/SzJ)^2. \tag{97}$$

The specific heat is seen to be largest on the flop side of the boundary.

We note that the effect of the spin-wave interactions in the paramagnetic phase of the antiferromagnet is more important than in the ferromagnet where the contribution of the spin-wave interactions to the specific heat is proportional to $(2kT/SzJ)^4$.[2,7,11] This is a consequence of the fact that in the antiferromagnet the low-lying excitations occur at the edge of the Brillouin zone whereas for the ferromagnet these occur for $k\approx 0$.

It is reasonable to assume that the discontinuity in the specific heat will persist along the whole phase boundary, indicating a true second-order phase transition, except possibly at the critical point $H=0$, $T=T_N$, where experimental and theoretical evidence suggest singular behavior when this point is approached along the temperature axis from both sides of T_N.

VI. SUSCEPTIBILITY

From the free energy, the magnetization and the static longitudinal susceptibility are obtained by means

---

[11] M. Wortis, Phys. Rev. **138**, A1126 (1956).

TABLE I. Specific heat to leading order in the $1/2S$ expansion.

Paramagnetic phase	$\mu(H-H_{c2,0})\gg kT$	$C=Nk(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}\exp[-\mu(H-H_{c2,0})/kT]\Gamma(3/2)[\mu(H-H_{c2,0})/kT]^2\{1+3[\mu(H-H_{c2,0})/kT]^{-1}+\cdots\}$
	$\mu(H-H_{c2,0})\ll kT$	$C=Nk(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}\{\Gamma(7/2)\zeta(5/2)-\frac{1}{2}\Gamma(5/2)\zeta(3/2)[\mu(H-H_{c2,0})/kT]+\cdots\}$
Flop phase	$\mu(H_{c2,0}-H)\ll kT$	$C=Nk(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}\{\Gamma(7/2)\zeta(5/2)-\frac{1}{2}\Gamma(5/2)\zeta(3/2)[\mu(H_{c2,0}-H)/kT]+\cdots\}$
	$\mu(H_{c2,0}-H)\gg kT$ and $\mu(H-H_{c1,0}{}^f)\gg kT$	$C=Nk(3^{3/2}/2\pi^2)(2kT/SzJ)^{3/2}\Gamma(5)\zeta(4)\{[kTH_{c2,0}/\mu(H_{c2,0}{}^2-H^2)]+\cdots$
	$\mu(H-H_{c1,0}{}^f)\ll kT$	$C=Nk(3^{3/2}/8\pi^2)(2kT/SzJ)^3(\eta_{13}/\eta_{14})^{3/2}\{\Gamma(5)\zeta(4)-\frac{1}{4}(\eta_{13}/\eta_{14}{}^2)\Gamma(2)\zeta(2)[\mu^2(H^2-(H_{c1,0}{}^f)^2)/(kT)^2]+\cdots\}$
Antiferromagnetic phase	$\mu(H_{c1,0}{}^a-H)\ll kT\ll 2\mu H_{c1,0}{}^a$	$C=Nk(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}(2H_{c1,0}{}^a/H_{c2,0})^{3/2}\eta_{14}{}^{3/2}\{\Gamma(7/2)\zeta(5/2)-\frac{1}{2}\Gamma(5/2)\zeta(3/2)[\mu(H_{c1,0}{}^a-H)/kT]+\cdots\}$
	$\mu(H_{c1,0}{}^a-H)\gg kT$	$C=Nk(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}(2H_{c1,0}{}^a/H_{c2,0})^{3/2}\eta_{14}{}^{3/2}\Gamma(3/2)\{\exp[-\mu(H_{c1,0}{}^a-H)/kT][\mu(H_{c1,0}{}^a-H)/kT]^2$ $\times\{1+3[\mu(H_{c1,0}{}^a-H)/kT]^{-1}+\cdots\}+\exp[-\mu(H_{c1,0}{}^a+H)/kT][\mu(H_{c1,0}{}^a+H)/kT]^2\{1+3[\mu(H_{c1,0}{}^a+H)/kT]^{-1}$ $+\cdots\}+\cdots\}$

of the relations

$$M=-\partial F/\partial H\,|_T, \qquad \chi=-\partial^2F/\partial H^2\,|_T.$$

We consider first only the leading terms in the $1/2S$ expansion. For the paramagnetic phase we obtain the familiar result for noninteracting spin waves,

$$M_0{}^p=\mu N[S-\sum_{\mathbf{k}}n_{\mathbf{k}}]. \tag{98}$$

For the flop phase we obtain

$$M_0{}^f=2(1-u^2)^{1/2}M_s, \tag{99}$$

where M_s is the sublattice magnetization,

$$M_s=\mu\tfrac{1}{2}N\left\{S+\tfrac{1}{2}-SzJ\left(\frac{2}{N}\right)\sum_{\mathbf{k}}\left[\frac{(A-e_{\mathbf{k}})}{\omega_1{}^0(\mathbf{k})}(n_1(\mathbf{k})+\tfrac{1}{2})\right.\right.$$
$$\left.\left.+\frac{(A+e_{\mathbf{k}})}{\omega_2{}^0(\mathbf{k})}(n_2(\mathbf{k})+\tfrac{1}{2})\right]\right\}, \tag{100}$$

with $\omega_{1,2}{}^0$ and $(A\pm e_{\mathbf{k}})$ as defined by Eqs. (71) and (72). This agrees with the result obtained in Ref. 8 for the case of L-type anisotropy. Already to this order in the $1/2S$ expansion it is necessary to take into account the corrections $\Delta_0{}^L$ and $\Delta_0{}^K$ in the relationship between the angle and the magnetic field Eq. (50) to obtain the correct expression for the magnetization from the free energy Eq. (92). For the antiferromagnetic phase we obtain from Eq. (89) the simple expression

$$M_0{}^a=\mu\sum_{\mathbf{k}}[n_1(\mathbf{k})-n_2(\mathbf{k})], \tag{101}$$

while the sublattice magnetizations are given by much more complicated expressions.

The explicit evaluations for M_0 and χ_0 are given in Tables II and III, respectively.

We note that χ_0 has the form

$$\chi_0=A(T)/[\beta\mu\,|\,H-H_c\,|]^{1/2} \tag{102}$$

in the neighborhood of each of the three critical fields, giving a square-root singularity at the phase boundaries. However, when we proceed to the next order in the $1/2S$ expansion, we find that the leading contribution to the susceptibility χ_1 is of the form

$$\chi_1=B(T)/[\beta\mu\,|\,H-H_c\,|]^{3/2}. \tag{103}$$

This suggests that the spin-wave result χ_0 is only the first term in a divergent series. Corresponding to the $[\beta\mu\,|\,H-H_c\,|]^{-3/2}$ divergent term in χ_1 the contribution of F_1 to the magnetization includes a term with a square-root divergence. Thus, while straightforward perturbation theory is sufficient to study the effect of spin-wave interactions on the specific heat in the neighborhood of the phase boundaries, for the magnetization and the susceptibility more sophisticated techniques will be necessary. This will presumably involve a direct resummation of the divergent series or the construction of suitable integral equations.

To pursue this point a little further, we write down

TABLE II. Magnetization to leading order in the $1/2S$ expansion.

Paramagnetic phase	$\mu(H-H_{c2,0})\gg kT$	$M=N\mu\{S-(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}\Gamma(3/2)\ \exp[-\mu(H-H_{c2,0})/kT]+\cdots\}$
	$\mu(H-H_{c2,0})\ll kT$	$M=N\mu\{S-(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}\Gamma(3/2)\ \mathbf{[}\zeta(3/2)-2\Gamma(1/2)[\mu(H-H_{c2,0})/kT]^{1/2}$ $-\zeta(1/2)[\mu(H-H_{c2,0})/kT]+\cdots\mathbf{]}\cdots\}$
Flop phase[a]	$\mu(H_{c2,0}-H)\ll kT$	$M=N\mu(H/H_{c2,0})\{S-s_0(H)+(3^{3/2}/4\pi^2)(2kT/SzJ)^{3/2}\mathbf{[}\Gamma(3/2)\zeta(3/2)-\pi[2\mu(H_{c2,0}-H)/kT]^{1/2}+\cdots\mathbf{]}\cdots\}$
	$\mu(H_{c2,0}-H)\gg kT$ and $\mu(H-H_{c1,0}^{f})\gg kT$	$M=N\mu(H/H_{c2,0})\{S-s_0(H)+(3^{3/2}/2\pi^2)\Gamma(4)\zeta(4)\ (2kT/SzJ)^{3/2}[kTH_{c2,0}/\mu(H_{c2,0}^2-H^2)]^{5/2}+\cdots\}$
	$\mu(H-H_{c1,0}^{f})\ll kT$	$M=N\mu(H/H_{c2,0})\{S-s_0(H)-(3^{3/2}/4\pi^2)\ (\eta_{13}/\eta_{14})^{1/2}\mathbf{[}\Gamma(2)\zeta(2)\ (2kT/SzJ)^2+\pi(2(\tilde{K}+2\tilde{L})/\eta_{14})^{1/2}(2kT/SzJ)^{3/2}$ $\times[\mu(H-H_{c1,0}^{f})kT]^{1/2}+\cdots\mathbf{]}\}$
Antiferromagnetic phase	$\mu(H_{c1,0}^{a}-H)\ll kT\ll 2\mu H_{c1,0}^{a}$	$M=N\mu(3^{3/2}/4\pi^2)\ (2kT/SzJ)^{3/2}(2H_{c1,0}^{a}/H_{c2,0})^{3/2}\eta_{14}^{3/2}\Gamma(3/2)\ \{\zeta(3/2)-2\Gamma(1/2)[\mu(H_{c1,0}^{a}-H)/kT]^{1/2}$ $-\zeta(1/2)[\mu(H_{c1,0}^{a}-H)/kT]+\cdots\}$
	$\mu(H_{c1,0}^{a}-H)\gg kT$	$M=N\mu(3^{3/2}/4\pi^2)\ (2kT/SzJ)^{3/2}(2H_{c1,0}^{a}/Hc_{2,0})^{3/2}\eta_{14}^{3/2}\Gamma(3/2)\ \exp(-\beta\mu H_{c1,0}^{a})2\sinh\beta\mu H+\cdots$

$$s_0(H)=\frac{1}{4\eta_{14}}\frac{2}{N}\sum_k\left(\frac{[1-\gamma(k)][\tilde{L}-\eta_2\gamma(k)]}{(1-\gamma(k))^{1/2}(1-(1-2\eta_2u^2)\gamma(k)-2\tilde{L}u^2)^{1/2}}+\frac{[1+\gamma(k)][\tilde{L}+\eta_2\gamma(k)]}{(1+\gamma(k))^{1/2}(1+(1-2\eta_2u^2)\gamma(k)-\tilde{L}u^2)^{1/2}}\right)-\frac{L}{2\eta_{14}}$$

$$s_0(H_{c2,0})=0;\qquad s_0(H_{c1,0}^{f})=0.127,\qquad L=0,\qquad K\neq0;\qquad s_0(H_{c1,0}^{f})=\tfrac{1}{2}(1-\tilde{L})^{-1}(1+\tilde{L}/1-\tilde{L})^{1/2}[(1+\tilde{L})0.156+0.097],\qquad K=0,\qquad L\neq0$$

[a] The magnetization at the critical fields to leading order in $1/2S$ is obtained by evaluating these expressions for $H=H_c(T)$.

TABLE III. Susceptibility near the phase boundaries to leading order in the $1/2S$ expansion.

Paramagnetic phase	$\mu(H-H_{c2,0})\ll kT$	$\chi=N(\mu^2/kT)\ (3^{3/2}/4\pi^2)\ (2kT/SzJ)^{3/2}\Gamma(3/2)\Gamma(1/2)[\mu(H-H_{c2,0})/kT]^{-1/2}+\cdots$
Flop phase	$\mu(H_{c2,0}-H)\ll kT$	$\chi=N(\mu^2/kT)\ (3^{3/2}/4\pi^2)\ (2KT/SzJ)^{3/2}\pi[2\mu(H_{c2,0}-H)/kT]^{-1/2}+\cdots$
	$\mu(H-H_{c1,0}^{f})\ll kT$	$\chi=N(\mu^2/kT)\ (3^{3/2}/4\pi^2)\ (2kT/SzJ)^{3/2}\pi(\eta_{13}(\tilde{K}+2\tilde{L})/2\eta_{14}^2)^{1/2}[\mu(H-H_{c1,0}^{f})/kT]^{-1/2}+\cdots$
Antiferromagnetic phase	$\mu(H_{c1,0}^{a}-H)\ll kT$	$\chi=N(\mu^2/kT)\ (3^{3/2}/4\pi^2)\ (2kT/SzJ)^{3/2}(2\eta_{14}H_{c1,0}^{a}/H_{c2,0})^{3/2}\Gamma(3/2)\Gamma(1/2)[\mu(H_{c1,0}^{a}-H)/kT]^{-1/2}+\cdots$

explicitly the expression for χ obtained in the paramagnetic phase for $\mu(H-H_{c2,0})\ll kT\ll 2SzJ$,

$$\chi_0=N\frac{\mu^2}{kT}\frac{3^{3/2}}{4\pi^2}\left(\frac{2kT}{SzJ}\right)^{3/2}\left\{\frac{\Gamma(1/2)}{[\mu(H-H_{c2,0})/kT]^{1/2}}+\zeta(1/2)+O[\mu(H-H_{c2,0})/kT]\right\},$$

$$\chi_1=-N\frac{\mu^2}{kT}\frac{1}{2S}\left(\frac{3}{2\pi}\right)^3\left(\frac{2kT}{SzJ}\right)^2$$

$$\times\left\{\frac{\zeta(3/2)\Gamma(1/2)}{[\mu(H-H_{c2,0})/kT]^{3/2}}+\frac{6\Gamma(1/2)\zeta(1/2)}{[\mu(H-H_{c2,0})/kT]^{1/2}}+4(\zeta(1/2)+\zeta(-1/2))+O[\mu(H-H_{c2,0})/kT]\right\}. \quad (104)$$

Because the anisotropy plays no essential role at the upper phase boundary, both L and K have been set equal to zero.

In a ferromagnet the susceptibility has a square-root singularity[11] $\chi\propto H^{-1/2}$ for $T<T_c$. In this temperature region $H=0$ represents the phase boundary between two oppositely aligned magnetic phases, and the square-root singularity is a consequence of the critical fluctuations near the phase boundary. In view of the formal similarity between the ferromagnet and the paramagnetic phase of the antiferromagnet, it might be expected that the susceptibility for $H\gtrsim H_{c2}$ would be of the form

$$\chi\propto[\mu(H-H_{c2}(T))/kT]^{-1/2},$$

where $H_{c2,0}$ in χ_0 has been replaced by the temperature-dependent critical field. The first few terms of the perturbation expansion do not, however, appear compatible with this simple form.

In the random-phase approximation (or self-consistent-field approximation), the susceptibility, which in a low-temperature expansion can be represented by a similarly divergent series, goes to a constant value as $H\to H_{c2}(T)$. That is, in this case the (divergent) series can easily be summed. It is by no means certain, however, that the series obtained using spin-wave theory will sum to a similar finite result.

ACKNOWLEDGMENT

We would like to thank Dr. H. Thomas for helpful discussions and for reading the manuscript.

APPENDIX A

The zero-order expectation values of pairs of spin-deviation operators will be calculated by means of Green's functions for noninteracting spin waves.

I. Antiferromagnetic Phase

We define the Green's functions

$$G_{11}(\mathbf{k},t-t')=-i\langle(a_{\mathrm{k}}(t)a_{\mathrm{k}}^{\dagger}(t'))_+\rangle, \quad (A1)$$

$$G_{12}(\mathbf{k},t-t')=-i\langle(b_{-\mathrm{k}}^{\dagger}(t)a_{\mathrm{k}}^{\dagger}(t'))_+\rangle, \quad (A2)$$

$$G_{21}(\mathbf{k},t-t')=-i\langle(a_{\mathrm{k}}^{\dagger}(t)b_{\mathrm{k}}^{\dagger}(t'))_+\rangle, \quad (A3)$$

$$G_{22}(\mathbf{k},t-t')=-i\langle(b_{-\mathrm{k}}(t)b_{-\mathrm{k}}^{\dagger}(t'))_+\rangle. \quad (A4)$$

Here the + denotes the Wick time-ordering operation. We shall calculate the Green's functions to lowest order in $1/2S$ only. The thermal expectation value is therefore to be taken with respect to $\mathcal{H}_0$, and the time development of the operators in the Green's functions is governed by $\mathcal{H}_0$ only. That is, the operators are to be taken in the interaction representation.

If we let

$$G_{AB}(t-t')=-i\langle(A(t)B(t'))_+\rangle, \quad (A5)$$

the equation of motion for the Green's function has, quite generally, the form

$$i(\partial/\partial t)G_{AB}(t-t')=\delta(t-t')\langle[A,B]\rangle$$
$$-i\langle([i\partial A(t)/\partial t]B(t'))_+\rangle, \quad (A6)$$

where

$$i\partial A/\partial t=[A,\mathcal{H}]. \quad (A7)$$

From $\mathcal{H}_0$ as given by Eq. (5) we obtain trivially the equations of motion for the Green's functions,

$$i(\partial/\partial t)G_{11}(\mathbf{k},t-t')=\delta(t-t')+(SzJ\eta_7+\mu H)$$
$$\times G_{11}(\mathbf{k},t-t')+SzJ\gamma(\mathbf{k})G_{12}(\mathbf{k},t-t'),$$
$$i(\partial/\partial t)G_{12}(\mathbf{k},t-t')=-SzJ\gamma(\mathbf{k})G_{11}(\mathbf{k},t-t')$$
$$-(SzJ\eta_7-\mu H)G_{12}(\mathbf{k},t-t'),$$
$$i(\partial/\partial t)G_{22}(\mathbf{k},t-t')=\delta(t-t')+(SzJ\eta_7-\mu H)$$
$$\times G_{22}(\mathbf{k},t-t')+SzJ\gamma(\mathbf{k})G_{21}(\mathbf{k},t-t'),$$
$$i(\partial/\partial t)G_{21}(\mathbf{k},t-t')=-SzJ\gamma(\mathbf{k})G_{22}(\mathbf{k},t-t')$$
$$-(SzJ\eta_7+\mu H)G_{21}(\mathbf{k},t-t'). \quad (A8)$$

Introducing the Fourier series[12]

$$G(\mathbf{k},t-t')=(-i\beta)^{-1}\sum_{\nu}e^{-i\omega_\nu(t-t')}G(\mathbf{k},\omega_\nu), \quad (A9)$$

where $\omega_\nu=\pi\nu/(-i\beta)$ and where ν is an even integer, then (A8) gives

$$G_{11}(\mathbf{k},\omega_\nu)=\frac{(\omega_\nu-\mu H+SzJ\eta_7)}{[\omega_\nu-\omega_2{}^0(\mathbf{k})][\omega_\nu+\omega_1{}^0(\mathbf{k})]},$$

$$G_{12}(\mathbf{k},\omega_\nu)=-\frac{SzJ\gamma(\mathbf{k})}{[\omega_\nu-\omega_2{}^0(\mathbf{k})][\omega_\nu+\omega_1{}^0(\mathbf{k})]},$$

$$G_{21}(\mathbf{k},\omega_\nu)=-\frac{SzJ\gamma(\mathbf{k})}{[\omega_\nu+\omega_2{}^0(\mathbf{k})][\omega_\nu-\omega_1{}^0(\mathbf{k})]},$$

$$G_{22}(\mathbf{k},\omega_\nu)=\frac{\omega_\nu+\mu H+SzJ\eta_7}{[\omega_\nu+\omega_2{}^0(\mathbf{k})][\omega_\nu-\omega_1{}^0(\mathbf{k})]}. \quad (A10)$$

---

[12] See, for example, L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin Inc., New York, 1962), Chap. 1.

Here

$$\omega_{1,2}{}^0(\mathbf{k})=\omega(\mathbf{k})\mp\mu H \tag{A11}$$

and

$$\omega(\mathbf{k})=SzJ[\eta_7{}^2-\gamma^2(\mathbf{k})]^{1/2}. \tag{A12}$$

From the definitions of the Green's function (A1) to (A4) it follows that

$$\begin{aligned}\langle a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}\rangle&=iG_{11}{}^<(\mathbf{k},t=t'),\\ \langle a_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger\rangle&=iG_{12}{}^{\gtrless}(\mathbf{k},t=t')=iG_{21}{}^{\gtrless}(\mathbf{k},t=t'),\\ \langle b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}\rangle&=iG_{22}{}^<\ \ (\mathbf{k},t=t'),\end{aligned} \tag{A13}$$

where

$$G^{\gtrless}(\mathbf{k},t=t')=\lim_{\epsilon\to0}[G(\mathbf{k},t\pm\epsilon,t)]. \tag{A14}$$

By inverting the Fourier series and evaluating the ν summations we obtain

$$\begin{aligned}iG_{11}{}^<(\mathbf{k},t=t')&=-(1/2\omega_{\mathbf{k}})[(\omega(\mathbf{k})-SzJ\eta_7)(1+n_1(\mathbf{k}))\\&\qquad-(\omega(\mathbf{k})+SzJ\eta_7)n_2(\mathbf{k})],\\ iG_{12}(\mathbf{k},t=t')&=iG_{21}(\mathbf{k},t=t')\\&=-[SzJ\gamma(\mathbf{k})/2\omega(\mathbf{k})][1+n_1(\mathbf{k})+n_2(\mathbf{k})],\\ iG_{22}{}^<(\mathbf{k},t=t')&=[1/2\omega(\mathbf{k})][(\omega(\mathbf{k})+SzJ\eta_7)n_1(\mathbf{k})\\&\qquad-(\omega(\mathbf{k})-SzJ\eta_7)(1+n_2(\mathbf{k}))],\end{aligned} \tag{A15}$$

where we have made use of the summation formulas

$$\begin{aligned}\lim_{\epsilon\to0}\beta^{-1}\sum_\nu(e^{+i\omega_\nu\epsilon}/\omega_\nu-\omega)&=n(\omega),\\ \lim_{\epsilon\to0}\beta^{-1}\sum_\nu(e^{-i\omega_\nu\epsilon}/\omega_\nu-\omega)&=n(\omega)+1,\\ \beta^{-1}\sum_\nu(\omega_\nu-\omega)^{-1}&=n(\omega)+\tfrac{1}{2}.\end{aligned} \tag{A16}$$

We note that all the expectation values are even in **k**, and that because the a and b operators commute, the order in which they appear is irrelevant. For instance, we have

$$\langle a_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger\rangle=\langle b_{\mathbf{k}}{}^\dagger a_{-\mathbf{k}}{}^\dagger\rangle. \tag{A17}$$

II. Flop Phase

We define the Green's functions

$$\begin{aligned}G_1{}^+(\mathbf{k},t-t')&=-i\langle(a_{\mathbf{k}}(t)a_{\mathbf{k}}{}^\dagger(t'))_+\rangle,\\ G_1{}^-(\mathbf{k},t-t')&=-i\langle(a_{-\mathbf{k}}{}^\dagger(t)a_{\mathbf{k}}{}^\dagger(t'))_+\rangle,\\ G_2{}^+(\mathbf{k},t-t')&=-i\langle(b_{\mathbf{k}}(t)a_{\mathbf{k}}{}^\dagger(t'))_+\rangle,\\ G_2{}^-(\mathbf{k},t-t')&=-i\langle(b_{-\mathbf{k}}{}^\dagger(t)a_{\mathbf{k}}{}^\dagger(t'))_+\rangle.\end{aligned} \tag{A18}$$

From the Hamiltonian (10) we obtain the equations of motion for the Green's functions. These are conveniently written in the matrix form

$$\begin{pmatrix}\omega_\nu/SzJ-A & -e_{\mathbf{k}} & -B & -d_{\mathbf{k}}\\ -e_{\mathbf{k}} & \omega_\nu/SzJ-A & -d_{\mathbf{k}} & -B\\ B & d_{\mathbf{k}} & \omega_\nu/SzJ+A & e_{\mathbf{k}}\\ d_{\mathbf{k}} & B & e_{\mathbf{k}} & \omega_\nu/SzJ+A\end{pmatrix}\begin{pmatrix}G_1{}^+(\mathbf{k},\omega_\nu)\\ G_2{}^+(\mathbf{k},\omega_\nu)\\ G_1{}^-(\mathbf{k},\omega_\nu)\\ G_2{}^-(\mathbf{k},\omega_\nu)\end{pmatrix}=\begin{pmatrix}1/SzJ\\0\\0\\0\end{pmatrix} \tag{A19}$$

with A, B, $d_{\mathbf{k}}$, and $e_{\mathbf{k}}$ as defined in the text.

Solving this set of equations for the Green's functions and performing the ν summations we obtain the required expectation values given by Eqs. (67) to (70) in the text.

Furthermore we note that

$$\begin{aligned}\langle a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}\rangle&=\langle b_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}\rangle,\\ \langle a_{\mathbf{k}}{}^\dagger a_{-\mathbf{k}}{}^\dagger\rangle&=\langle b_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger\rangle=\langle a_{-\mathbf{k}}a_{\mathbf{k}}\rangle=\langle b_{-\mathbf{k}}b_{\mathbf{k}}\rangle,\\ \langle a_{\mathbf{k}}{}^\dagger b_{\mathbf{k}}\rangle&=\langle b_{\mathbf{k}}a_{\mathbf{k}}{}^\dagger\rangle=\langle b_{\mathbf{k}}{}^\dagger a_{\mathbf{k}}\rangle=\langle a_{\mathbf{k}}b_{\mathbf{k}}{}^\dagger\rangle,\\ \langle a_{\mathbf{k}}{}^\dagger b_{-\mathbf{k}}{}^\dagger\rangle&=\langle b_{-\mathbf{k}}{}^\dagger a_{\mathbf{k}}{}^\dagger\rangle=\langle b_{-\mathbf{k}}a_{\mathbf{k}}\rangle=\langle a_{\mathbf{k}}b_{-\mathbf{k}}\rangle,\end{aligned} \tag{A20}$$

and that all these expectation values are even in **k**.

APPENDIX B

For completeness we list here the constants η_i appearing in the text:

$$\eta_1=1+\tilde{K},\quad \eta_2=1+\tfrac{1}{2}\tilde{K},\quad \eta_3=1+\tilde{K}+2\tilde{L},\quad \eta_4=1+\tilde{K}-2\tilde{L},$$

$$\eta_5=1+\tilde{K}+4\tilde{L},\quad \eta_6=1+\tilde{K}-4\tilde{L},\quad \eta_7=1+\tilde{K}+2\tilde{L}\xi^2,\quad \eta_8=1+\tilde{K}-2\tilde{L}\xi^2,$$

$$\eta_9=1+\tfrac{1}{2}\tilde{K}-\tfrac{3}{2}\tilde{L}\xi^2,\quad \eta_{10}=1+\tfrac{1}{2}\tilde{K}-\tilde{L}\xi^2,\quad \eta_{11}=1+\tfrac{1}{2}\tilde{K}-5\tilde{L},\quad \eta_{12}=1+\tilde{K}-\tilde{L},$$

$$\eta_{13}=1+\tfrac{1}{2}\tilde{K}+\tilde{L},\quad \eta_{14}=1+\tfrac{1}{2}\tilde{K}-\tilde{L},\quad \eta_{15}=1+\tfrac{1}{2}\tilde{K}-\tfrac{5}{6}\tilde{L}.$$

We give here also the explicit expression for the term G in Eq. (92),

$$G=\tfrac{1}{8}NzJ\{(\eta_1-2\eta_2u^2)[1-(\mu+\tilde{\mu}_1)+\tfrac{1}{4}(\mu_1+\tilde{\mu}_2)^2+\tfrac{1}{4}(\mu_2+\tilde{\mu}_2)^2+\tfrac{1}{4}(\nu_2-\tilde{\nu}_2)^2]-4\tilde{L}(1-\tfrac{3}{2}u^2)$$
$$\times[1-(\mu_1+\tilde{\mu}_1)+\tfrac{1}{4}(\mu+\tilde{\mu}_1)^2+\tfrac{1}{8}(\nu_1-\tilde{\nu}_1)^2]+(1-\eta_2u^2)[-(\mu_2+\tilde{\mu}_2)+\tfrac{1}{2}(\mu_1+\tilde{\mu}_1)(\mu_2+\tilde{\mu}_2)+\tfrac{1}{4}(\nu_1+\tilde{\nu}_1)(\nu_2+\tilde{\nu}_2)]+\eta_2u^2$$
$$\times[(\nu_2+\tilde{\nu}_2)+\tfrac{1}{2}(\mu_1+\tilde{\mu}_1)(\nu_2+\tilde{\nu}_2)-\tfrac{1}{4}(\nu_1+\tilde{\nu}_1)(\mu_2+\tilde{\mu}_2)]+\tfrac{3}{2}\tilde{L}u^2[-(\nu_1+\tilde{\nu}_1)+\tfrac{1}{2}(\mu_1+\tilde{\mu}_1)(\nu_1+\tilde{\nu}_1)]\},$$

where

$$\mu_1=\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(A-e_{\mathbf{k}})}{\omega_1(\mathbf{k})}, \qquad \tilde{\mu}_1=\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(A+e_{\mathbf{k}})}{\omega_2(\mathbf{k})},$$

$$\mu_2=\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(A-e_{\mathbf{k}})}{\omega_1(\mathbf{k})}\gamma_{\mathbf{k}}, \qquad \tilde{\mu}_2=-\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(A+e_{\mathbf{k}})}{\omega_2(\mathbf{k})}\gamma_{\mathbf{k}},$$

$$\nu_1=\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(B-d_{\mathbf{k}})}{\omega_1(\mathbf{k})}, \qquad \tilde{\nu}_1=\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(B+d_{\mathbf{k}})}{\omega_2(\mathbf{k})},$$

$$\nu_2=\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(B-d_{\mathbf{k}})}{\omega_1(\mathbf{k})}\gamma_{\mathbf{k}}, \qquad \tilde{\nu}_2=-\frac{2}{N}\sum_{\mathbf{k}}\frac{SzJ(B+d_{\mathbf{k}})}{\omega_2(\mathbf{k})}\gamma_{\mathbf{k}}.$$

Errata

Paramagnetic-Resonance Study of Crystal-Field and Exchange Effects in an Excited State of TmN, BERNARD R. COOPER, R. C. FEDDER, AND D. P. SCHUMACHER [Phys. Rev. **163,** 506 (1967)]. The numerical values found for W and x in the point-charge calculation as given at the top of p. 515 are incorrect. This resulted from using an incorrect value for d, the experimental distance from a Tm site to nearest-neighbor nitrogen sites. The relationship between W and n should be $W/k=-0.786n$, rather than that given by Eq. (48) on p. 515. Thus, the experimental value of W would require a charge of 4.4 electronic charges on the surrounding nitrogen sites. The correct value of x for the point-charge calculation is -0.929, only slightly different from that previously given on p. 515. We are grateful to Dr. S. K. Malik for bringing this error to our attention.

Electric Field Gradients in Dilute Alloys of Copper with Nonmagnetic Transition Impurities, M. T. BÉAL-MONOD [Phys. Rev. **164,** 360 (1967)]. A misprint was introduced in Eq. (4), which should read as follows:

$$\Delta\rho=(4\pi c/Z_0k_F)\sum_l l\,\sin^2(\eta_{l-1}-\eta_l)_{E_F}. \qquad (4)$$