

Helical Spin Ordering—I Theory of Helical Spin Configurations

TAKEO NAGAMIYA

*Department of Material Physics, Faculty of Engineering Science, Osaka University
Toyonaka, Japan*

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Introduction

The development of the study of helical spin ordering is a rather recent event. The first report on this subject appeared in 1958 at the International Congress on Magnetism in Grenoble, France, in a talk given by the present author. This report contained the prediction of a helical spin arrangement in MnO_2 , the theory having been worked out by A. Yoshimori in collaboration with the writer. Earlier observations on MnO_2 with neutron diffraction made by R. A. Erickson, which had given puzzling results, gave support to this prediction. It appeared that the report received little attention at that time. Since then, however, observations with neutron diffraction of satellite lines in chromium metal, made at Brookhaven, and findings at Oak Ridge of such lines in heavy rare earth metals that correspond to helical and modified-helical spin arrangements, as well as a study of MnAu_2 under external magnetic field at Saclay, gave a good deal of impetus to a further development of the subject both theoretically and experimentally. In the meantime, the spin arrangement in chromium, which is sinusoidal, has come to be regarded as a spin density wave of the conduction electrons as proposed by A. W. Overhauser.

In the present article, the writer has made an effort to give a complete description of the theory of helical and modified-helical spin ordering, confining himself, however, to molecular field treatments (except for a note in Section VIII) and spin-wave calculations. Modifications of a helical spin order arise from anisotropy energies and an external magnetic field. Also, a description of the theory of complex helical spin configurations in complex crystalline lattices is given. A basic assumption made is that there exist isotropic exchange interactions between atomic spin moments of further neighbors as well as between neighboring moments. The coefficients of these exchange interactions are assumed as given constants. In this sense, the spin system dealt with may be called the Heisenberg magnet. Experimental observations relevant to the theory are referred to as far as they are known to the writer and are useful to elucidate the theory.

Part 2 of this article, in preparation, will deal with the role of conduction electrons in the exchange interaction, will review and discuss the observed magnetic and other properties of heavy and light rare-earth metals, and finally will discuss the spin density wave in chromium.

Mention should be made that the original plan of writing this article was laid in 1963 while the writer was enjoying a stay in the Department of Physics of the University of California at Berkeley as the result of an

invitation extended by Professors C. Kittel and A. M. Portis. The writer would like to take this opportunity to express his warm thanks to these individuals. The text presented here deviates almost completely from the writer's lecture notes at Berkeley. Part 2 however, will cover the latter to a large extent.

I. Elementary Theory of Helical Spin Ordering

In a ferromagnet that consists of atomic spins coupled with each other by exchange forces, the spin vectors are aligned parallel to each other, and there are spin-wave excitations. A spin wave is a motion such that a spin at a position \mathbf{R}_n precesses about the direction of the alignment with a phase angle $\mathbf{q} \cdot \mathbf{R}_n - \omega t$, where \mathbf{q} is the wave vector of the spin wave and ω its frequency, so that the spin vectors describe at each instant a helix in space. In order that the ferromagnetic state be stable, ω^2 must be a positive function of \mathbf{q} . If, however, ω vanishes for a certain \mathbf{q} , one may imagine that the spin wave with this \mathbf{q} would stand still with an arbitrary amplitude. One may further ask what would happen if ω^2 were negative in a certain range of \mathbf{q} . The ferromagnetic state would then be unstable, and a helical ordering of the spins with the maximum possible amplitude would be realized as the stable state.

Yoshimori¹ looked into this problem in detail and predicted the existence of a helical ordering of spins in rutile-type crystals having magnetic ions as cations and nonmagnetic ions as anions. He analyzed the neutron diffraction lines of MnO_2 observed by Erickson^{2a} and found that its spin structure is helical; the spins in the same c -plane of this crystal point in the same direction perpendicular to the c -axis, and this direction turns from one plane to the next by an angle of 129° ($= 5\pi/7$). It had already been pointed out^{2b} that in MnF_2 , which is also of rutile structure and has a collinear antiferromagnetic spin arrangement, the spin-wave frequencies become negative in a region of \mathbf{q} -space for a certain range of exchange constants. Soon after Yoshimori published his paper, Kaplan³ proposed a simple theory of helical ordering to interpret the neutron diffraction lines from chromium, and Villain⁴ also predicted helical ordering by a treatment of molecular field. Although the three papers have common features, Yoshimori's paper has been worked out most extensively, including discussions of anisotropy energy, susceptibility, the spin-wave spectrum, and neutron diffraction. Yoshimori confined himself to rutile-type crystals,

¹ A. Yoshimori, *J. Phys. Soc. Japan* **14**, 807 (1959); T. Nagamiya, *Proc. Colloq. Intern. Magnétisme, Grenoble, 1958*, p. 15 [or see *J. Phys. Radium* **20**, 70 (1959)].

^{2a} R. A. Erickson, private communication (1957) (cited in Yoshimori¹).

^{2b} T. Nakamura and O. Nagai, private communication (1957).

³ T. A. Kaplan, *Phys. Rev.* **116**, 888 (1959).

⁴ J. Villain, *Phys. Chem. Solids* **11**, 303 (1959).

but his theory is applicable to all lattices with one magnetic atom per unit cell.

1. SIMPLE HELICAL SPIN ORDERING AT ABSOLUTE ZERO

Consider a lattice of magnetic atoms, such as that of manganese in MnF_2 and MnO_2 , in which the unit cell can be so chosen that it contains only one magnetic atom. We put aside anisotropy energy for a moment and consider only exchange forces. On each magnetic atom we assume a classical spin. Between spins \mathbf{S}_m and \mathbf{S}_n at positions \mathbf{R}_m and \mathbf{R}_n , an exchange potential of the form

$$-2J(\mathbf{R}_{mn})\mathbf{S}_m \cdot \mathbf{S}_n \quad (\mathbf{R}_{mn} = \mathbf{R}_m - \mathbf{R}_n) \quad (1.1)$$

will be assumed, where $J(-\mathbf{R}_{mn}) = J(\mathbf{R}_{mn})$. It is essential that this exchange potential is not confined to nearest neighbors in order that we obtain a helical ordering, as we shall soon see. We make Fourier transformations of the exchange coefficients and spins:

$$J(\mathbf{q}) = \sum_n J(\mathbf{R}_n) \exp(-i\mathbf{q} \cdot \mathbf{R}_n), \quad (1.2)$$

$$\mathbf{S}_q = N^{-1/2} \sum_n \mathbf{S}_n \exp(-i\mathbf{q} \cdot \mathbf{R}_n) \quad (\mathbf{S}_{-\mathbf{q}} = \mathbf{S}_q^*), \quad (1.3)$$

where we denote by N the total number of atoms and we assume that one atom is at the origin and $J(\mathbf{R}_n) = 0$ for $\mathbf{R}_n = 0$. It is easy to see that $J(-\mathbf{q}) = J(\mathbf{q})$. The total exchange energy

$$-\sum_m \sum_n J(\mathbf{R}_{mn}) \mathbf{S}_m \cdot \mathbf{S}_n \quad (1.4)$$

can then be written as

$$-\sum_q J(\mathbf{q}) \mathbf{S}_q \cdot \mathbf{S}_{-\mathbf{q}}. \quad (1.5)$$

We look for the lowest minimum of (1.4) or (1.5) subject to the condition $\mathbf{S}_n^2 = \text{const} = S^2$ for all n . Instead of this condition, we shall first impose a milder condition

$$\sum_n \mathbf{S}_n^2 = \text{const}, \quad (1.6)$$

which can be written with Fourier components as

$$\sum_q \mathbf{S}_q \cdot \mathbf{S}_{-\mathbf{q}} = \text{const}. \quad (1.7)$$

Under this milder condition, the minimum of (1.5) is attained simply by taking only that \mathbf{q} for which $J(\mathbf{q})$ is the highest maximum. Denoting this \mathbf{q} by \mathbf{Q} ($\mathbf{q} = -\mathbf{Q}$ being equally allowed), we have the minimum value

of (1.5) as^{4a}

$$-J(\mathbf{Q})(\mathbf{S}_Q \cdot \mathbf{S}_{-Q} + \mathbf{S}_{-Q} \cdot \mathbf{S}_Q). \quad (1.8)$$

Also, we obtain from (1.3)

$$\mathbf{S}_n = N^{-1/2}[\mathbf{S}_Q \exp(i\mathbf{Q} \cdot \mathbf{R}_n) + \mathbf{S}_{-Q} \exp(-i\mathbf{Q} \cdot \mathbf{R}_n)]. \quad (1.9)$$

This equation can be written in components as

$$\begin{aligned} S_{nx} &= A \cos(\mathbf{Q} \cdot \mathbf{R}_n + \alpha), \\ S_{ny} &= B \cos(\mathbf{Q} \cdot \mathbf{R}_n + \beta), \\ S_{nz} &= C \cos(\mathbf{Q} \cdot \mathbf{R}_n + \gamma), \end{aligned} \quad (1.10)$$

where A , B , and α , β , γ are arbitrary constants. Equations (1.10) represent a general elliptic helical ordering of spins with wave vector \mathbf{Q} : that is, the spin vector \mathbf{S}_n rotates and describes an ellipse as the position \mathbf{R}_n advances in the direction of \mathbf{Q} . Now, to satisfy the conditions $\mathbf{S}_n^2 = \text{const}$, this ellipse must be a circle. Taking the z -axis perpendicular to the plane of the circle, we have, instead of (1.10),

$$\begin{aligned} S_{nx} &= S \cos(\mathbf{Q} \cdot \mathbf{R}_n + \alpha), \\ S_{ny} &= S \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha), \\ S_{nz} &= 0. \end{aligned} \quad (1.11)$$

The corresponding energy, (1.8), is calculated, with the use of (1.3), to be

$$-NS^2J(\mathbf{Q}). \quad (1.12)$$

Yoshimori called the spin ordering represented by (1.11) the *screw structure* of spins, since *screw* means a combination of rotation and translation. Generally, (1.11) represents an oblique screw structure, as the direction of \mathbf{Q} is not necessarily perpendicular to the rotation plane. The relative orientation of the screw axis and the rotation plane may be fixed by an anisotropy energy; for instance, when there is an easy plane of magnetization in the crystal, the spins will rotate in this plane, whereas the direction of \mathbf{Q} may be determined by an anisotropy in $J(\mathbf{R}_{mn})$. The structure in which the rotation plane is perpendicular to \mathbf{Q} may be called the *proper screw structure* and the structure in which they are parallel the *cycloidal structure* (after Yoshimori).

A characteristic feature of the screw structure, or the *helical arrangement of spins*, as it is more commonly called, is that the period of the arrange-

^{4a} When $\mathbf{Q} = 0$ or \mathbf{Q} is a vector at such a special point on the Brillouin zone boundary that \mathbf{Q} and $-\mathbf{Q}$ are equivalent vectors, there is an additional factor $\frac{1}{2}$ on the right-hand side of (1.8) and of (1.9). In this case, too, (1.12) is valid.

ment is, in general, incommensurable with the lattice period, since the magnitude of \mathbf{Q} is determined solely by the exchange coefficients.

Example: Consider a layer crystal with interlayer spacing c . The direction of \mathbf{Q} will be assumed to be perpendicular to the layers. We sum $J(\mathbf{R}_{mn})$ over all sites m in the layer that contains the site n and denote this sum by J_0 ; the sum over a neighboring layer we denote by J_1 ; for a next-neighboring layer, we define J_2 , and so on. Then, for a \mathbf{q} that is perpendicular to the layers, we can write

$$\begin{aligned} J(q) &= \sum_{\nu=-\infty}^{\infty} J_\nu \exp(-ivcq) \\ &= J_0 + 2J_1 \cos(cq) + 2J_2 \cos(2cq) + \dots \end{aligned} \quad (1.13)$$

If we retain only J_0 , J_1 , and J_2 , we have from $J(q) = \max$ the following equation with $q = Q$:

$$[J_1 + 4J_2 \cos(cQ)] \sin(cQ) = 0.$$

This gives solutions $Q = 0$ (ferromagnetic spin arrangement), $Q = \pi/c$ (antiferromagnetic spin arrangement), and a helical arrangement having \mathbf{Q} given by

$$\cos(cQ) = -J_1/4J_2 \quad \text{when } |J_1| < |4J_2|. \quad (1.14)$$

It can be seen easily that the value of $J(Q) = J_0 - (J_1^2/4J_2) - 2J_2$ calculated from (1.14) is greater than both $J(0) = J_0 + 2J_1 + 2J_2$ and $J(\pi/c) = J_0 - 2J_1 + 2J_2$, provided that J_2 is negative. Thus, when $J_2 < 0$ and $|J_1| < |4J_2|$, the helical state has the lowest energy. When J_2 is positive, the ferromagnetic or antiferromagnetic state is more stable according as J_1 is positive or negative; this is because spins on next-neighboring layers wish to be parallel or antiparallel, depending on the sign of J_1 . On the other hand, for sufficiently large negative J_2 , there must be a compromise between the forces acting between next-neighboring layers to make spins antiparallel and the forces acting between neighboring layers to make spins either parallel or antiparallel, and as a result one has a helical arrangement.

2. MOLECULAR FIELD THEORY FOR FINITE TEMPERATURE

The effect of a finite temperature, T , may be considered with the approximation of the Weiss molecular field. Denoting the thermal average of \mathbf{S}_m by $\langle \mathbf{S}_m \rangle$, we can write the exchange magnetic field acting on \mathbf{S}_n as

$$\mathbf{H}_{ex,n} = -(g\mu_B)^{-1} 2 \sum_m J(\mathbf{R}_{mn}) \langle \mathbf{S}_m \rangle. \quad (2.1)$$

Here g is the spectroscopic splitting factor and $\mu_B = e\hbar/2mc$ the Bohr

magneton. When there is, further, an external field \mathbf{H} , the effective field acting on \mathbf{S}_n is given by $\mathbf{H} + \mathbf{H}_{\text{ex},n}$. The thermal average of \mathbf{S}_n must point in the direction of this field, and its magnitude is given by

$$|\langle \mathbf{S}_n \rangle| = S\sigma_n = \frac{\sum_{m=-S}^S m \exp(g\mu_B m |\mathbf{H} + \mathbf{H}_{\text{ex},n}|/kT)}{\sum_{m=-S}^S \exp(g\mu_B m |\mathbf{H} + \mathbf{H}_{\text{ex},n}|/kT)}, \quad (2.2)$$

namely,

$$\sigma_n = B_S(x_n) = \frac{2S+1}{2S} \coth \frac{2S+1}{2S} x_n - (2S)^{-1} \coth(2S)^{-1} x_n, \quad (2.3)$$

$$x_n = g\mu_B S |\mathbf{H} + \mathbf{H}_{\text{ex},n}|/kT, \quad (2.4)$$

where $B_S(x)$ is the Brillouin function. When $\mathbf{H} = 0$, we may anticipate a helical ordering as a solution of Eqs. (2.3) and (2.4), and we may put $\sigma_n = \text{const} = \sigma$. Then, (2.3) and (2.4) become

$$\sigma = B_S(x), \quad x = g\mu_B S |\mathbf{H}_{\text{ex}}|/kT, \quad (2.5)$$

where, from (2.1) and $\langle \mathbf{S}_m \rangle = S\sigma \exp[i(\mathbf{Q} \cdot \mathbf{R}_m + \alpha)]$, $\langle \mathbf{S}_m \rangle$ being expressed as a vector in the complex plane, one has

$$\begin{aligned} |\mathbf{H}_{\text{ex}}| &= (g\mu_B)^{-1} 2 \left| \sum_m J(\mathbf{R}_{mn}) S\sigma \exp[i(\mathbf{Q} \cdot \mathbf{R}_m + \alpha)] \right| \\ &= (g\mu_B)^{-1} 2 \left| \sum_m J(\mathbf{R}_{mn}) S\sigma \exp(i\mathbf{Q} \cdot \mathbf{R}_{mn}) \cdot \exp[i(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)] \right| \\ &= (g\mu_B)^{-1} 2J(\mathbf{Q}) S\sigma. \end{aligned} \quad (2.6)$$

Hence, from (2.5) follows

$$\sigma = B_S[2J(\mathbf{Q}) S^2 \sigma / kT]. \quad (2.7)$$

Since $B_S(x) = [(S+1)/3S]x - O(x^3)$ for small x , this gives immediately the Néel temperature by

$$T_N = [S(S+1)/3k]2J(\mathbf{Q}). \quad (2.8)$$

Here \mathbf{Q} has to be such that $J(\mathbf{Q})$ is maximum (and positive), since then this equation gives the highest, and thus the real, Néel temperature. Below this Néel temperature, σ is given as the solution of (2.7). It varies with T like the saturation magnetization of a ferromagnet.

Above the Néel temperature, the thermal average of each spin vanishes for no external field. For a finite field, the thermal averages of the spin vectors should be equal and parallel to the field direction; hence we put $\mathbf{Q} = 0$ in (2.6) and obtain from (2.3) and (2.4)

$$\sigma = B_S\{[g\mu_B SH + 2J(0)S^2\sigma]/kT\}, \quad (2.9)$$

which determines σ as a function of H and T . For small H/T , we have

$$\sigma = [(S + 1)/3SkT][g\mu_B SH + 2J(0)S^2\sigma],$$

so that

$$\chi = g\mu_B S\sigma/H = C/(T - \theta_p), \quad (2.10)$$

$$C = (g\mu_B)^2 S(S + 1)/3k, \quad \theta_p = 2[S(S + 1)/3k]J(0), \quad (2.11)$$

where χ is the susceptibility.

The effect of an external field below the Néel temperature will be discussed in Section IV.

II. Spin Waves in the Screw Structure

We shall discuss here the modes and frequencies of spin waves that can be excited in the proper screw structure.^{1,5,6} We shall assume the screw axis to be in the z -direction so that the spins rotate in the xy -plane as the position advances in the z -direction, as expressed by (1.11). We can introduce an anisotropy energy for each spin of the form DS_z^2 with positive D , without disturbing the assumed proper screw structure. This anisotropy energy gives an effective field of $-2DS_z$ in the z -direction for each spin.

For convenience of calculation, we shall introduce a local coordinate system ξ, η, ζ in such a way that the ζ -axis coincides with the equilibrium spin direction at each lattice point, the ξ -axis is perpendicular to this direction in the xy -plane, and the η -axis is parallel to the z -axis. Then the relative orientation of the ζ, ξ -axes at \mathbf{R}_m and those at \mathbf{R}_n is such that the former is rotated from the latter by an angle of $\mathbf{Q} \cdot \mathbf{R}_{mn}$. We have, therefore, the coordinate transformation

$$\begin{aligned} \zeta_n &= \zeta_m \cos(\mathbf{Q} \cdot \mathbf{R}_{mn}) - \xi_m \sin(\mathbf{Q} \cdot \mathbf{R}_{mn}), \\ \xi_n &= \zeta_m \sin(\mathbf{Q} \cdot \mathbf{R}_{mn}) + \xi_m \cos(\mathbf{Q} \cdot \mathbf{R}_{mn}). \end{aligned} \quad (II.1)$$

For small spin oscillations, the local ζ -component of each spin can be regarded as a constant, S , and the equation of motion (torque equation) can be written as

$$\begin{aligned} \hbar \dot{S}_{n\xi} &= S_{n\eta} H_{n\xi} - S_{n\xi} H_{n\eta}, \\ \hbar \dot{S}_{n\eta} &= S_{n\xi} H_{n\xi} - S_{n\xi} H_{n\eta}, \end{aligned} \quad (II.2)$$

where we have omitted a factor $-g\mu_B$ in the right-hand side. ξ, ξ stand for ζ_n, ξ_n , so that $S_{n\xi} = S$, and $H_{n\xi}, H_{n\eta}, H_{n\xi}$ are the three components of the effective field acting on the n th spin. This effective field consists of

⁵ K. Yosida and H. Miwa, *J. Appl. Phys.* **32**, 88 (1961).

⁶ T. A. Kaplan, *Phys. Rev.* **124**, 329 (1961).

the exchange field given by (2.1), the thermal average sign being dropped, and the anisotropy field $-2DS_{n\eta}$, namely,

$$\begin{aligned} H_{n\xi} &= \sum_m 2J(\mathbf{R}_{mn}) [S_{m\xi} \cos (\mathbf{Q} \cdot \mathbf{R}_{mn}) - S_{m\xi} \sin (\mathbf{Q} \cdot \mathbf{R}_{mn})], \\ H_{n\xi} &= \sum_m 2J(\mathbf{R}_{mn}) [S_{m\xi} \sin (\mathbf{Q} \cdot \mathbf{R}_{mn}) + S_{m\xi} \cos (\mathbf{Q} \cdot \mathbf{R}_{mn})], \\ H_{n\eta} &= \sum_m 2J(\mathbf{R}_{mn}) S_{m\eta} - 2DS_{n\eta}. \end{aligned} \quad (\text{II.3})$$

A factor $-(g\mu_B)^{-1}$ was omitted in the right-hand side, and we understand by suffixes ξ and η to H_n and S_m the respective local coordinates, i.e., ξ_n , ξ_m and ξ_m , ξ_m , respectively. Replacing $S_{n\xi}$ and $S_{m\xi}$ by S in (II.3) and writing

$$S_{n\xi} + iS_{n\eta} = (2S)^{1/2}a_n, \quad S_{n\xi} - iS_{n\eta} = (2S)^{1/2}a_n^*, \quad (\text{II.4})$$

we have from (II.2) and (II.3) the following equation, after neglecting second-order terms in a and a^* :

$$\begin{aligned} \hbar\dot{a}_n &= -2iSa_n \left[\sum_m J(\mathbf{R}_{mn}) \cos (\mathbf{Q} \cdot \mathbf{R}_{mn}) \right] \\ &\quad + iS \sum_m J(\mathbf{R}_{mn}) (a_m + a_m^*) \cos (\mathbf{Q} \cdot \mathbf{R}_{mn}) \\ &\quad + iS \sum_m J(\mathbf{R}_{mn}) (a_m - a_m^*) - iSD(a_n - a_n^*). \end{aligned} \quad (\text{II.5})$$

A similar equation in which a and a^* are interchanged and i is replaced by $-i$ is also obtained. These equations are further simplified by making the Fourier transformation:

$$\begin{aligned} a_{\mathbf{q}} &= N^{-1/2} \sum_n a_n \exp (-i\mathbf{q} \cdot \mathbf{R}_n), \\ a_{-\mathbf{q}}^* &= N^{-1/2} \sum_n a_n^* \exp (-i\mathbf{q} \cdot \mathbf{R}_n). \end{aligned} \quad (\text{II.6})$$

Observing that $\exp(-i\mathbf{q} \cdot \mathbf{R}_n) = \exp(i\mathbf{q} \cdot \mathbf{R}_{mn}) \cdot \exp(-i\mathbf{q} \cdot \mathbf{R}_m)$ and referring to (1.2), we obtain from (II.5)

$$\begin{aligned} \hbar\dot{a}_{\mathbf{q}} &= -2iSa_{\mathbf{q}}J(\mathbf{Q}) \\ &\quad + \frac{1}{2}iS(a_{\mathbf{q}} + a_{-\mathbf{q}}^*)[J(\mathbf{Q} + \mathbf{q}) + J(\mathbf{Q} - \mathbf{q})] \\ &\quad + iS(a_{\mathbf{q}} - a_{-\mathbf{q}}^*)[J(\mathbf{q}) - D], \\ \hbar\dot{a}_{-\mathbf{q}}^* &= +2iSa_{-\mathbf{q}}^*J(\mathbf{Q}) \\ &\quad - \frac{1}{2}iS(a_{\mathbf{q}} + a_{-\mathbf{q}}^*)[J(\mathbf{Q} + \mathbf{q}) + J(\mathbf{Q} - \mathbf{q})] \\ &\quad + iS(a_{\mathbf{q}} - a_{-\mathbf{q}}^*)[J(\mathbf{q}) - D]. \end{aligned} \quad (\text{II.7})$$

Putting $\dot{a}_q = -i\omega_q a_q$ and $\dot{a}_{-q}^* = -i\omega_q a_{-q}^*$ and solving the resulting homogeneous equations for a_q and a_{-q}^* , we finally obtain the frequency

$$\hbar\omega_q = 2S\{[J(\mathbf{Q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q})] \cdot [J(\mathbf{Q}) - J(\mathbf{q}) + D]\}^{1/2} \quad (\text{II.8})$$

and the amplitude ratio

$$\begin{aligned} (a_q + a_{-q}^*)/(a_q - a_{-q}^*) &= S_{q\xi}/iS_{q\eta} \\ &= [J(\mathbf{Q}) - J(\mathbf{q}) + D]^{1/2}/[J(\mathbf{Q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q})]^{1/2}. \end{aligned} \quad (\text{II.9})$$

In (II.9), $S_{q\xi}$ and $S_{q\eta}$ are the Fourier transforms of $S_{n\xi}$ and $S_{n\eta}$, respectively.

In this way, we have obtained formulas for the spin waves to be excited in the proper screw structure. ω_q given by (II.8) vanishes for $\mathbf{q} = 0$, in which case $S_{q\eta}$ also vanishes, as can be seen from (II.9), and the mode of oscillation is such that the whole spin system rotates as a rigid body about the screw axis. Another special case of interest is $\mathbf{q} = \pm\mathbf{Q}$ when $D = 0$, in which case, too, the frequency vanishes and $S_{q\xi}$ also vanishes. In this case, the spins oscillate with phase angles $\mathbf{q} \cdot \mathbf{R}_n$ in the direction parallel to the screw axis; this implies a small canting of the plane in which the spin vectors rotate. If D is small but not zero, the spin waves with $\mathbf{q} = \pm\mathbf{Q}$ have a small but finite frequency, and the spin vectors oscillate elliptically with a small amplitude perpendicular to the screw axis and a large amplitude parallel to it. In this case, there is an oscillating component of the total spin perpendicular to the screw axis, so that the mode should be active to an imposed oscillating electromagnetic field. This can be seen in the following way. Since the x -component of the oscillating part of the n th spin is $-S_{n\xi} \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$, the total x -component can be calculated to be

$$\begin{aligned} \sum_n S_{nx} &= \sum_n -S_{n\xi} \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) \\ &= \sum_n -N^{-1/2} S_{q\xi} \exp(i\mathbf{q} \cdot \mathbf{R}_n) \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) \\ &= \mp(i/2) N^{1/2} S_{q\xi} \exp(\mp i\alpha) \quad \text{for } \mathbf{q} = \pm\mathbf{Q}. \end{aligned} \quad (\text{II.10})$$

Thus, the amplitude of the total x -component is $\frac{1}{2}N^{1/2} |S_{q\xi}|$, which is nonvanishing for $D \neq 0$.

In the Holstein-Primakoff formalism, the spin operators are written with annihilation and creation Bose-operators, a_n and a_n^* , as

$$S_{n\xi} + iS_{n\eta} = (2S)^{1/2}(1 - a_n^* a_n / 2S)^{1/2} a_n, \quad (\text{II.11})$$

$$S_{n\xi} - iS_{n\eta} = (2S)^{1/2} a_n^* (1 - a_n^* a_n / 2S)^{1/2},$$

$$S_{n\xi} = S - a_n^* a_n, \quad (\text{II.12})$$

where

$$[a_n, a_n^*] = 1. \quad (\text{II.13})$$

This way of writing spin operators in terms of a_n and a_n^* is exact so long as all the states with the number of Bose particles greater than $2S$ are disregarded. If the factor $(1 - a_n^* a_n / 2S)^{1/2}$ is approximated by 1, Eqs. (II.11) reduce to Eqs. (II.4).

From (II.6) and (II.13) follows also

$$[a_q, a_q^*] = 1. \quad (\text{II.14})$$

The procedure of solving Eqs. (II.7) is equivalent to transforming a_q and a_q^* to a new set of variables (the normal coordinates), α_q and α_q^* :

$$\begin{aligned} a_q &= \alpha_q \cosh \theta_q + \alpha_{-q}^* \sinh \theta_q, \\ a_q^* &= \alpha_q^* \cosh \theta_q + \alpha_{-q} \sinh \theta_q, \end{aligned} \quad (\text{II.15})$$

where

$$\coth 2\theta_q = (A + B)/(A - B), \quad (\text{II.16})$$

$$\begin{aligned} A &= J(\mathbf{Q}) - J(\mathbf{q}) + D, \\ B &= J(\mathbf{Q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q}). \end{aligned} \quad (\text{II.17})$$

To see the foregoing, one may notice first that

$$[\alpha_q, \alpha_q^*] = 1, \quad (\text{II.18})$$

which follows from $\cosh^2 \theta_q - \sinh^2 \theta_q = 1$. Second, from (II.9), the transformation coefficients $\cosh \theta_q$ and $\sinh \theta_q$ have to be in the ratio

$$\cosh \theta_q / \sinh \theta_q = (A^{1/2} + B^{1/2}) / (A^{1/2} - B^{1/2}),$$

from which follows $\coth 2\theta_q = (A + B)/(A - B)$.

The thermal average length of each spin in the helical ordering can be calculated in the following way. By (II.12) and (II.6),

$$\langle S_{n\xi} \rangle = S - \langle a_n^* a_n \rangle = S - N^{-1} \sum_{q,q'} \langle a_q^* a_{q'} \rangle \exp [i(\mathbf{q}' - \mathbf{q}) \cdot \mathbf{R}_n],$$

which can be written with α_q and α_q^* as

$$\begin{aligned} S - N^{-1} \sum_{q,q'} &\langle (\alpha_q^* \cosh \theta_q + \alpha_{-q} \sinh \theta_q) (\alpha_{q'} \cosh \theta_{q'} + \alpha_{-q'}^* \sinh \theta_{q'}) \rangle \\ &\cdot \exp [i(\mathbf{q}' - \mathbf{q}) \cdot \mathbf{R}_n]. \end{aligned}$$

Since there is no phase relation between spin waves with different \mathbf{q} 's, terms in this expression other than those with $\mathbf{q} = \mathbf{q}'$ vanish; in other words, given a set of boson occupation numbers, $\{n_q\}$, where $n_q = \alpha_q^* \alpha_q$, the traces of $\alpha_q^* \alpha_{q'}$ and $\alpha_{-q} \alpha_{-q'}^*$ vanish for $\mathbf{q} \neq \mathbf{q}'$, and the traces of $\alpha_q^* \alpha_{-q'}$

and $\alpha_{-\mathbf{q}}\alpha_{\mathbf{q}'}$ all vanish. Thus, we are left with

$$S = N^{-1} \sum_{\mathbf{q}} [\langle n_{\mathbf{q}} \rangle \cosh^2 \theta_{\mathbf{q}} + (\langle n_{\mathbf{q}} \rangle + 1) \sinh^2 \theta_{\mathbf{q}}],$$

or

$$S = N^{-1} \sum_{\mathbf{q}} [(\langle n_{\mathbf{q}} \rangle + \frac{1}{2}) \cosh 2\theta_{\mathbf{q}} - \frac{1}{2}], \quad (\text{II.19})$$

where

$$\langle n_{\mathbf{q}} \rangle = [\exp(\hbar\omega_{\mathbf{q}}/kT) - 1]^{-1}. \quad (\text{II.20})$$

At low temperatures, only those spin waves that have small $\omega_{\mathbf{q}}$ are important. Confining ourselves to this case and assuming that D is non-vanishing, we may consider only small values of \mathbf{q} . If \mathbf{q} is small, $\coth 2\theta_{\mathbf{q}}$ can be approximated by $1 + 2(B/A)$, since A and B can be approximated as

$$A = J(\mathbf{Q}) - J(0) + D, \quad B = -\frac{1}{2} \sum_{i,j} [\partial^2 J(\mathbf{q})/\partial q_i \partial q_j] |_{\mathbf{Q}} \times q_i q_j,$$

where $i, j = x, y, z$, so that B is small (positive). $\cosh 2\theta_{\mathbf{q}}$ is then approximately equal to $(4B/A)^{-1/2}$. So we obtain finally for the thermal average length of each spin

$$\begin{aligned} S &= (16\pi^3 N)^{-1} \iiint (\cosh 2\theta_{\mathbf{q}} - 1) d^3 \mathbf{q} - (8\pi^3 N)^{-1} \iiint \langle n_{\mathbf{q}} \rangle \cosh 2\theta_{\mathbf{q}} d^3 \mathbf{q} \\ &= \text{const} - (8\pi^3 N)^{-1} \iiint \left(\frac{A}{4B}\right)^{1/2} \frac{d^3 \mathbf{q}}{\exp(\hbar\omega_{\mathbf{q}}/kT) - 1}. \end{aligned} \quad (\text{II.21})$$

The constant term represents the spin length contracted by zero-point motion, and the second term is proportional to T^2 , since $\hbar\omega_{\mathbf{q}} = 2S(AB)^{1/2}$ and $B^{1/2}$ is proportional to $|\mathbf{q}|$.

III. Effect of Anisotropy Energy on Spin Configuration

In this part, we shall be concerned with the alteration of a helical spin configuration by anisotropy energy. First, we consider an anisotropy energy with uniaxial symmetry giving rise to an *easy axis* of spin orientation, second, an anisotropy energy of twofold, fourfold, or sixfold symmetry giving rise to a sort of *easy plane*, and third, a more complex anisotropy energy giving rise to an *easy cone*. We assume that all these anisotropy energies are of one-atom type originating in the crystalline electric field acting on individual atoms and perturbing their spin states. Other anisotropy energies, such as magnetic dipolar,⁷ electric quadrupolar,^{7,8} and

⁷ R. J. Elliott, *Phys. Rev.* **124**, 346 (1961).

⁸ H. Miwa and K. Yosida, *Progr. Theoret. Phys. (Kyoto)* **26**, 693 (1961).

anisotropic exchange⁹ interactions will not be considered. The theory of the effect of anisotropy energy on the helical spin configuration has been developed by Kaplan,⁶ Elliott,⁷ and Miwa and Yosida.⁸ They explained successfully the various spin configurations and phase changes observed in heavy rare-earth metals. The description given here is somewhat different from theirs and somewhat more general, although essentially the same.

3. UNIAXIAL ANISOTROPY ENERGY WITH AN EASY AXIS; OSCILLATION IN THE LONGITUDINAL COMPONENT

The easy axis will be denoted as z . The simplest form of this type of anisotropy energy may be written as

$$w(S_z) = D[S_z^2 - \frac{1}{3}S(S+1)], \quad (3.1)$$

with *negative* D , which vanishes for $S = \frac{1}{2}$, however. The general form of the anisotropy energy to be considered can be written as an even polynomial of S_z of degree at most equal to $2S$.^{9a} With this general function, $w(S_z)$, we shall proceed to the approximation of the Weiss molecular field.

The exchange field acting on the n th spin is given by (2.1). For brevity, we shall omit the factor $-(g\mu_B)^{-1}$ from it (and consequently the factor $-g\mu_B$ from the magnetic moment) and also eliminate the suffix "ex." Then, it is given in components as

$$H_{ni} = 2 \sum_m J(R_{mn}) S_{\sigma_{mi}}, \quad S_{\sigma_{mi}} = \langle S_{mi} \rangle, \quad i = z, x, y. \quad (3.2)$$

The equations to determine σ_{ni} ($i = z, x, y$) are

$$S_{\sigma_{ni}} = \frac{\text{tr } S_i \exp \beta [H_{nz}S_z + H_{nx}S_x + H_{ny}S_y - w(S_z)]}{\text{tr } \exp \beta [H_{nz}S_z + H_{nx}S_x + H_{ny}S_y - w(S_z)]}, \quad (3.3)$$

where $\beta = 1/kT$ and "tr" means "trace." To find the Néel temperature and the behavior of the spin arrangement below but close to it, we assume the exchange field to be small, and we expand the exponential function in (3.3) in powers of it. If we set $H_{nx} = H_{ny} = 0$, a simple calculation yields the following result for the z -component:

$$S_{\sigma_{nz}} = \frac{\beta H_{nz} \text{tr } S_z^2 \exp [-\beta w(S_z)] + \frac{1}{6}\beta^3 H_{nz}^3 \text{tr } S_z^4 \exp [-\beta w] + \dots}{\text{tr } \exp [-\beta w(S_z)] + \frac{1}{2}\beta^2 H_{nz}^2 \text{tr } S_z^2 \exp [-\beta w] + \dots}. \quad (3.4)$$

If we retain only linear terms, we have simply

$$S_{\sigma_{nz}} = \beta H_{nz} \text{tr } S_z^2 \exp [-\beta w(S_z)] / \text{tr } \exp [-\beta w(S_z)],$$

⁹ T. A. Kaplan and D. H. Lyons, *Phys. Rev.* **129**, 2072 (1963).

^{9a} Any function of S_z takes $2S + 1$ eigenvalues and thus is expressible as a linear combination of $2S + 1$ bases, for which we may take 1, S_z , S_z^2 , ..., S_z^{2S} .

and substituting (3.2) into this we obtain a system of linear homogeneous equations for σ_{nz} 's. So we put

$$\sigma_{nz} = \sigma_z \cos (\mathbf{q} \cdot \mathbf{R}_n + \alpha),$$

and utilize the definition of $J(\mathbf{q})$, Eq. (1.2). With the relation

$$\mathbf{R}_m = \mathbf{R}_{mn} + \mathbf{R}_n,$$

we then have

$$\sigma_z = 2\beta J(\mathbf{q})\sigma_z \operatorname{tr} S_z^2 \exp [-\beta w] / \operatorname{tr} \exp [-\beta w].$$

Eliminating σ_z from both sides, we are left with the equation to determine the Néel temperature for the z -component of the spins. Since the last factor (the quotient of traces) is a decreasing function of T , being equal to S^2 for $T = 0$ and to $\frac{1}{3}S(S+1)$ for $T = \infty$, the highest Néel temperature, i.e., the real Néel temperature, is obtained for the maximum of $J(\mathbf{q})$. So we put $\mathbf{q} = \mathbf{Q}$ and have

$$kT_N = 2J(\mathbf{Q}) \{ \operatorname{tr} S_z^2 \exp [-w(S_z)/kT_N] / \operatorname{tr} \exp [-w(S_z)/kT_N] \}. \quad (3.5)$$

If one assumes $w(S-1) - w(S) \gg kT_N$, this equation reduces to

$$kT_N = 2J(\mathbf{Q})S^2, \quad (3.6)$$

and in the opposite case to

$$kT_N = 2J(\mathbf{Q}) \left\{ \frac{1}{3}S(S+1) + \frac{\operatorname{tr} [\frac{1}{3}S(S+1) - S_z^2]w(S_z)}{(2S+1)kT_N} \right\}. \quad (3.7)$$

For $w = DS_z^2$, or (3.1), we have for (3.7) the following:

$$kT_N = 2J(\mathbf{Q})\frac{1}{3}S(S+1) \left\{ 1 - \frac{D}{kT_N} \frac{4S(S+1)-3}{15} \right\} \quad (D < 0). \quad (3.8)$$

As we shall see later, a similar calculation for the x or y component leads to another Néel temperature, which is, however, lower than that just calculated. Therefore, below T_N only the z -component oscillates, down to another critical temperature. In this temperature range, the oscillation of the z -component can be determined from Eq. (3.4) by putting $H_{nx} = H_{ny} = 0$. Immediately below T_N we can put

$$\sigma_{nz} = \sigma_z \cos (\mathbf{Q} \cdot \mathbf{R}_n + \alpha), \quad (3.9)$$

$$H_{nz} = 2J(\mathbf{Q})S\sigma_z \cos (\mathbf{Q} \cdot \mathbf{R}_n + \alpha). \quad (3.10)$$

More precisely, because of the presence of H_{nz}^3 and higher-order terms in (3.4), we have to include the third and higher odd harmonics in both σ_{nz} and H_{nz} . However, since with terms up to H_{nz}^3 we can determine only the amplitude of the fundamental, we neglect terms beyond H_{nz}^3 in (3.4), substitute (3.9) and (3.10) therein, and pick up only terms of the funda-

mental. Then we solve the equation to determine σ_z . Equation (3.4) can be written up to H_{nz}^3 as

$$S\sigma_{nz} = \beta H_{nz}\varphi_1(T) - \frac{1}{6}\beta^3 H_{nz}^3\varphi_3(T), \quad (3.11)$$

where

$$\varphi_1(T) = \text{tr } S_z^2 \exp[-\beta w] / \text{tr exp}[-\beta w], \quad (3.12)$$

$$\begin{aligned} \varphi_3(T) &= 3(\text{tr } S_z^2 \exp[-\beta w])^2 / (\text{tr exp}[-\beta w])^2 \\ &\quad - \text{tr } S_z^4 \exp[-\beta w] / \text{tr exp}[-\beta w]. \end{aligned} \quad (3.13)$$

Proceeding as mentioned, we obtain, using (3.5),

$$\sigma_z^2 = \frac{2}{[J(Q)S]^2} \left\{ \frac{(kT)^3}{\varphi_3(T)} \frac{d}{dT} \left(-\frac{\varphi_1(T)}{kT} \right) \right\}_{T=T_N} \cdot (T_N - T). \quad (3.14)$$

Thus, σ_z varies as $(T_N - T)^{1/2}$, as might be expected.

It can be shown that the amplitude of the third harmonic varies as $(T_N - T)^{3/2}$, that of the fifth harmonic as $(T_N - T)^{5/2}$, and so on. Eventually, if the period of the oscillation is equal to seven layers of atomic plane, as in rare-earth metals Er and Tm, the fifth harmonic is equivalent to the second harmonic and the seventh one equivalent to the zeroth one; the zeroth harmonic is the ferromagnetic component of the whole system, and, it increases with decreasing temperature as $(T_N - T)^{7/2}$.

4. UNIAXIAL ANISOTROPY ENERGY WITH AN EASY AXIS; OSCILLATION IN THE TRANSVERSE COMPONENT

We now ask about oscillations in the x or y component. For this purpose, we return to Eq. (3.3) and consider the equation to determine σ_{nx} . To first order in H_{nx} and H_{ny} , this equation can be written as^{9b}

$$\begin{aligned} S\sigma_{nx} &= \text{tr} \{ S_x \exp \beta [H_{nz}S_z - w(S_z)] \int_0^\beta \exp(-\lambda) [H_{nz}S_z - w] \\ &\quad \cdot (H_{nx}S_x + H_{ny}S_y) \exp \lambda [H_{nz}S_z - w] d\lambda \} / \text{tr exp} \beta [H_{nz}S_z - w]. \end{aligned}$$

^{9b} For noncommutative H_0 and H_1 , we have to first order in H_1

$$\exp[-\beta(H_0 + H_1)] = \exp(-\beta H_0) - \exp(-\beta H_0) \int_0^\beta \exp(\lambda H_0) H_1 \exp(-\lambda H_0) d\lambda.$$

This follows by putting $\exp[-\beta(H_0 + H_1)] = \exp(-\beta H_0)G(\beta)$, differentiating both the sides of this equation with respect to β , thus obtaining $-H_1 \exp(-\beta H_0)G(\beta) = \exp(-\beta H_0)G'(\beta)$, i.e., $-\exp(\beta H_0)H_1 \exp(-\beta H_0)G(\beta) = G'(\beta)$, and then integrating $G'(\beta)$ with the initial condition $G(0) = 1$. Thus, we have

$$\begin{aligned} G(\beta) &= 1 - \int_0^\beta \exp(\lambda H_0) H_1 \exp(-\lambda H_0) G(\lambda) d\lambda \\ &= 1 - \int_0^\beta \exp(\lambda H_0) H_1 \exp(-\lambda H_0) d\lambda + O(H_1^2). \end{aligned}$$

It can be shown that the term proportional to H_{ny} vanishes. So we have, with (3.2), linear homogeneous equations for σ_{nx} . The preceding equation can be written, after a series of elementary calculations, in the following form:

$$\begin{aligned} S\sigma_{nx} = \frac{1}{2}H_{nx} & \sum_{S_z=-S+1}^S \left\{ (S - S_z + 1)(S + S_z) \exp \beta[H_{nz}S_z - w(S_z)] \right. \\ & \cdot \frac{1 - \exp(-\beta)[H_{nz} + w(S_z - 1) - w(S_z)]}{H_{nz} + w(S_z - 1) - w(S_z)} \Big\} \\ & \cdot \left\{ \sum_{S_z=-S}^S \exp \beta[H_{nz}S_z - w(S_z)] \right\}^{-1}. \end{aligned} \quad (4.1)$$

If we assume $H_{nz} + w(S_z - 1) - w(S_z) \ll kT$ for all the S_z values, the factor in the second line reduces to β . Then, in the approximation of neglecting the third and higher orders of $\beta[H_{nz}S_z + w(S_z - 1) - w(S_z)]$, Eq. (4.1) is written as

$$\begin{aligned} S\sigma_{nx} = \frac{1}{2}\beta H_{nx} & \{\text{tr} \exp \beta[H_{nz}S_z - w(S_z)]\}^{-1} \\ & \times \text{tr} ([S(S + 1) - S_z^2] \exp \beta[H_{nz}S_z - w(S_z)]) \\ & \cdot \{1 - (\beta/4)[w(S_z + 1) + w(S_z - 1) - 2w(S_z)] \\ & + (\beta^2/12)([H_{nz} - w(S_z) + w(S_z - 1)])^2 + [H_{nz} - w(S_z + 1) \\ & + w(S_z)]^2\} - (\beta/4)S_z \exp \beta[H_{nz}S_z - w(S_z)] \\ & \cdot \{2H_{nz} - w(S_z + 1) + w(S_z - 1) - (\beta/3)([H_{nz} - w(S_z) \\ & + w(S_z - 1)])^2 - [H_{nz} - w(S_z + 1) + w(S_z)]^2\}. \end{aligned} \quad (4.2)$$

It can be seen easily that (4.1) and (4.2) are even functions of H_{nz} . H_{nz} is a periodic function of \mathbf{R}_n with wave vector \mathbf{Q} according to the preceding discussion. Hence either (4.1) or (4.2) may be expanded in a Fourier series with $2Q$ as the fundamental wave vector. In these equations, H_{nx} can be written by (3.2) as

$$H_{nx} = 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_{mx}, \quad (4.3)$$

and, in order to solve the equations, σ_{mx} and σ_{nx} may also be expanded in Fourier series with \mathbf{Q} as the fundamental wave vector.

If we retain the first two terms in the right-hand side of (4.2) and expand it in powers of H_{nz} , the zeroth-order term will become $\frac{1}{2}\beta H_{nx}$ times

$$\text{tr} [S(S + 1) - S_z^2] \exp [-\beta w(S_z)] / \text{tr} \exp [-\beta w(S_z)], \quad (4.4)$$

and the second-order term $(\frac{1}{2}\beta H_{nx})(\frac{1}{2}\beta^2 H_{nz}^2)$ times

$$\begin{aligned} \text{tr}\{\frac{1}{3}S(S+1) + [S(S+1) - \frac{4}{3}]S_z^2 - S_z^4\} \exp(-\beta w) \\ \times [\text{tr exp}(-\beta w)]^{-1} - \text{tr}\{S(S+1) - S_z^2\} \exp(-\beta w) \\ \times [\text{tr } S_z^2 \exp(-\beta w)] / [\text{tr exp}(-\beta w)]^2. \end{aligned} \quad (4.5)$$

Abbreviating $[\text{tr } S_z^2 \exp(-\beta w)] / [\text{tr exp}(-\beta w)] = \langle S_z^2 \rangle_w$ and

$$[\text{tr } S_z^4 \exp(-\beta w)] / [\text{tr exp}(-\beta w)] = \langle S_z^4 \rangle_w,$$

we can write (4.4) and (4.5) as

$$S(S+1) - \langle S_z^2 \rangle_w \quad (4.4a)$$

and

$$\frac{1}{3}S(S+1) - \frac{4}{3}\langle S_z^2 \rangle_w + (\langle S_z^2 \rangle_w)^2 - \langle S_z^4 \rangle_w. \quad (4.5a)$$

It is remarked here that (4.4a) is smaller than $2\langle S_z^2 \rangle_w$, i.e.,

$$3\langle S_z^2 \rangle_w > S(S+1)$$

and that

$$\langle S_z^4 \rangle_w > (\langle S_z^2 \rangle_w)^2,$$

so that (4.5a) is a negative quantity. The first inequality is obvious, since $w(S_z)$ is a decreasing function of S_z^2 (the z-axis is the easy axis), and the second inequality follows from

$$\begin{aligned} & [\text{tr } S_z^4 \exp(-\beta w)] [\text{tr exp}(-\beta w)] - [\text{tr } S_z^2 \exp(-\beta w)]^2 \\ &= \frac{1}{2} \sum_{S_z} \sum_{S_z'} (S_z^4 + S_z'^4) \exp(-\beta w - \beta w') - \sum_{S_z} \sum_{S_z'} S_z^2 S_z'^2 \\ &\quad \times \exp(-\beta w - \beta w') \\ &= \frac{1}{2} \sum_{S_z} \sum_{S_z'} (S_z^2 - S_z'^2)^2 \exp(-\beta w - \beta w') > 0, \end{aligned}$$

where $w = w(S_z)$ and $w' = w(S_z')$.

Using (4.3), (4.4a), and (4.5a), we can write (4.2) as

$$\begin{aligned} \sigma_{nx} &= \beta \sum_m J(\mathbf{R}_{mn}) \sigma_{mx} [\underline{S(S+1)} - \langle S_z^2 \rangle_w] \\ &\quad + \frac{1}{2}\beta^2 H_{nz}^2 [\frac{1}{3}S(S+1) - \frac{4}{3}\langle S_z^2 \rangle_w + (\langle S_z^2 \rangle_w)^2 - \langle S_z^4 \rangle_w]. \end{aligned} \quad (4.6)$$

To solve (4.6) approximately, we substitute (3.10) and expand σ_{nx} in a Fourier series:

$$\sigma_{nx} = \sum_{-\infty}^{\infty} \sigma_{\nu} \exp(i\nu \mathbf{Q} \cdot \mathbf{R}_n) \quad \text{where } \sigma_{-\nu} = \sigma_{\nu}^*. \quad (4.7)$$

Then we obtain a set of linear relations between the Fourier coefficients as

follows:

$$\begin{aligned}\beta^{-1}\sigma_\nu = J(\nu\mathbf{Q})\sigma_\nu a_0 + \frac{1}{2}J[(\nu - 2)\mathbf{Q}]\sigma_{\nu-2}a_2 \exp(2i\alpha) \\ + \frac{1}{2}J[(\nu + 2)\mathbf{Q}]\sigma_{\nu+2}a_2 \exp(-2i\alpha),\end{aligned}\quad (4.8)$$

where

$$a_0 = S(S + 1) - \langle S_z^2 \rangle_w + a_2, \quad (4.9)$$

$$a_2 = \beta^2[J(\mathbf{Q})S\sigma_z]^2[\frac{1}{3}S(S + 1) - \frac{4}{3}\langle S_z^2 \rangle_w + (\langle S_z^2 \rangle_w)^2 - \langle S_z^4 \rangle_w]. \quad (4.10)$$

The infinite determinant of the homogeneous equations (4.8) should vanish, giving eigenvalues of β^{-1} . Since it was assumed that $J(\mathbf{q})$ takes its largest value for $\mathbf{q} = \pm\mathbf{Q}$, the largest eigenvalue of $\beta^{-1}(= kT)$, which would give the second Néel temperature, is obtained mainly from the coefficients of σ_{-1} and σ_1 . Neglecting other coefficients, we have from (4.8) the following two equations:

$$\begin{aligned}kT\sigma_{-1} = J(\mathbf{Q})\sigma_{-1}a_0 + \frac{1}{2}J(\mathbf{Q})\sigma_1a_2 \exp(-2i\alpha), \\ kT\sigma_1 = J(\mathbf{Q})\sigma_1a_0 + \frac{1}{2}J(\mathbf{Q})\sigma_{-1}a_2 \exp(2i\alpha).\end{aligned}\quad (4.11)$$

Thus,

$$\begin{vmatrix} J(\mathbf{Q})a_0 - kT & \frac{1}{2}J(\mathbf{Q})a_2 \exp(-2i\alpha) \\ \frac{1}{2}J(\mathbf{Q})a_2 \exp(2i\alpha) & J(\mathbf{Q})a_0 - kT \end{vmatrix} = 0,$$

$$kT = J(\mathbf{Q})(a_0 \pm \frac{1}{2}a_2).$$

Since a_2 is a negative quantity as mentioned before, we have to take the lower sign, and this determines the second Néel temperature, T_N' , where the x -component of the spins starts to oscillate. Thus, the equation to determine T_N' is

$$kT_N' = J(\mathbf{Q})(a_0 - \frac{1}{2}a_2), \quad (4.12)$$

where a_0 and a_2 are defined by (4.9) and (4.10), the temperature at which these are evaluated being T_N' . The main term in the right-hand side of (4.12) is $J(\mathbf{Q})[S(S + 1) - \langle S_z^2 \rangle_w]$, and when we compare this with the right-hand side of Eq. (3.5), which is $2J(\mathbf{Q})\langle S_z^2 \rangle_w$, we see that T_N' is lower than T_N .

Furthermore, from (4.11) we have the ratio of σ_1 and σ_{-1} to be $-\exp(i\alpha)$ to $\exp(-i\alpha)$, which means that σ_{nz} is proportional to $\sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$. In other words, ordering in the x -component of the spins sets in below T_N' in regions where the z -component is disordered, as the latter is given by (3.9) and is proportional to $\cos(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$.

If we neglect a_2 completely, we have for the second Néel temperature
 $kT_{N'} = J(\mathbf{Q})\{\frac{2}{3}S(S+1) + \text{tr}[S_z^2 - \frac{1}{3}S(S+1)]w(S_z)/(2S+1)kT_N'\}.$

(4.13)

In the case of $w = D[S_z^2 - \frac{1}{3}S(S+1)]$, this becomes

$$kT_{N'} = 2J(\mathbf{Q})\frac{1}{3}S(S+1) \left\{ 1 + \frac{D}{kT_N'} \frac{4S(S+1) - 3}{30} \right\}. \quad (4.14)$$

Comparing (4.14) with (3.8), we see that $T_{N'}$ is lower than T_N , since D is negative. The neglected terms will further lower $T_{N'}$, and in the case of a large anisotropy energy the second Néel temperature might not exist.

Examples of the theoretical results of Sections 3 and 4 are found in rare-earth metals Er and Tm. Neutron diffraction experiments have shown that in Er below 85°K and Tm below 56°K a longitudinal sinusoidal spin ordering sets in along the hexagonal axis (the crystal being hcp) with a period of seven hexagonal layers. In Er, the perpendicular component begins to oscillate at 53°K and, furthermore, below 20°K the spin structure becomes conical. In Tm, the perpendicular component never starts to oscillate, presumably because of a large anisotropy energy, and with decreasing temperature the spin structure tends to a repetition of (+ + + + - - -), four layers of up-spins and three layers of down-spins. Thus, in Tm the phase constant α , which was arbitrary in the foregoing arguments, becomes fixed in such a way that nodal planes do not coincide with atomic planes. According to experiment, a ferromagnetic component is perceptible below 40°K. The integral period of seven layers is evidently favorable for such to occur, and conversely the spin system seems to be stabilized by assuming an integral period and adjusting α so as to lower the energy.¹⁰ More discussions of rare-earth metals will be given in the forthcoming Part 2.

5. ANISOTROPY ENERGY OF TWOFOLD, FOURFOLD, AND SIXFOLD SYMMETRY WITH AN EASY PLANE

We consider an anisotropy energy of p -fold symmetry ($p = 2, 4, 6$) about the z axis. This anisotropy energy is assumed to make the xy plane the plane of easy magnetization. The simplest form of such an anisotropy Hamiltonian may be written as

$$w(S_z) + \text{const} \cdot [(S_x + iS_y)^p + (S_x - iS_y)^p], \quad (5.1)$$

where $w(S_z)$ is assumed to be the smallest for the smallest value of S_z^2 . The constant in the second term should be small. This second term vanishes

¹⁰ T. Nishikubo and T. Nagamiya, *J. Phys. Soc. Japan* **20**, 808 (1965).

for $S < p/2$. In the case of $p = 2$, this term reduces to $\text{const} \cdot (S_x^2 - S_y^2)$, and in this case we can expect results similar to those that we have found in the preceding two sections, namely, below a certain Néel temperature, T_N , only the y component oscillates (if the const is positive), and below another Néel temperature, $T_{N'}$, the x -component comes into oscillation, the resulting elliptic oscillation being confined to the xy plane. Similar calculations should be possible for this case. Thus, we shall henceforth be concerned only with the cases of $p = 4$ and $p = 6$.

In the case of $p = 4$ or 6 , a helical ordering in the xy plane appears below the Néel temperature if the second term of (5.1) is small. This can be seen by expanding the exponential factor in (3.3), $w(S_z)$ being replaced by (5.1), in powers of exchange field components and retaining the constant and linear terms, and then expanding these terms in powers of $(S_x \pm iS_y)^p$ and retaining the constant and linear terms; since $(S_x \pm iS_y)^p$ are operators to change S_z to $S_z \pm p$, and since S_x and S_y appear at most twice after the symbol tr, no contribution appears from the latter linear term when $p > 2$. We are therefore led to Eq. (4.1) or (4.2), with $H_{nz} = 0$. Thus, the Néel temperature is determined by the procedure that followed these equations. Assuming

$$kT_N = J(\mathbf{Q}) \frac{\text{tr} [S(S+1) - S_z^2] \exp [-w(S_z)/kT_N]}{\text{tr} \exp [-w(S_z)/kT_N]}, \quad (5.2)$$

we obtain

$$w(S_z) - w(S_z - 1) \ll kT_N,$$

it would seem that either the x or y component, or any one component in the xy plane, comes into oscillation below T_N , but higher-order terms in the expansion in powers of $H_{nx}S_x + H_{ny}S_y$ will couple the x and y components to give a helical ordering, i.e., if the x component has started to oscillate as $\cos(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$, then the y component must at the same time have started to oscillate as $\pm \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$ to ensure the minimum free energy.

Thus, below T_N we have helical ordering in which the thermal average spin vectors rotate in the xy plane. With decreasing temperature, however, the anisotropy energy of fourfold or sixfold symmetry comes into play. If the *turn angle* of the helix between successive atomic xy layers, which is \mathbf{Q} times the interlayer distance, is close to 60° in the case of $p = 6$, we may expect trapping of the spin vectors in the successive six potential valleys. If the turn angle is close to 30° , trapping of each pair of spin vectors in the successive six valleys may occur, as actually observed in the case of Ho below 20°K (although the structure is conical). For smaller turn angles, trapping of all spin vectors in a single valley is a possibility; this is the transition from the helical state to the ferromagnetic state observed in Tb and Dy. We shall discuss this ferromagnetic transition in some detail in the following.

We shall first consider the situation at absolute zero or at very low temperatures. In the helical state, the anisotropy energy within the easy plane may modify the spin arrangement in such a way that the $(p - 1)$ th and $(p + 1)$ th harmonics are mixed to the fundamental, but this will only slightly change the energy, so that we shall simply take the energy to be $-S^2J(\mathbf{Q})$ per atom [see (1.12); $2J(\mathbf{Q})S$ is the magnitude of the exchange field, and the energy per atom is minus half the product of this with the spin length S]. In the ferromagnetic state, the energy due to the exchange interaction is $-S^2J(0)$, which is higher than $-S^2J(\mathbf{Q})$, but we have an additional negative energy due to the minimum of the anisotropy energy, which we shall denote by $w_{\min} < 0$. Therefore, the ferromagnetic state is more stable if

$$S^2[J(\mathbf{Q}) - J(0)] < |w_{\min}|. \quad (5.3)$$

We now ask if the ferromagnetic state is really stable under this condition; i.e., we ask if all the ferromagnetic spin-wave frequencies are positive. For this purpose, we shall assume an out-of-plane anisotropy energy of the form of DS_z^2 with positive D , confining ourselves to small spin deviations from the xy plane, and an in-plane anisotropy energy of the form of GS_y^2 with positive G , considering only small spin deviations from the equilibrium x direction. These energies would give anisotropy fields $-2DS_z$ in the z direction and $-2GS_y$ in the y direction. If one assumes the in-plane anisotropy energy to be a classical function of azimuth φ and to be simply proportional to $\cos(p\varphi)$, namely,

$$w(\varphi) = w_{\min} \cos(p\varphi), \quad (5.4)$$

then

$$G = (p^2/2S^2) |w_{\min}|, \quad (5.5)$$

since $S_\varphi = S_y$.

Now, the effective field acting on the n th spin can be written in components as

$$\begin{aligned} H_{nx} &= 2J(0)S + H, & H_{ny} &= 2 \sum_m J(\mathbf{R}_{mn}) S_{my} - 2GS_{ny}, \\ H_{nz} &= 2 \sum_m J(\mathbf{R}_{mn}) S_{mz} - 2DS_{nz}, \end{aligned} \quad (5.6)$$

where we have included an external field H in the x -component. The equations of motion for the spins are

$$\hbar \dot{S}_{ny} = S_{nx}H_{nx} - S_{nx}H_{nz}, \quad \hbar \dot{S}_{nz} = S_{nx}H_{ny} - S_{ny}H_{nx}.$$

Substituting here (5.6) and putting $S_{nx} = S$, $S_{ny} = b \exp(i\mathbf{q} \cdot \mathbf{R}_n - i\omega t)$, $S_{nz} = c \exp(i\mathbf{q} \cdot \mathbf{R}_n - i\omega t)$, where b and c are constants, we obtain im-

mediately the frequency formula:

$$(\hbar\omega/2S)^2 = [J(0) + (H/2S) - J(\mathbf{q}) + D][J(0) + (H/2S) - J(\mathbf{q}) + G]. \quad (5.7)$$

We assume that D is so large that the first factor in the right-hand side of (5.7) is positive for any \mathbf{q} . Then, the condition for stability is

$$J(0) + (H/2S) - J(\mathbf{q}) + G > 0 \quad \text{for all } \mathbf{q}.$$

With the use of (5.5), and remembering that $J(Q)$ is the maximum of $J(\mathbf{q})$, this turns out to be

$$S^2[J(Q) - J(0)] < \frac{1}{2}p^2 |w_{\min}| + \frac{1}{2}SH. \quad (5.8)$$

This inequality is surely satisfied when (5.3) holds and $p > 2$.

We are thus assured of the stability of the ferromagnetic state. If we now consider w_{\min} to be a function of temperature, we see that a transition occurs from the helical state to the ferromagnetic state at the temperature at which the left and right sides of (5.3) become equal. An external field of course enhances this transition, in which case we have an extra positive term $SH - \frac{1}{2}\chi_h H^2$ in the right-hand side of (5.3), where χ_h is the susceptibility per atom of the helical state to be discussed in Part IV, Section 7. However, the inequality with this additional term in the right-hand side does not always assure the inequality (5.8), so that the ferromagnetic state may not be stable. In fact, as we shall discuss in the next part, there is the interesting possibility of a *fan structure* in the presence of a magnetic field.

In Dy, the Néel temperature is 179°K, whereas the temperature of transition to the ferromagnetic state is 85°K, where the turn angle of the helix is 26.5°. The foregoing consideration may be applicable to this case. In Tb, the Néel temperature is 228°K, and only 7° below this the transition to the ferromagnetic state occurs; at the latter temperature, the turn angle of the helix is 18°. For such a case, we have to make a high-temperature approximation, as we shall do below. In both Dy and Tb, the lowering of the (free) energy of the ferromagnetic state due to a magnetostrictive effect may play a considerable role, as was pointed out by Enz.¹¹

We shall briefly discuss the ferromagnetic transition at a high temperature. For this purpose, we compare the free energies of the two states. In the approximation of the Weiss field, with a negligible anisotropy energy, the free energy of the helical state can be expressed as

$$F_h = -kT \ln \operatorname{tr} \exp (\beta H_h S_x) + \frac{1}{2}H_h S \sigma_h, \quad (5.9)$$

¹¹ U. Enz, *Physica* **26**, 698 (1960).

with

$$S\sigma_h = [d/d(\beta H_h)] \ln \text{tr} \exp (\beta H_h S_z), \quad (5.10)$$

where H_h means the magnitude of the internal field in this state. The free energy, F_f , of the ferromagnetic state can be expressed with H_f and σ_f in place of H_h and σ_h , with an additional anisotropy energy term w_{\min} of the given temperature. This w_{\min} is the minimum value of the p -fold anisotropy Hamiltonian averaged over the Boltzmann distribution of the spins in the ferromagnetic Weiss field corresponding to an arbitrary direction of magnetization.

We expand these free energy expressions in powers of H_h or H_f . To the lowest power, we have

$$F_h = -\beta^{-1} \ln (2S + 1) - \frac{1}{180} \beta^3 H_h^4 S(S + 1)(S^2 + S + \frac{1}{2}) \quad (5.11)$$

and a similar expression for F_f with an additional term w_{\min} . On the other hand, assuming a Brillouin function, we have

$$S\sigma = \frac{1}{3}\beta HS(S + 1) - \frac{1}{45}(\beta H)^3 S(S + 1)(S^2 + S + \frac{1}{2}) + \dots \quad (5.12)$$

for $\sigma = \sigma_h$ and $H = H_h$ and also for $\sigma = \sigma_f$ and $H = H_f$. Now,

$$H_h = 2J(Q)S\sigma_h, \quad H_f = 2J(0)S\sigma_f, \quad (5.13)$$

$$kT_N = \frac{2}{3}J(Q)S(S + 1), \quad kT_C = \frac{2}{3}J(0)S(S + 1). \quad (5.14)$$

Hence, using (5.12), we can express H_h as a function of T and T_N and H_f as a function of T and T_C . A calculation gives the results:

$$H_h^2 = 15(kT)^2(1 - T/T_N)(S^2 + S + \frac{1}{2})^{-1} \quad (T < T_N),$$

$$H_f^2 = 15(kT)^2(1 - T/T_C)(S^2 + S + \frac{1}{2})^{-1} \quad (T < T_C).$$

Thus, in order that $F_h > F_f$, the following inequality must hold for $T < T_C$ and $T < T_N$:

$$\frac{5}{2} \frac{S(S + 1)}{S^2 + S + \frac{1}{2}} kT^2(T_C^{-1} - T_N^{-1})[1 - \frac{1}{2}T(T_C^{-1} + T_N^{-1})] < |w_{\min}|. \quad (5.15)$$

Since T_C is lower than T_N according to (5.14), the left-hand side of (5.15) is positive for $T < T_C$ and has a maximum at

$$T = 4T_N T_C / 3(T_N + T_C).$$

This maximum is small when T_C is close to T_N . On the other hand, $|w_{\min}|$ vanishes at T_N and increases with decreasing temperature. If this increase is rapid, the inequality (5.15) may become satisfied, or the ferromagnetic state may become more stable, at temperatures that are below the critical

temperature determined by the equality of the two sides of (5.15). It is emphasized that T_C must be close to T_N in order that the ferromagnetic transition of this kind is predicted. In other words, $J(0)$ must be close to $J(\mathbf{Q})$, according to (5.14). Magnetostrictive energy in the ferromagnetic state may be included in w_{\min} (cf. Note added in proof on page 403).

6. ANISOTROPY ENERGY HAVING AN EASY CONE

Neutron diffraction experiments have shown that in the rare-earth metals Ho and Er the spin structures below 20°K are such that the spins in each hexagonal basal plane are ferromagnetically ordered, and they precess on a circular cone in going from one plane to the next. The cone axis is the c -axis, and the vertex angle of the cone is about 75° for Ho and 25° for Er. The turn angle in the projection of the spins on the c -plane is 30° and 44°, respectively. In Ho, speaking in more detail, the turn angle deviates from 30° alternatingly by 2δ and -2δ , with $\delta = 9^\circ$ at 4.2°K, because of the anisotropy energy of sixfold symmetry.¹² (For references, see the forthcoming Part 2.)

We shall discuss briefly in this section the transition to such a conical structure due to a gradual change in the anisotropy energy with lowering temperature. Confining ourselves to low temperatures, we may assume that each atomic plane perpendicular to the vector \mathbf{Q} (the hexagonal basal plane in the case of rare-earth metals) has a ferromagnetic saturation moment equal to that at absolute zero, and the anisotropy energy is a classical function of the direction of this moment and of temperature. Such an anisotropy energy for each atom may be written as

$$w(\cos^2 \theta, \sin^p \theta \cos p\varphi; T), \quad (6.1)$$

where θ and φ are the polar and azimuthal angles of the atomic moment under consideration, and $p = 6$ in the case of rare-earth metals.

If w is a small quantity, we may have a helical arrangement whose rotation plane is determined by the minimum of w averaged over the orientations of the moments in that arrangement. Let us consider the simplest case that w is independent of φ and has a minimum at $\theta = \theta_0$ (so that it has also the same minimum at $\theta = \pi - \theta_0$). The polar angle of the normal to the rotation plane of the helix will be denoted by Θ ; the azimuth should be indeterminate in this case. Since $\cos \theta$ for the moment of the atom at \mathbf{R}_n is $\sin \Theta \cos (\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$ and the angle $\mathbf{Q} \cdot \mathbf{R}_n + \alpha$ is distributed uniformly in the angular interval of 2π , provided that the period of the helix is not a simple rational multiple of the lattice spacing along \mathbf{Q} , we have for

¹² W. C. Koehler, J. W. Cable, M. K. Wilkinson, and E. O. Wollan, *Phys. Rev.* **151**, 414 (1966).

the average of the anisotropy energy

$$\langle w(\sin^2 \Theta \cos^2 (\mathbf{Q} \cdot \mathbf{R}_n + \alpha); T) \rangle_n = \frac{1}{2\pi} \int_0^{2\pi} w(\sin^2 \Theta \cos^2 \varphi; T) d\varphi. \quad (6.2)$$

The minimum of this integral, with respect to Θ , determines Θ . It is easy to see that the rotation plane of the helix determined in this way cuts the easy cone determined by $w(\cos^2 \theta) = \min$, with $\theta = \theta_0$, the plane being closer to the polar axis. Eventually, if $\theta_0 = 0$, the plane will contain the polar axis, and, if $\theta_0 = \pi/2$, it will be perpendicular to the polar axis.

On the other hand, when the minimum of w at θ_0 is deep enough, the structure should be a conical one. The vertex angle of this cone must be somewhat greater than θ_0 because of the exchange interaction. This vertex angle will be denoted by θ_1 . Assuming again the independence (or a weak dependence) of w on φ , we can determine θ_1 by minimizing the exchange plus anisotropy energy, which can be written as

$$-S^2 \sin^2 \theta_1 J(\mathbf{Q}) - S^2 \cos^2 \theta_1 J(0) + w(\cos^2 \theta_1; T), \quad (6.3)$$

where $S \sin \theta_1$ is the rotating spin component and $S \cos \theta_1$ the parallel, or ferromagnetic, component. The minimization of this with respect to θ_1 gives

$$S^2 [J(\mathbf{Q}) - J(0)] + [d/d(\cos^2 \theta_1)] w(\cos^2 \theta_1; T) = 0. \quad (6.4)$$

The first term is positive, so that the second term must be negative, which means that $\cos^2 \theta_1$ must be smaller than $\cos^2 \theta_0$. We cannot go further without knowing the functional form of w .

The structural change in Ho from a proper helix to a cone may be due to a deepening of the minimum of w at θ_0 with lowering temperature. θ_0 should be smaller than 75° ($=\theta_1$). Since the observed structure change is an abrupt one¹² and since the rotation plane of the helical structure above 20°K remains perpendicular to the c -axis, there being no helix with an oblique normal as that predicted with (6.2), one must imagine that w has another well-defined minimum at $\theta = \pi/2$ and that the minimum at θ_0 gets deeper than this as the temperature is lowered. Furthermore, as mentioned before, the turn angle in the conical structure is alternately smaller and greater than 30° , whereas the turn angle just above 20°K is a little greater than 30° . The fitting of each pair of moments successively into potential valleys of the anisotropy energy must give a further stabilization of the conical structure below 20°K .

In Er, the structure above 20°K is such that components both parallel and perpendicular to the c axis oscillate with a common wave number. This wave number joins smoothly to that of the conical structure below

20°K. A longitudinal sinusoidal structure at high temperatures first changes, with decreasing temperature, to an elliptic structure at T_N' discussed in Section 4, and then the plane of the ellipse gradually deviates from the c axis because the minimum of w moves from $\theta = 0$ to a nonzero θ value, and, finally, as the minimum deepens, there occurs an abrupt change into a conical structure. This theoretical interpretation is consistent with all observations on Er.

We shall mention the principle of calculating the energy of the structure in Ho below 20°K. The relative azimuthal angle between the moment of the atom at \mathbf{R}_n and the moment of the atom at the origin can be written as $\mathbf{Q}_{30} \cdot \mathbf{R}_n - \delta + \delta \cos(6\mathbf{Q}_{30} \cdot \mathbf{R}_n)$, where \mathbf{Q}_{30} is the wave vector representing the helical structure with a turn angle of 30°. To calculate the exchange energy for the rotating components, we have to take the cosine of this angle, multiply it by $-S^2 \sin^2 \theta_1 J(\mathbf{R}_n)$, and sum it over \mathbf{R}_n . The cosine of the angle can be expanded into a Fourier series with coefficients expressed in terms of Bessel functions. The Fourier components consist of the fundamental having \mathbf{Q}_{30} , the fifth, seventh, ..., harmonics, and, in general, $(6 \times \text{integer } \pm 1)$ th harmonics. Correspondingly, the exchange energy can be expressed by $J(\mathbf{Q}_{30})$, $J(5\mathbf{Q}_{30})$, $J(7\mathbf{Q}_{30})$, etc., and by Bessel functions of the zeroth, first, etc., degrees. The result should replace the first term of (6.3). In the anisotropy energy, given by (6.1), θ is replaced by θ_1 and $\cos 6\varphi$ by $\sin 6\delta$. However, actual calculations are not yet meaningful, because we do not know the exact form of w for Ho. On the other hand, the mentioned expansion of the angle into a Fourier series is useful in analyzing neutron diffraction data.

IV. Effect of External Field on Spin Configurations

A weak external field applied to a helically ordered spin system induces a magnetization that is proportional to the field strength. At high fields, however, structure changes appear, some of which have no analogs in ferromagnetism and antiferromagnetism. Experiments to show these structure changes are still few, and in this section we shall be concerned mostly with the theory.^{11,13-16}

¹³ A. Herpin, P. Mériel, and J. Villain, *Compt. Rend.* **249**, 1334 (1959); A. Herpin and P. Mériel, *ibid.* **250**, 1450 (1960); *J. Phys. Radium* **22**, 337 (1961).

¹⁴ T. Nagamiya, K. Nagata, and Y. Kitano, *Progr. Theoret. Phys. (Kyoto)* **27**, 1253 (1962).

¹⁵ Y. Kitano and T. Nagamiya, *Progr. Theoret. Phys. (Kyoto)* **31**, 1 (1964).

¹⁶ H. Thomas and P. Wolf, *Proc. Intern. Conf. Magnetism, Nottingham, 1964*, p. 731. Inst. Phys. Phys. Soc., London, 1965.

7. FIELD APPLIED PERPENDICULAR TO THE PLANE OF SPIN ROTATION

We shall first neglect the anisotropy energy and consider a field applied perpendicular to the plane in which the spin vectors rotate.

The exchange field acting on each spin in the absence of external magnetic field has a magnitude of $2J(\mathbf{Q}) |\langle \mathbf{S} \rangle|$, the factor $-(g\mu_B)^{-1}$ being here again omitted for convenience. If a perpendicular external field of magnitude H is superposed on this, one will have a conical structure with the thermal average of any one spin having certain common components $\langle S \rangle_{||}$ and $\langle S \rangle_{\perp}$ parallel and perpendicular to the field direction. The effective field on each spin will have the components

$$2J(0)\langle S \rangle_{||} + H \quad \text{parallel to the field,}$$

$$2J(\mathbf{Q})\langle S \rangle_{\perp} \quad \text{perpendicular to the field.}$$

Then, one should have

$$\frac{\langle S \rangle_{||}}{\langle S \rangle_{\perp}} = \frac{2J(0)\langle S \rangle_{||} + H}{2J(\mathbf{Q})\langle S \rangle_{\perp}}. \quad (7.1)$$

Thus, the effective field is $2J(\mathbf{Q})\langle \mathbf{S} \rangle$, where $\langle \mathbf{S} \rangle$ is the thermal average of each spin vector. The magnitude of $\langle \mathbf{S} \rangle$ is determined by the equation

$$|\langle \mathbf{S} \rangle| = SB_S(x), \quad x = 2J(\mathbf{Q}) |\langle \mathbf{S} \rangle| S/kT. \quad (7.2)$$

One sees that $|\langle \mathbf{S} \rangle|$ is unaffected by the field. This is, however, valid only when the field is so weak that the spins are not parallel to the field direction. At the critical field, H_0 , at which the spins come to point all parallel to the field direction, one has $\langle S \rangle_{||} = |\langle \mathbf{S} \rangle|$. From (7.1) follows

$$H = 2\langle S \rangle_{||}[J(\mathbf{Q}) - J(0)]. \quad (7.3)$$

Thus,

$$H_0 = 2|\langle \mathbf{S} \rangle|[J(\mathbf{Q}) - J(0)]. \quad (7.4)$$

(The right-hand sides of these equations is divided by $g\mu_B$ in the original notation.) Furthermore, from (7.3), one obtains the susceptibility perpendicular to the plane of spin rotation, which we shall denote by χ_z , namely,

$$\chi_z = \langle S \rangle_{||}/H = \frac{1}{2}[J(\mathbf{Q}) - J(0)]^{-1} \quad (\text{per atom}). \quad (7.5)$$

[In the original notation, the right-hand side of (7.5) acquires a factor $(g\mu_B)^2$.]

The foregoing has a close analog in the two-sublattice antiferromagnetism, χ_z corresponding to the perpendicular susceptibility, χ_{\perp} , of the latter. In actuality, the presence of an anisotropy energy modifies expression (7.5) to some extent.

8. FIELD APPLIED IN THE PLANE OF SPIN ROTATION

When the external field is in the plane in which the spin vectors rotate, we encounter a more complicated situation. The theoretical treatment of this case was first made by Yoshimori,¹ then by Herpin and Mériel,¹³ who discovered the fan structure to be discussed below, by Enz,¹¹ and by Nagamiya *et al.*,¹⁴ and later most generally by Kitano and Nagamiya.¹⁵ The plane of spin rotation will be taken as the xy plane, and the field is supposed to be in the x direction. At no field, the spin at \mathbf{R}_n has components

$$\begin{aligned}\langle S_{nx} \rangle &= S\sigma_{nx} = S\sigma \cos (\mathbf{Q} \cdot \mathbf{R}_n + \alpha), \\ \langle S_{ny} \rangle &= S\sigma_{ny} = S\sigma \sin (\mathbf{Q} \cdot \mathbf{R}_n + \alpha).\end{aligned}\quad (8.1)$$

In the presence of a field, \mathbf{H} , these will change to^{16a}

$$\begin{aligned}S\sigma_{nx} &= S[\sigma_{x0} + \sigma_{x1} \cos (\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \sigma_{x2} \cos 2(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots], \\ S\sigma_{ny} &= S[\sigma_{y1} \sin (\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \sigma_{y2} \sin 2(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots],\end{aligned}\quad (8.2)$$

σ_{nx} being an even periodic function of the angle $\mathbf{Q} \cdot \mathbf{R}_n + \alpha$ and σ_{ny} an odd periodic function. Correspondingly, the effective field acting on the n th spin will become

$$\begin{aligned}H_{nx} &= H + 2J(0)S\sigma_{x0} + 2J(Q)S\sigma_{x1} \cos (\mathbf{Q} \cdot \mathbf{R}_n + \alpha) \\ &\quad + 2J(2Q)S\sigma_{x2} \cos 2(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots,\end{aligned}\quad (8.3)$$

$$H_{ny} = 2J(Q)S\sigma_{y1} \sin (\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + 2J(2Q)S\sigma_{y2} \sin 2(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots.$$

If we write

$$(\sigma_{nx}^2 + \sigma_{ny}^2)^{1/2} = \sigma_n, \quad (H_{nx}^2 + H_{ny}^2)^{1/2} = H_n, \quad (8.4)$$

then σ_n will be given by the Brillouin function of SH_n/kT , and, conversely, H_n will be given by the inverse Brillouin function of σ_n , multiplied by kT/S . Even in the case where we have an anisotropy energy, the magnitude of the effective field (exchange plus external field), H_n , is given by a certain function of σ_n and T similar to that for the case of no anisotropy energy. We shall write it as $H^*(\sigma_n, T)$. Then we have, in components, the following equations to determine σ_{nx} and σ_{ny} :

$$H_{nx} = (\sigma_{nx}/\sigma_n)H^*(\sigma_n, T), \quad H_{ny} = (\sigma_{ny}/\sigma_n)H^*(\sigma_n, T), \quad (8.5)$$

where σ_{nx} and σ_{ny} are given by (8.2) and by (8.3).

If the external field H is small, σ_n will not be much different from σ ; at low temperatures, σ_n will be practically equal to σ for any strength of the external field; at high temperatures, both σ_n and σ will be small, so

^{16a} There will be a small change in the \mathbf{Q} value when there is a field, which we shall neglect.

far as H is not extremely high. In all these cases, we can expand $H^*(\sigma_n, T)$ in powers of $\sigma_n - \sigma$ and can confine ourselves to terms of lower powers. Also, it will be shown later that σ_n coincides with σ at a critical field, H_0 , where transition occurs from the *fan structure* to the *ferromagnetic structure*, so that in the neighborhood of this critical field we can make the same treatment. Since by reversing the direction of the effective field the magnetization also reverses its sign, $H^*(\sigma_n, T)/\sigma_n$ is an even function of σ_n , or a function of σ_n^2 . Thus, we can make the following expansion:

$$\frac{H^*(\sigma_n, T)}{\sigma_n} = \frac{H^*(\sigma, T)}{\sigma} + \frac{\partial}{\partial(\sigma^2)} \left[\frac{H^*(\sigma, T)}{\sigma} \right] \cdot (\sigma_n^2 - \sigma^2) + \dots \quad (8.6)$$

The first term of this expansion is equal to the exchange field at no external field, divided by σ , so that it can be identified with $2J(\mathbf{Q})S$. With two terms of (8.6), we can therefore write Eqs. (8.5) as

$$H_{ni} = \sigma_{ni} \{ 2J(\mathbf{Q})S + [\partial/\partial(\sigma^2)] [H^*(\sigma, T)/\sigma] \cdot (\sigma_n^2 - \sigma^2) \}, \quad (8.7)$$

where $i = x, y$.

We substitute (8.2) and (8.3) into (8.7) and compare the Fourier coefficients of both sides. Then we obtain equations to determine σ_{x0} , σ_{z1} , σ_{y1} , etc. Leaving the details of the calculation to Kitano and Nagamiya,^{15,16} we mention here and in the next section the results only. In the Appendix, we shall develop another mathematical method and derive susceptibility formulas.

For weak field, we obtain the susceptibility formula for the helical state for a field in the plane:

$$\chi_h = \frac{S\sigma_{x0}}{H} = \frac{2\beta_2 + \gamma}{2\beta_2 + (1 + \beta_2)\gamma} \chi_z, \quad (8.8)$$

where

$$\beta_2 = [J(\mathbf{Q}) - J(2\mathbf{Q})]/[J(\mathbf{Q}) - J(0)], \quad (8.9)$$

$$\gamma = \frac{\sigma^2}{S[J(\mathbf{Q}) - J(0)]} \frac{\partial}{\partial(\sigma^2)} \frac{H^*(\sigma, T)}{\sigma}. \quad (8.10)$$

In the present case, where we have no anisotropy energy, $H^*(\sigma, T)$ is $(kT/S)B_S^{-1}(\sigma)$, as mentioned before. In this case, γ can be expressed, near the Néel temperature, as

$$\gamma \approx \frac{2J(\mathbf{Q})}{J(\mathbf{Q}) - J(0)} \frac{T_N - T}{T_N}. \quad (8.11)$$

χ_h and χ_z thus coincide with each other at T_N . Below T_N , χ_h is smaller¹⁶ $J(\mathbf{q})$ and \mathbf{q}_0 in Kitano and Nagamiya¹⁵ correspond to our $2S^2J(\mathbf{q})$ and \mathbf{Q} , respectively.

than χ_z according to (8.8) and (8.11). For $T \rightarrow 0$, γ tends to infinity, and one has $\chi_h = \chi_z/(1 + \beta_2)$.

Because χ_h is smaller than χ_z below T_N , a field applied in the plane of the spin rotation will make the spins flip in such a way that the rotation plane become perpendicular to the field direction, provided there is no anisotropy energy to keep the spins in the original plane. If this anisotropy energy is not zero but small, the flip will occur at a critical field, H_c , which one obtains by equating $\frac{1}{2}(\chi_z - \chi_h)H_c^2$ to the difference between the anisotropy energy values for the two configurations. This is analogous to the well-known spin-flip phenomenon in antiferromagnetism.

When we are at temperatures sufficiently close to T_N , the series (8.6) is well convergent, since both σ_n^2 and σ^2 are small, and the coefficients of the expansion are finite. It can be shown also that the Fourier expansions (8.2) and (8.3) converge well in this case. The calculation of the Fourier coefficients can be carried out completely after neglecting the second and higher harmonics, and the result is the following.¹⁵

At zero field, the hodograph described by the spin vectors is a circle of radius σ . A weak field applied in this plane changes this circle into an ellipse whose minor axis along the field direction has a semiaxial length of $\sigma_{z1} = (\sigma^2 - 4\sigma_0^2)^{1/2}$ (σ_{z0} being abbreviated as σ_0), according to the calculation, and whose major axis perpendicular to the field direction has the constant semiaxial length σ . σ_0 increases with increasing field strength, first linearly, corresponding to the susceptibility χ_h , and then more rapidly. When σ_0 reaches $\frac{1}{2}\sigma$, σ_{z1} vanishes, and the magnetization curve then follows another branch. In the latter region, we have a *fan structure*, in which only

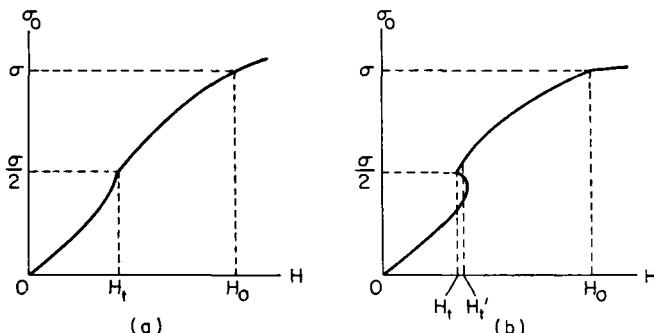


FIG. 1. Magnetization curve of a helical spin system for a field in the plane that contains the spin vectors. (a) At a temperature close to the Néel temperature ($\gamma < 8/11$); helical below H_t , fan between H_t and H_0 , ferromagnetic above H_0 . (b) At a low temperature ($\gamma > 8/11$); helical below H_t' , fan between H_t' and H_0 , ferromagnetic above H_0 . H_0 is proportional to the thermal average spin length for zero field.

the y component of the spin vectors oscillates, whereas the x component is a constant, so that the hodograph described by the spin vectors is a line. The x component still increases with increasing field strength, whereas the y component diminishes as $\sigma_{y1} = [\frac{4}{3}(\sigma - \sigma_0^2)]^{1/2}$, until σ_0 reaches σ . Above this field, which is equal to H_0 given by (7.4), the spins all align parallel to the field, and their magnitude increases with increasing field as in an ordinary ferromagnet.

The transition from the helical structure to the fan structure at $\sigma_0 = \frac{1}{2}\sigma$ occurs at a field H_t given by

$$H_t = \left(\frac{1}{2} - \frac{\gamma}{16} \right) H_0. \quad (8.12)$$

The magnetization curve looks like Fig. 1a or b, according to whether γ is smaller or larger than $8/11$. In the latter case, the actual transition from helix to fan should take place at a field H_t' at which the vertical line in Fig. 1b divides the triple-valued part of the magnetization curve into equal enclosed areas. The transition is, therefore, of the first kind.

We shall discuss these situations in more detail with different mathematics in the next section.

9. STRUCTURE CHANGES IN THE PLANE (NO IN-PLANE ANISOTROPY, ARBITRARY TEMPERATURE)

We would like to recall Eq. (5.7) of the preceding part for the spin-wave frequency of the ferromagnetic state in which the moments are all aligned parallel in one of the easy directions in the easy plane. In that equation, D and G were so defined that DS_z^2 is the out-of-plane anisotropy energy and GS_y^2 the in-plane anisotropy energy, both for small deviations from the easy direction, x . When D is large enough, positive frequencies are obtained only when the field strength exceeds a value H_0 defined by

$$H_0 = 2S[J(Q) - J(0) - G]. \quad (9.1)$$

We see that here D does not enter into H_0 . When $G = 0$, (9.1) is nothing but Eq. (7.4) for absolute zero. Below H_0 , defined by (9.1), the ferromagnetic state is unstable, and we may have a fan structure. We might also infer that for an arbitrary temperature and no anisotropy within the plane we have ferromagnetic structure above H_0 , defined by (7.4), and fan structure below H_0 . In fact, this inference can be verified in the following way in the approximation of the molecular field.

We once again consider Eq. (3.3). There $w(S_z)$ is the anisotropy energy that makes the xy plane the easy plane. H_{nx} , H_{ny} , H_{nz} are the components

of the effective field (exchange plus external field in the present case) acting on the n th spin. They are given by

$$\begin{aligned} H_{nx} &= 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_{mx} + H, \\ H_{ny} &= 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_{my}, \\ H_{nz} &= 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_{mz}. \end{aligned} \quad (9.2)$$

We consider that σ_{mx} , as a function of the position \mathbf{R}_m , fluctuates about a constant value σ_0 that satisfies the equation

$$S\sigma_0 = \frac{\text{tr } S_x \exp \beta[H^*S_x - w(S_z)]}{\text{tr } \exp \beta[H^*S_x - w(S_z)]}, \quad (9.3)$$

where

$$\begin{aligned} H^* &= 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_0 + H \\ &= 2J(0)S\sigma_0 + H. \end{aligned} \quad (9.4)$$

We consider also that σ_{my} and σ_{mz} fluctuate about zero. We shall be able to show that all these fluctuations vanish above H_0 defined by (7.4) and that below H_0 oscillation begins in σ_{my} with the wave vector \mathbf{Q} and with an amplitude proportional to $(H_0 - H)^{1/2}$. The oscillation in σ_{my} and a second-order variation in σ_{mx} below H_0 form a fan structure. σ_{mz} vanishes above H_0 and in a certain range below H_0 .

Putting $\sigma_{nx} = \sigma_0 + \sigma_{nx}'$ and $H_{nx}' = 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_{mx}'$, we can write the energy of the spin at \mathbf{R}_n as

$$-[H^*S_x + H_{nx}'S_x + H_{ny}S_y + H_{nz}S_z - w(S_z)].$$

We substitute this into (3.3) and expand the exponential function in powers of H_{nx}' , H_{ny} , H_{nz} , using the mathematical method described in footnote 9b, page 319. Picking up only linear terms, we obtain equations of the following form:

$$S\sigma_{nx}' = H_{nx}'A(\beta, H^*), \quad S\sigma_{ny} = H_{ny}B(\beta, H^*), \quad S\sigma_{nz} = H_{nz}C(\beta, H^*), \quad (9.5)$$

where

$$\begin{aligned} A(\beta, H^*) &= \text{tr } S_x \int_0^\beta \exp [(\beta - \lambda)(H^*S_x - w)] S_x \\ &\times \exp [\lambda(H^*S_x - w)] d\lambda / \text{tr } \exp [\beta(H^*S_x - w)] - S^2\sigma_0^2, \end{aligned}$$

$$\begin{aligned}
 B(\beta, H^*) &= \text{tr } S_y \int_0^\beta \exp [(\beta - \lambda)(H^*S_x - w)] S_y \\
 &\quad \times \exp [\lambda(H^*S_x - w)] d\lambda / \text{tr} \exp [\beta(H^*S_x - w)], \\
 C(\beta, H^*) &= \text{tr } S_z \int_0^\beta \exp [(\beta - \lambda)(H^*S_x - w)] S_z \\
 &\quad \times \exp [\lambda(H^*S_x - w)] d\lambda / \text{tr} \exp [\beta(H^*S_x - w)].
 \end{aligned} \tag{9.6}$$

These are certain susceptibilities and are decreasing functions of H^* . Namely, with increasing H^* , the thermal average spin increases in the x direction, so that the squared spin deviations in the y and z directions decrease, and hence B and C decrease. It may also be imagined that the spin deviation in the x direction from its average value, S_{σ_0} , decreases with increasing H^* , and hence A decreases. A proof of these will be given in Section A1 of the Appendix. One should also be able to show that B is the largest, C the medium, and A the smallest ($B > C > A$); this follows from the fact that the susceptibility in the y direction is larger than that in the z direction (since there is no anisotropy energy to prevent the rotation of the spin in the xy plane, whereas there is such an anisotropy energy in the xz plane) and that the susceptibility in the x direction (which is the parallel susceptibility in the ferromagnetic state) is expected to be very small (see also the Appendix for the proof of $B > A$). It can be shown further that

$$B(\beta, H^*) = S_{\sigma_0}/H^*. \tag{9.7}$$

This can be proved in the following way. Consider

$$\text{tr } S_y \exp \beta[H^*S_x - w(S_z)], \tag{9.8}$$

and transform each of the operators after the symbol tr by $\exp(i\theta S_z)$, which means a rotation by an angle of θ about the z axis. We know that

$$\begin{aligned}
 \exp(i\theta S_z) S_x \exp(-i\theta S_z) &= S_x \cos \theta - S_y \sin \theta, \\
 \exp(i\theta S_z) S_y \exp(-i\theta S_z) &= S_x \sin \theta + S_y \cos \theta.
 \end{aligned}$$

Then, expression (9.8) can be written, for small θ , as

$$\text{tr } (\theta S_x + S_y) \exp \beta[H^*S_x - H^*\theta S_y - w(S_z)]. \tag{9.9}$$

Expanding the exponential in powers of θ and taking the linear terms, we obtain

$$\begin{aligned}
 \text{tr } \theta S_x \exp [\beta(H^*S_x - w)] - \text{tr } H^*\theta S_y \int_0^\beta \exp [(\beta - \lambda)(H^*S_x - w)] S_y \\
 \times \exp [\lambda(H^*S_x - w)] d\lambda.
 \end{aligned} \tag{9.10}$$

Now, we note that (9.8) vanishes, which can be seen by a similar transformation with $\theta = \pi$ about the x axis, by which S_y and S_z change sign. Hence, (9.10) also vanishes. Therefore, by the definition of $S\sigma_0$, (9.3), and the definition of B , (9.6), we obtain the result (9.7). It might be added that we can show in a similar way, starting with $\text{tr } S_z \exp \beta[H^*S_x - w(S_z)]$, that

$$\text{tr } S_z \int_0^\beta \exp [(\beta - \lambda)(H^*S_x - w)] S_y \exp [\lambda(H^*S_x - w)] d\lambda = 0. \quad (9.11)$$

S_z and S_y can be interchanged in this expression.

Now, to solve Eqs. (9.5), we put σ_{nx}' , σ_{ny} , σ_{nz} all proportional to $\exp(i\mathbf{q} \cdot \mathbf{R}_n)$, with amplitudes σ_x' , σ_y , σ_z , respectively, and substitute Eqs. (9.2) for H_{ny} and H_{nz} and $H_{nx}' = 2 \sum_m J(\mathbf{R}_{mn}) S\sigma_{mx}'$ into (9.5). Then we have

$$\begin{aligned} \sigma_x' &= 2J(\mathbf{q})A(\beta, H^*)\sigma_z', \\ \sigma_y &= 2J(\mathbf{q})B(\beta, H^*)\sigma_y, \\ \sigma_z &= 2J(\mathbf{q})C(\beta, H^*)\sigma_z. \end{aligned} \quad (9.12)$$

These equations give vanishing σ_x' , σ_y , and σ_z unless

$$1 = 2J(\mathbf{q})A, \quad 1 = 2J(\mathbf{q})B, \quad \text{and} \quad 1 = 2J(\mathbf{q})C,$$

respectively. However, in actuality, owing to the existence of nonlinear terms in H_{nx}' , H_{ny} , H_{nz} which we have neglected, we have nonvanishing σ_x' , etc., when, and only when, $1 \leq 2J(\mathbf{Q})A$, etc. This situation is similar to that we encountered in Chapters I and III in determining the Néel temperature. Furthermore, to determine the highest field H^* , i.e., the real critical field, above which the fluctuations vanish, we have to take the maximum of $J(\mathbf{q})$, since A , etc., are decreasing functions of H^* . So we put $\mathbf{q} = \mathbf{Q}$.

Since B is the largest, there is a field at which $1 = 2J(\mathbf{Q})B$, whereas $1 > 2J(\mathbf{Q})A$ and $1 > 2J(\mathbf{Q})C$. Below this field, σ_{ny} should become oscillating, whereas σ_{nx}' and σ_{nz} remain still vanishing as far as first-order changes are concerned. By virtue of (9.4) and (9.7), the condition $1 = 2J(\mathbf{Q})B$ can be rewritten as

$$H^* \equiv 2J(0)S\sigma_0 + H = 2J(\mathbf{Q})S\sigma_0,$$

or

$$H = 2[J(\mathbf{Q}) - J(0)]S\sigma_0.$$

In this equation, $S\sigma_0$ can be put equal to $S\sigma$, where $S\sigma$ is the thermal average magnitude of spins in the helical state in *no external field*. This is because the effective field in the present case is $H^* = 2J(\mathbf{Q})S\sigma_0$, and the exchange field in the helical state is $2J(\mathbf{Q})S\sigma$, and hence the equations to

determine σ_0 and σ are the same. Writing H_0 for H , we have, therefore,

$$H_0 = 2[J(\mathbf{Q}) - J(0)]S\sigma. \quad (9.13)$$

This is nothing but Eq. (7.4).

We can proceed to determining the amplitude of σ_{ny} , as well as σ_{nx}' , below H_0 by taking account of higher powers of H_{ny} and H_{nz}' in the molecular-field self-consistency equations. It turns out that H_{nz}' is of the order of H_{ny}^2 below H_0 . It can be shown also that σ_{nz} and H_{nz} vanish throughout. These calculations are described in the Appendix. We shall mention here the results only.

Below H_0 , the y component, σ_{ny} , oscillates with the wave vector \mathbf{Q} and an amplitude proportional to $(H_0 - H)^{1/2}$. The x component, σ_{nx}' , consists of a constant part, σ_{x0}' , and an oscillating part with wave vector $2\mathbf{Q}$, both parts being proportional to $H_0 - H$. From σ_{x0}' , the susceptibility of the fan structure can be calculated, and it is given by

$$\chi_{\text{fan}} = \frac{3\beta_2 + 2\gamma}{3\beta_2 + (2 + \beta_2)\gamma} \chi_z, \quad (9.14)$$

where β_2 and γ are defined by (8.9) and (8.10), γ being zero at T_N and infinite at $T = 0$.

We can now discuss the transition between the helical state and the fan state. The free energy of the helical state at a field H , relative to that at $H = 0$, is $-\frac{1}{2}\chi_h H^2$. At $H = H_0$, we have seen that the effective field has the same magnitude as that at $H = 0$, so that the value of the entropy at $H = H_0$ and that at $H = 0$ are equal. The energy at $H = H_0$ is $-\frac{1}{2}S\sigma H_0$. Thus, the energy of the fan state at a field H can be written approximately as

$$-\frac{1}{2}S\sigma H_0 + S\sigma(H_0 - H) - \frac{1}{2}\chi_{\text{fan}}(H_0 - H)^2. \quad (9.15)$$

The field of transition, H_t' , which has the same meaning as that illustrated in Fig. 1b, can be obtained by equating (9.15) with $-\frac{1}{2}\chi_h H^2$. We have

$$H_t' = \frac{[(\chi_z - \chi_{\text{fan}})(\chi_z - \chi_h)]^{1/2} - (\chi_z - \chi_{\text{fan}})}{\chi_{\text{fan}} - \chi_h} H_0. \quad (9.16)$$

This falls between $0.5H_0$ and $0.414H_0$.

If the out-of-plane anisotropy energy is not large enough to keep the spin vectors in the plane, there will be a field below H_0 where the z -component starts to oscillate, giving rise to an elliptic oscillation. Upon further lowering the field, this elliptic oscillation will discontinuously transform to the helical ordering. These transitions were discussed by Nagamiya *et al.*¹⁴ for $T = 0$, but we shall not go into this subject here.

10. STRUCTURE CHANGES WITH ANISOTROPY IN THE PLANE (LOW TEMPERATURES)

We shall briefly consider magnetization processes in a system in which there is initially a helical ordering and there is an anisotropy in the plane of the spin rotation. Also, in the next section, we shall briefly discuss the case of a conical structure. Since these are of interest mainly at low temperatures, the entropy term in the free energy may be neglected, and the consideration may be confined to the problem of minimum energy. An external magnetic field is supposed to be perpendicular to the screw axis.

Consider the case of a helical structure with an in-plane anisotropy energy. For a strong field applied in one of the easy directions in the plane, the spins may be aligned all parallel to the field direction. The spin-wave frequencies in this case were calculated in Section 5, and it was mentioned in Section 9 that some of the spin waves become unstable below H_0 given by (9.1). We may then have a fan structure. We shall here consider this problem in some detail.

We have an anisotropy energy GS_y^2 for small deviations of a spin in the y -direction from the easy direction x . For a deviation of an angle φ , this energy can be written as $GS^2\varphi^2$; the corresponding restoring torque is $-2GS^2\varphi$. An equivalent torque is obtained when a field of magnitude $2GS$ is applied in the x -direction. From this consideration follows the term $-2GS$ in H_0 , Eq. (9.1). Now we assume that φ oscillates sinusoidally as

$$\varphi_n = \eta \sin (\mathbf{Q} \cdot \mathbf{R}_n). \quad (10.1)$$

Then, the mean value of $GS^2\varphi_n^2$ over n is

$$GS^2\langle\varphi_n^2\rangle_n = \frac{1}{2}GS^2\eta^2. \quad (10.2)$$

The mean value of the exchange energy per atom up to $O(\eta^4)$ is

$$\begin{aligned} -S^2\langle\sum_m J(\mathbf{R}_{mn}) \cos(\varphi_m - \varphi_n)\rangle_n \\ = -S^2J(0) + \frac{1}{2}S^2\langle\sum_m J(\mathbf{R}_{mn})(\varphi_m - \varphi_n)^2\rangle_n \\ - \frac{1}{24}S^2\langle\sum_m J(\mathbf{R}_{mn})(\varphi_m - \varphi_n)^4\rangle_n \\ = -S^2J(0) - \frac{1}{2}S^2\eta^2[J(\mathbf{Q}) - J(0)] \\ + \frac{1}{32}S^2\eta^4[4J(\mathbf{Q}) - 3J(0) - J(2\mathbf{Q})]. \end{aligned} \quad (10.3)$$

The mean interaction energy of the spins with the external field, also up to $O(\eta^4)$, is

$$-HS\langle\cos\varphi_n\rangle_n = -HS + \frac{1}{2}HS\eta^2 - \frac{1}{64}HS\eta^4. \quad (10.4)$$

The sum of these three energies, (10.2), (10.3), and (10.4), may be minimized with respect to η . Then we have the following equation:

$$\eta^2 = \frac{8(H_0 - H)}{2S[4J(\mathbf{Q}) - 3J(0) - J(2\mathbf{Q})] - H}, \quad (10.5)$$

where (9.1) was used for H_0 . Assuming that H is close to H_0 , we may replace H in the denominator by H_0 . Then,

$$\eta^2 = \frac{8(H_0 - H)}{2S[3J(\mathbf{Q}) - 2J(0) - J(2\mathbf{Q}) + G]}. \quad (10.6)$$

From this, we can derive the differential susceptibility of the fan structure. Namely, the mean x component of the spins is $S\langle\cos\varphi_n\rangle_n$, which is $S(1 - \eta^2/4)$ to order η^2 , so that

$$\chi_{\text{fan}} = [3J(\mathbf{Q}) - 2J(0) - J(2\mathbf{Q}) + G]^{-1} \quad (10.7)$$

[multiplied by $(g\mu_B)^2$ in the original notation].

Equations (10.7) and (9.14) become identical in the limit of $T = 0$ ($\gamma = \infty$) and $G = 0$. Since H_0 becomes lower and χ_{fan} smaller by the introduction of G , the field of transition, H_t' , between the fan and the helix, relative to H_0 , becomes higher. This prediction is of course based on the assumption that the amplitude of the fan at the transition field is still small and the in-plane anisotropy energy can be written as $GS^2\varphi^2$.

If, however, the anisotropy energy has a φ^4 -term, the situation becomes a little different. Assume, for instance, an anisotropy energy of the form

$$\frac{1}{8}GS^2[1 - \cos 6\varphi]. \quad (10.8)$$

Expansion of this with respect to φ gives $GS^2\varphi^2 - 3GS^2\varphi^4 + \dots$. Correspondingly, we have, by a calculation similar to that we have made the following susceptibility formula:

$$\chi_{\text{fan}} = [3J(\mathbf{Q}) - 2J(0) - J(2\mathbf{Q}) - 35G]^{-1}. \quad (10.9)$$

Notice that the G -term in the denominator changed from $+G$ to $-35G$. It would then be quite possible that the denominator becomes *negative* for somewhat large G . In such a case, the magnetization curve would look like Fig. 2. We may then have a transition of the first kind between the fan state and the ferromagnetic state at a field H_0' which is higher than H_0 . For high values of G , we might even have a direct transition between the helical state and the ferromagnetic state without an intermediate fan state. In fact, these transitions have been predicted by Kitano and Nagamiya¹⁵ by calculations in which the anisotropy energy (10.8) was exactly taken into account, but the fan state was approximated by a

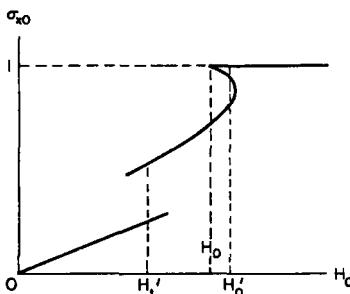


FIG. 2. Magnetization curve of a helical spin system for a field in the easy plane, in the case where there is a large anisotropy energy within the plane; the field is applied along one of the easy axes; the temperature is absolute zero.

purely sinusoidal oscillation, and for the helical state the in-plane anisotropy energy was neglected. Actually, however, there should be certain modulations of the fan and helical configurations because of the anisotropy energy of a multifold symmetry. A more exact mathematical treatment is thus desirable.

In the case where the external field is along one of the hard axes in the easy plane, discontinuous transitions helix-fan-ferro are predicted, as long as the anisotropy energy in the plane is not too large. On the other hand, if the anisotropy energy is large, one obtains a parallel alignment not in the field direction but near that potential minimum which is closest to the field direction. This parallel alignment appears between the helix and the fan.

These results are shown by the examples in Figs. 3a and 3b.

11. STRUCTURE CHANGES OF A CONICAL ARRANGEMENT

Magnetization processes of a conical structure for a field applied perpendicular to the cone axis involve even more complex structure changes. For an axially symmetric anisotropy energy, it was predicted¹⁵ that, depending on the functional form of this anisotropy energy, some or all of the following intermediate structures appear with increasing field (see Fig. 4):

- (1) Conical structure, a little distorted and inclined toward the field direction.
- (2) Fan on a conical surface whose vertex angle, θ , is smaller than the angle of the original cone, θ_1 , but tends to θ_1 for vanishing width of the fan.
- (3) Ferromagnetic alignment oblique to the field direction.
- (4) Fan in the meridian plane, obliquely disposed to the field direction.

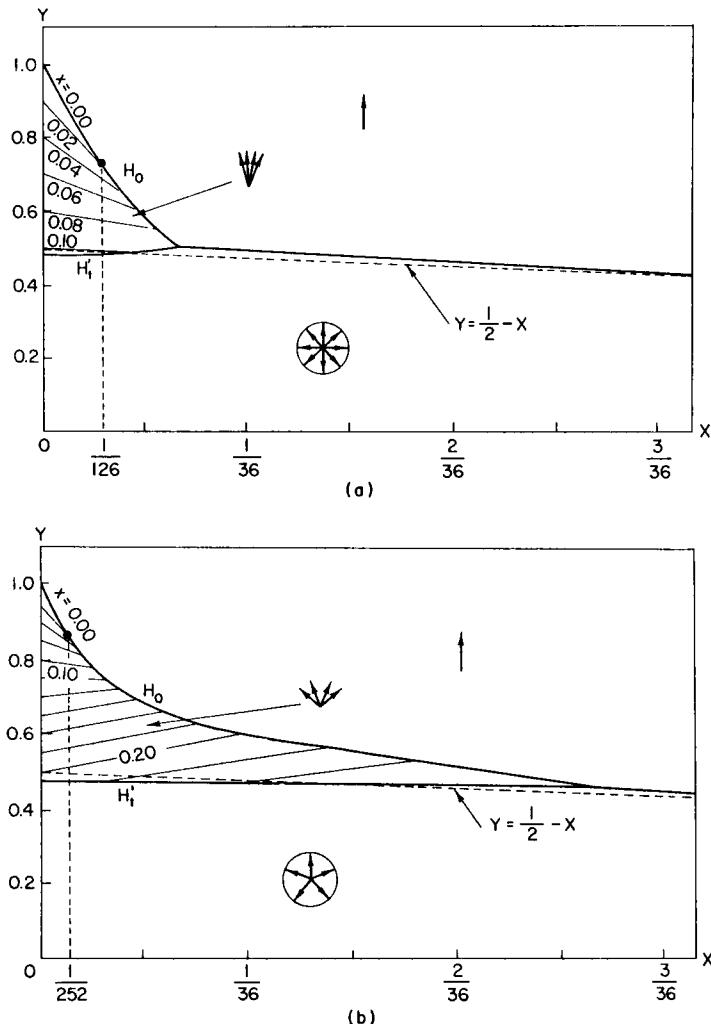


FIG. 3. Examples of structure changes due to a field applied along one of the easy axes in the easy plane (after Kitano and Nagamiya). (a) $\beta_2 = [J(\mathbf{Q}) - J(2\mathbf{Q})]/[J(\mathbf{Q}) - J(0)] = 8$; (b) $\beta_2 = 3$. The abscissa measures the strength of the anisotropy energy of sixfold symmetry within the easy plane: $X = V_6/2S[J(\mathbf{Q}) - J(0)]$, where V_6 is the coefficient of the one-ion anisotropy energy of the form $-V_6 \cos 6\phi$. The ordinate measures the field strength: $Y = H/2S[J(\mathbf{Q}) - J(0)]$. When H is increased, a first-order transition from helix to fan occurs at H_t' , and then a second-order transition (left of the broken vertical line) or a first-order transition (right of the same) occurs from fan to ferromagnetic alignment. For high values of X , a direct first-order transition occurs from helix to ferromagnetic alignment. A broken, nearly horizontal line is an approximation to H_t' and to the field of direct transition helix-ferromagnetism. Lines in the fan region are contours of constant amplitude of the fan, the attached numerical values indicating ξ defined by $\sin(\phi_n/2) = \xi \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha)$.

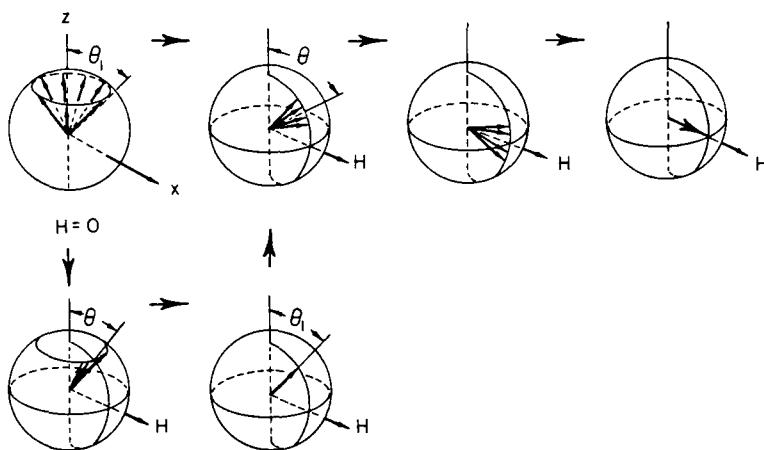


FIG. 4. Structure changes from a conical spin arrangement due to an applied field. The anisotropy energy is assumed to be a function of the polar angle θ only, to have a single minimum at an angle between $\theta = 0$ and $\theta = \pi/2$, and to vary smoothly without showing pronounced fluctuations. Arrows indicate the path of structure changes with increasing field. The direct path going to the right is taken when the minimum of the anisotropy energy is shallow, and the downward round path is taken when the minimum is deep.

(5) Fan in the meridian plane, symmetrically disposed with respect to the field direction.

(6) Ferromagnetic alignment in the field direction.

The fifth structure may appear when the anisotropy energy has a maximum at the horizontal plane ($\theta = \pi/2$). If it has a minimum (the second minimum) at $\theta = \pi/2$, a fan in the horizontal plane, rather than in the meridian plane, may appear. A brief discussion of the appearance of these structures will be given below.

Denoting by θ_n and φ_n the polar and azimuthal angles of the n th spin, we can write the total energy of the system as

$$E = -S^2 \sum_m \sum_n J(\mathbf{R}_{mn}) [\cos \theta_m \cos \theta_n + \sin \theta_m \sin \theta_n \cos (\varphi_m - \varphi_n)] \\ - SH \sum_n \sin \theta_n \cos \varphi_n + \sum_n w(\cos^2 \theta_n). \quad (11.1)$$

To derive a fan structure, we put

$$\theta_n = \theta + 2\xi \cos (\mathbf{R}_n \cdot \mathbf{Q} + \alpha), \quad (11.2)$$

$$\varphi_n = 2\xi \sin (\mathbf{R}_n \cdot \mathbf{Q} + \beta),$$

and substitute these into (11.1). Then, up to the second powers of ξ and ξ , we have

$$\begin{aligned} E/N = & -S^2 J(0) - SH \sin \theta + w(\cos^2 \theta) \\ & + [-2S^2\{J(\mathbf{Q}) - J(0)\} \sin^2 \theta + SH \sin \theta] \xi^2 \\ & + [-2S^2\{J(\mathbf{Q}) - J(0)\} + SH \sin \theta + (d^2 w/d\theta^2)] \xi^2. \end{aligned} \quad (11.3)$$

If ξ^2 and ξ^2 are neglected, the value of θ is determined from the minimum of the first line of (11.3), namely, from

$$-SH \cos \theta - 2w'(\cos^2 \theta) \cos \theta \sin \theta = 0,$$

or

$$SH = -2w'(\cos^2 \theta) \sin \theta. \quad (11.4)$$

The coefficient of ξ^2 in (11.3) can, therefore, be written as

$$-[2S^2\{J(\mathbf{Q}) - J(0)\} + 2w'(\cos^2 \theta)] \sin^2 \theta. \quad (11.5)$$

By (6.4), this vanishes at $\theta = \theta_1$, where θ_1 is the angle of the cone at zero field. Since w has a minimum near this angle, $dw/d\theta$ is an increasing function of θ near θ_1 , namely, $w'(\cos^2 \theta)$ is a decreasing function of θ near θ_1 . Thus, (11.5) is negative for $\theta < \theta_1$ and positive for $\theta > \theta_1$. This would mean that ξ is nonvanishing for $\theta < \theta_1$ and vanishing for $\theta > \theta_1$. The field, H_1 , corresponding to θ_1 can be calculated from (11.4) to be

$$H_1 = 2S[J(\mathbf{Q}) - J(0)] \sin \theta_1, \quad (11.6)$$

where $-2w'(\cos^2 \theta_1)$ was replaced by $2S^2[J(\mathbf{Q}) - J(0)]$, since (11.5) vanishes for $\theta = \theta_1$. Below this field, ξ would be nonvanishing, and one might have structure 2, whereas above this field structure 3 might be realized. However, before concluding this, one has to make a few more considerations. The coefficient of ξ^2 in (11.3) must be positive in the neighborhood of θ_1 in order to have structures 2 and 3, as otherwise ξ would be nonvanishing, and an oscillation in the θ -direction would take place simultaneously. Furthermore, the energy (11.3) at θ_1 with $\xi = \xi = 0$ must be lower than the energy of structure 1 extrapolated to $H = H_1$. It can be shown after some calculations that the former energy is actually lower than the latter energy when the coefficient of ξ^2 is positive at θ_1 . Thus, with the assumption of the positive sign of the coefficient of ξ^2 at θ_1 , one obtains structures 2 and 3.

The amplitude ξ , of the φ -oscillation, can be calculated when the ξ^4 -term is included in the energy expression. It is more convenient to define ξ by

$$\sin(\varphi_n/2) = \xi \sin(\mathbf{R}_n \cdot \mathbf{Q} + \beta) \quad (11.7)$$

than by (11.2). With this definition of ξ , the ξ^4 -term can be calculated

rather easily, and it can be shown that this term is expressed as

$$\frac{1}{2}S^2[3J(Q) - 2J(0) - J(2Q)] \sin^2 \theta \cdot \xi^4, \quad (11.8)$$

which is positive. Thus, we can calculate ξ^2 below H_1 as a function of H or θ by minimizing the energy with respect to ξ .

When the coefficient of ξ^2 in (11.3) at θ_1 is negative, it can be shown that the energy (11.3) at θ_1 with $\xi = \zeta = 0$ is higher than the energy of structure 1 extrapolated to $H = H_1$. Although no detailed calculations have been made in this case, it is very likely that structure 1 persists beyond H_1 and transforms discontinuously at a higher field to structure 4 or 5, or even 6.

We can write the coefficient of ξ^2 in (11.3), using (11.4), as

$$-2S^2[J(Q) - J(0)] - 2w'(\cos^2 \theta) \cos^2 \theta + 4w''(\cos^2 \theta) \sin^2 \theta \cos^2 \theta.$$

This is equal to $-2S^2[J(Q) - J(0)]$ at $\theta = \pi/2$, which is negative. For θ close to $\pi/2$, it must also be negative. Thus, one has an oscillation in the θ -direction at a field that makes θ close to or equal to $\pi/2$. Structures 4 and 5 result in this way. At a field higher than that which makes θ equal to $\pi/2$, (11.4) is no longer valid, but the coefficient of ξ^2 in (11.3) will still remain negative until the field reaches a value, H_0 , given by

$$H_0 = 2S[J(Q) - J(0)] - S^{-1}(d^2w/d\theta^2) |_{\theta=\pi/2}. \quad (11.9)$$

At this field, structure 6, namely, the ferromagnetic alignment in the field direction, will set in, and it will persist beyond H_0 . These predictions are, however, valid only when the anisotropy energy, w , has a maximum at $\theta = \pi/2$, in which case the last term of (11.9) is positive. If w has a minimum at $\theta = \pi/2$, this term is negative and the coefficient of ξ^2 in (11.3) for $\theta = \pi/2$ is negative at H_0 , given by (11.9), and will remain negative up to a higher field equal to $2S[J(Q) - J(0)]$. Thus, in this case, one should have a fan in the horizontal plane, rather than in the meridian plane, up to $H = 2S[J(Q) - J(0)]$.

12. EXPERIMENTAL OBSERVATIONS

The first example in which magnetization processes were observed by neutron diffraction is an ordered alloy MnAu₂. This was studied earlier by magnetic measurements by Meyer and Taglang¹⁷: there was a rather steep rise in the magnetization curve at about 10 kOe, followed by a gradual increase going to saturation (polycrystalline material). This behavior of the magnetization curve is called *metamagnetic*. By neutron diffraction experiment, Herpin and Mériel¹⁸ studied structure changes in this alloy with

¹⁷ A. J. P. Meyer and P. Taglang, *J. Phys. Radium* **17**, 457 (1956).

the application of a magnetic field, and they discovered the fan structure as the intermediate phase. They were also the first to propose the theory of the transitions from helix to fan and then to ferromagnetic alignment.

The crystal of MnAu_2 consists of a tetragonal body-centered lattice of Mn having two (001) layers of Au between adjacent (001) layers of Mn ($a = 3.37 \text{ \AA}$, $c/a = 2.60$ at room temperature). The Néel temperature T_N is 365°K . The spin structure is helical, rotating in the c -plane and propagating in the c -direction. The turn angle measured by neutron diffraction is 51° at 300°K , which corresponds to a period of seven layers of Mn, but decreases to 46° at 125°K and then increases to 47° at 87°K . The moment value of each spin extrapolated to $T = 0$ is $3.5 \mu_B$. With an applied field, the helical structure changes abruptly to the fan structure at 10 kOe, the angle of the fan diminishing with increasing field and finally vanishing at about 15 kOe. Although their samples were polycrystalline, they could select, by applying the field parallel or perpendicular to the scattering vector, reflections only from those particles for which the field was within the c -plane. However, there must have been particles for which the field was along the easy, hard, and intermediate directions in the c -plane.

For rare-earth metals Dy and Ho, structure changes with applied magnetic field have been observed in detail with single crystals. These will be described in the forthcoming Part 2. A brief account will be given here, however. In Dy, in the range of temperature where the spin structure is helical at zero field, there is evidence that simple helix-fan-ferro transitions occur when the field is applied in the hexagonal basal plane (the plane of the spin rotation). In Ho, in the range between 133°K (T_N) and 80°K , in which there is no appreciable anisotropy energy within the hexagonal basal plane, also simple helix-fan-ferro transitions have been observed, but below 80°K , where there is an appreciable anisotropy energy in the plane, two intermediate fan structures have been observed. The original helical structure and these two fan structures all have different periods in space. In our theory, the function $J(\mathbf{q})$ was assumed to be independent of the applied field, and so was the value of \mathbf{Q} which makes $J(\mathbf{q})$ maximum. The variation of \mathbf{Q} with changes in the spin structure must be related to the state of the conduction electrons, but this is not yet fully understood. Also, our theory does not predict the appearance of two intermediate fan structures.

In Ho, in a small temperature range above 20°K , there is a single transition from helix to ferromagnetic alignment. This can be understood theoretically (see Figs. 3a and 3b in a high-anisotropy range). Below 20°K , we have a conical spin structure in zero field, and this transforms to a ferromagnetic alignment oblique to the field (structure 3) and then to the ferromagnetic alignment parallel to the field (structure 6).

V. Spin Waves in Various Configurations in an Applied Field

In this Chapter, we shall study spin waves in the following three spin configurations:

- (1) Conical arrangement arising from applied field or anisotropy energy or both,
- (2) Fan arrangement due to an applied field,
- (3) Helical arrangement in a weak field.

We shall particularly study those modes and frequencies that are resonant to an imposed oscillating magnetic field, for which we have to take account of the oscillating demagnetizing field. Spin waves in these structures have been studied mostly by Cooper, Elliott, and co-workers.^{18,19,19a}

Taking a particular spin, \mathbf{S}_n , we denote its equilibrium direction by ξ , the direction perpendicular to this and the z -axis (cone axis in the first case, normal to the plane containing the spin vectors in the second and third cases) by ξ , and the direction perpendicular to ξ and η by η ; the positive ξ -direction will be assumed to coincide with the direction of right-hand rotation about the z -axis. Then, referring to Chapter II, we can write

$$\begin{aligned} S_{n\xi} &= S - a_n^* a_n, \\ S_{n\xi} &= (\frac{1}{2}S)^{1/2}(a_n^* + a_n), \\ S_{n\eta} &= i(\frac{1}{2}S)^{1/2}(a_n^* - a_n). \end{aligned} \quad (V.1)$$

If we denote by θ and φ_n the polar and azimuthal angles of the equilibrium direction of \mathbf{S}_n (the polar axis being the z -axis), we can write

$$\begin{aligned} S_{nx} &= -S_{n\xi} \sin \varphi_n - S_{n\eta} \cos \theta \cos \varphi_n + S_{n\xi} \sin \theta \cos \varphi_n, \\ S_{ny} &= +S_{n\xi} \cos \varphi_n - S_{n\eta} \cos \theta \sin \varphi_n + S_{n\xi} \sin \theta \sin \varphi_n, \\ S_{nz} &= S_{n\eta} \sin \theta + S_{n\xi} \cos \theta. \end{aligned} \quad (V.2)$$

With these expressions, we write the exchange energy, the anisotropy energy, the Zeeman energy, and the demagnetizing energy in terms of a_n^* and a_n . In calculating the spin-wave frequencies and modes, we take up only those terms which are quadratic in a_n^* and a_n , and transform these variables to a_q^* and a_q , defined by (II.6). Then we diagonalize the resulting Hamiltonian with another transformation. The condition for equilib-

¹⁸ B. R. Cooper, R. J. Elliott, S. J. Nettel, and H. Suhl, *Phys. Rev.* **127**, 57 (1962); B. R. Cooper, *Proc. Phys. Soc. (London)* **80**, 1225 (1962).

¹⁹ B. R. Cooper and R. J. Elliott, *Phys. Rev.* **131**, 1043 (1963).

^{19a} Most of their results had been obtained by A. Watabe, H. Miwa, and K. Yosida [reported in a now discontinued mimeograph journal, *Busseiron Kenkyu* (in Japanese), April 1961].

rium follows either by putting the linear terms of the Hamiltonian equal to zero or by minimizing the constant term. We take the Hamiltonian to be

$$\begin{aligned} \mathcal{H} = & - \sum_{m,n} J(\mathbf{R}_{mn}) \mathbf{S}_m \cdot \mathbf{S}_n + \sum_n [w(\mathbf{S}_n) - \mathbf{H} \cdot \mathbf{S}_n] \\ & + \text{demagnetizing energy}, \end{aligned} \quad (\text{V.3})$$

where $w(\mathbf{S}_n)$ is a one-atom type anisotropy energy, and we have written \mathbf{H} for $g\mu_B \mathbf{H}$ (more exactly, $-g\mu_B \mathbf{H}$, but then we may reverse the direction of \mathbf{H}). We shall consider the oscillating demagnetizing field only for *uniform modes*. Assuming an ellipsoidal shape of the sample, whose principal axes are along x , y , z , and the volume of the sample to be 1, we can write the demagnetizing energy as

$$\frac{1}{2} [N_x(g\mu_B \sum_n S_{nx})^2 + N_y(g\mu_B \sum_n S_{ny})^2 + N_z(g\mu_B \sum_n S_{nz})^2],$$

where N_x , N_y , N_z are the demagnetizing factors ($N_x + N_y + N_z = 4\pi$). However, to simplify the notation, we shall write this as

$$(1/2N) [N_x(\sum_n S_{nx})^2 + N_y(\sum_n S_{ny})^2 + N_z(\sum_n S_{nz})^2], \quad (\text{V.4})$$

where N is the number of atoms in unit volume; N_x stands for $N(g\mu_B)^2 N_x$, etc.

13. CONICAL ARRANGEMENT; FIELD PARALLEL TO z

If we have initially a proper screw structure, a field applied normal to the screw plane, namely, parallel to z , will change it to a conical one. If we have initially a conical arrangement, such a field will decrease the cone angle. In either case, the cone angle, θ , is given as a function of H , and it depends also on the functional form of $w(\mathbf{S}_n)$. We assume that $w(\mathbf{S}_n)$ is axially symmetric so that it is a function of S_{nz} only (an even function of S_{nz}). The equilibrium direction of \mathbf{S}_n , or the value of θ , can be determined by considering \mathbf{S}_n as a classical vector and minimizing the total energy, (V.3), with respect to θ , where φ_n is put equal to $\mathbf{Q} \cdot \mathbf{R}_n$.

To determine θ , we put $S_{n\xi} = S$, $S_{n\xi} = S_{n\eta} = 0$, $\varphi_n = \mathbf{Q} \cdot \mathbf{R}_n$ in the expression of the total energy and have

$$\begin{aligned} & -N[S^2 \sin^2 \theta J(\mathbf{Q}) + S^2 \cos^2 \theta J(0)] + Nw(S \cos \theta) - NH S \cos \theta \\ & + \frac{1}{2} NN_z S^2 \cos^2 \theta. \end{aligned} \quad (13.1)$$

We differentiate this expression with respect to θ and put it equal to zero:

$$-2S \cos \theta [J(\mathbf{Q}) - J(0)] - w'(S \cos \theta) + H - N_z S \cos \theta = 0. \quad (13.2)$$

Furthermore, out of the total energy we pick up

$$\sum_n [w(S_{nz}) - HS_{nz}] + (1/2N)N_z(\sum_n S_{nz})^2 \quad (13.3)$$

and expand this around $S_{n\xi} = S$, $S_{n\eta} = 0$ in powers of $S - S_{n\xi}$ and $S_{n\eta}$. Since $S - S_{n\xi} = a_n^* a_n$ is a second-order quantity in the spin-deviation operators, a_n^* and a_n , and $S_{n\eta}$ is a first-order quantity, we keep only those terms which are linear in $S - S_{n\xi}$ and $S_{n\eta}^2$. Then we have for (13.3)

$$\begin{aligned} & \sum_n \{-w'(S \cos \theta) \cos \theta (S - S_{n\xi}) + \frac{1}{2}w''(S \cos \theta) \sin^2 \theta S_{n\eta}^2 \\ & + H \cos \theta (S - S_{n\xi}) - N_z S \cos^2 \theta (S - S_{n\xi})\} \\ & + (1/2N)N_z \sin^2 \theta (\sum_n S_{n\eta})^2, \end{aligned}$$

or, with the use of (13.2),

$$\begin{aligned} & \sum_n \{2S \cos^2 \theta [J(\mathbf{Q}) - J(0)](S - S_{n\xi}) + DS_{n\eta}^2\} \\ & + (1/2N)N_z \sin^2 \theta (\sum_n S_{n\eta})^2, \end{aligned} \quad (13.4)$$

where

$$D = \frac{1}{2}w''(S \cos \theta) \sin^2 \theta. \quad (13.5)$$

It is noted that H has disappeared in (13.4), although θ is a function of H determined by (13.2).

The total energy is now the sum of the exchange energy, the energy (13.4), and the x and y parts of the demagnetizing energy. Those terms of the total energy which are quadratic in a_n^* and a_n can be calculated to be the following:

$$\begin{aligned} & \sum_{m,n} J(\mathbf{R}_{mn}) S \{ [\cos^2 \theta + \sin^2 \theta \cos \mathbf{Q} \cdot \mathbf{R}_{mn}] (a_m^* a_m + a_n^* a_n) \\ & - \frac{1}{2} [\sin^2 \theta + (1 + \cos^2 \theta) \cos \mathbf{Q} \cdot \mathbf{R}_{mn}] (a_m^* a_n + a_n^* a_m) \\ & + \frac{1}{2} \sin^2 \theta [1 - \cos \mathbf{Q} \cdot \mathbf{R}_{mn}] (a_m^* a_n^* + a_m a_n) \\ & + i \cos \theta \sin \mathbf{Q} \cdot \mathbf{R}_{mn} (a_m^* a_n - a_n^* a_m) \} \\ & + \sum_n 2S \cos^2 \theta [J(\mathbf{Q}) - J(0)] a_n^* a_n - \sum_n \frac{1}{2} D S (a_n^* - a_n)^2 \\ & + (1/4N)N_x S [\sum_n (a_n^* + a_n) \sin \mathbf{Q} \cdot \mathbf{R}_n + i(a_n^* - a_n) \cos \theta \sin \mathbf{Q} \cdot \mathbf{R}_n]^2 \\ & + (1/4N)N_y S [\sum_n (a_n^* + a_n) \cos \mathbf{Q} \cdot \mathbf{R}_n - i(a_n^* - a_n) \cos \theta \sin \mathbf{Q} \cdot \mathbf{R}_n]^2 \\ & - (1/4N)SN_z \sin^2 \theta [\sum_n (a_n^* - a_n)]^2. \end{aligned} \quad (13.6)$$

We make here the Fourier transformations:

$$a_n = N^{-1/2} \sum_{\mathbf{q}} a_{\mathbf{q}} \exp(i\mathbf{q} \cdot \mathbf{R}_n), \quad a_n^* = N^{-1/2} \sum_{\mathbf{q}} a_{\mathbf{q}}^* \exp(-i\mathbf{q} \cdot \mathbf{R}_n).$$

Then, (13.6) becomes

$$\begin{aligned} & \sum_{\mathbf{q}} 2S a_{\mathbf{q}}^* a_{\mathbf{q}} \{ [J(\mathbf{Q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q})] \\ & \quad - \frac{1}{2} \sin^2 \theta [J(\mathbf{q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q})] \\ & \quad + \frac{1}{2}D + \frac{1}{2} \cos \theta [J(\mathbf{Q} - \mathbf{q}) - J(\mathbf{Q} + \mathbf{q})] \} \\ & + \sum_{\mathbf{q}} S (a_{\mathbf{q}}^* a_{-\mathbf{q}}^* + a_{\mathbf{q}} a_{-\mathbf{q}}) \\ & \quad \times \{ \frac{1}{2} \sin^2 \theta [J(\mathbf{q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q})] - \frac{1}{2}D \} \\ & - \frac{1}{16} SN_z [(a_Q^* - a_Q)(1 - \cos \theta) - (a_{-Q}^* - a_{-Q})(1 + \cos \theta)]^2 \\ & + \frac{1}{16} SN_y [(a_Q^* + a_Q)(1 - \cos \theta) + (a_{-Q}^* + a_{-Q})(1 + \cos \theta)]^2 \\ & - \frac{1}{4} SN_z \sin^2 \theta (a_0^* - a_0)^2. \end{aligned} \quad (13.7)$$

Excepting the terms with $\mathbf{q} = 0$ and $\mathbf{q} = \pm \mathbf{Q}$, we write this as

$$\sum_{\mathbf{q}} 2S [A_{\mathbf{q}} a_{\mathbf{q}}^* a_{\mathbf{q}} + \frac{1}{2}B_{\mathbf{q}} (a_{\mathbf{q}}^* a_{-\mathbf{q}}^* + a_{\mathbf{q}} a_{-\mathbf{q}})]. \quad (13.7a)$$

It is noted that $A_{\mathbf{q}} \neq A_{-\mathbf{q}}$ due to the existence of a $\cos \theta$ term in the coefficient of $a_{\mathbf{q}}^* a_{\mathbf{q}}$ in (13.7), but $B_{\mathbf{q}} = B_{-\mathbf{q}}$.

To diagonalize (13.7a), we make the following transformation (see Chapter II):

$$a_{\mathbf{q}} = \alpha_{\mathbf{q}} \cosh \theta_{\mathbf{q}} + \alpha_{-\mathbf{q}}^* \sinh \theta_{\mathbf{q}}, \quad (13.8)$$

$$a_{\mathbf{q}}^* = \alpha_{\mathbf{q}}^* \cosh \theta_{\mathbf{q}} + \alpha_{-\mathbf{q}} \sinh \theta_{\mathbf{q}},$$

with

$$\coth 2\theta_{\mathbf{q}} = (A_{\mathbf{q}} + A_{-\mathbf{q}})/(-2B_{\mathbf{q}}). \quad (13.9)$$

Then (13.7a) becomes

$$\sum_{\mathbf{q}} 2S [A_{\mathbf{q}} \cosh^2 \theta_{\mathbf{q}} + A_{-\mathbf{q}} \sinh^2 \theta_{\mathbf{q}} + 2B_{\mathbf{q}} \cosh \theta_{\mathbf{q}} \sinh \theta_{\mathbf{q}}] \alpha_{\mathbf{q}}^* \alpha_{\mathbf{q}},$$

or

$$\sum_{\mathbf{q}} S [(A_{\mathbf{q}} - A_{-\mathbf{q}}) + (A_{\mathbf{q}} + A_{-\mathbf{q}}) \cosh 2\theta_{\mathbf{q}} + 2B_{\mathbf{q}} \sinh 2\theta_{\mathbf{q}}] \alpha_{\mathbf{q}}^* \alpha_{\mathbf{q}}. \quad (13.10)$$

Since

$$\cosh 2\theta_q = (1 - \tanh^2 2\theta_q)^{-1/2} = \frac{A_q + A_{-q}}{[(A_q + A_{-q})^2 - 4B_q^2]^{1/2}},$$

$$\sinh 2\theta_q = (\cosh^2 2\theta_q - 1)^{-1/2} = \frac{-2B_q}{[(A_q + A_{-q})^2 - 4B_q^2]^{1/2}},$$

provided that $A_q + A_{-q} > 0$,^{19b} the coefficient of $\alpha_q^* \alpha_q$ in (13.10), which is \hbar times the frequency, is calculated to be the following:

$$\hbar\omega_q = S(A_q - A_{-q}) + S[(A_q + A_{-q})^2 - 4B_q^2]^{1/2}, \quad (13.11)$$

or

$$\begin{aligned} \hbar\omega_q = & S \cos \theta [J(Q - q) - J(Q + q)] \\ & + 2S[J(Q) - \frac{1}{2}J(Q - q) - \frac{1}{2}J(Q + q)]^{1/2} \\ & \times \{J(Q) - \sin^2 \theta J(q) - \frac{1}{2} \cos^2 \theta [J(Q - q) + J(Q + q)] + D\}^{1/2}. \end{aligned} \quad (13.12)$$

In order that the assumed conical configuration be stable, all the frequencies must be positive. The first term of (13.11) or (13.12) changes sign when q and $-q$ are interchanged, so that the absolute value of this term must be smaller than the second term. For small values of q , the first term is of order of q^3 , since $J(q)$ is maximum at $q = Q$, whereas the second term is of order of q , so that the first term is surely smaller than the second term for small q . Also, near $q = Q$ and $q = -Q$, it is easy to see that this condition is satisfied. The frequency near $q = \pm Q$ is small when θ is close to $\pi/2$ and D is small.

We are particularly interested in the cases of $q = 0$ and $q = \pm Q$. In the case of $q = 0$, we have the last term of (13.7), but despite this, $A_0 + B_0$ vanishes when the Hamiltonian is written in the form of (13.7a); hence the frequency, of the form of (13.11), vanishes. This is to be expected, since the mode of oscillation for $q = 0$ is such that the cone rotates as a rigid body about its axis (the z -axis). If we had an axial anisotropy (of hexagonal symmetry, for instance), the azimuthal, as well as the polar, distribution of the spin vectors at equilibrium would not be uniform; nevertheless, a rotation of the system about the z axis, such that the nonuniform distribution of the spin vectors on the undulatory cone surface remains unchanged in the vector space, would produce just a shift of the spin pattern along the z axis, so that the frequency of such a spin-wave mode would

^{19b} If $A_q + A_{-q}$ is negative, we have a minus sign before the square root quantity; in this case, we have negative frequencies, so that the system is unstable.

be zero.^{19c} However, this is possible only when the period of the spin pattern is irrational in units of the lattice spacing, in which case the spin vectors cover the cone surface uniformly. If the period is rational, a finite number of spin vectors will fit in each potential valley, and the rotational oscillation will be coupled with a pulsation of the cone angle; hence the frequency will be nonvanishing. Evidently, such a motion would be active to oscillating magnetic field parallel to the z -axis.

For $\mathbf{q} = \pm\mathbf{Q}$, we have extra terms in (13.7) due to the demagnetizing fields in the x and y directions. Thus, the total expression (13.7) has the form

$$\begin{aligned} A^+a_Q^*a_Q + A^-a_{-Q}^*a_{-Q} + B(a_Q^*a_{-Q}^* + a_Qa_{-Q}) \\ + C(a_Q^*a_{-Q} + a_{-Q}^*a_Q) + \frac{1}{2}D^+(a_Q^*a_Q^* + a_Qa_Q) \\ + \frac{1}{2}D^-(a_{-Q}^*a_{-Q}^* + a_{-Q}a_{-Q}). \end{aligned} \quad (13.13)$$

In the case of a sample that is axially symmetric about the z -axis, C , D^+ , and D^- all vanish ($N_x = N_y$), so that the frequency is expressed in the same form as (13.11) with $\mathbf{q} = \mathbf{Q}$ or $\mathbf{q} = -\mathbf{Q}$. However, in this case, $A_{\pm Q}$ and B_Q contain demagnetizing terms:

$$\begin{aligned} A_{\pm Q} &= S\{(1 + \cos^2 \theta)[J(\mathbf{Q}) - \frac{1}{2}J(0) - \frac{1}{2}J(2\mathbf{Q}) + \frac{1}{8}(N_x + N_y)] + D\} \\ &\pm S \cos \theta[J(0) - J(2\mathbf{Q}) - \frac{1}{4}(N_x + N_y)], \end{aligned} \quad (13.14)$$

$$B_Q = S\{\sin^2 \theta[J(\mathbf{Q}) - \frac{1}{2}J(0) - \frac{1}{2}J(2\mathbf{Q}) - \frac{1}{8}(N_x + N_y)] - D\}. \quad (13.15)$$

Corresponding to ω_Q and ω_{-Q} there are two modes. For ω_Q , one puts $\alpha_{-Q} = \alpha_{-Q}^* = 0$ in (13.8) and obtains $a_Q/a_{-Q}^* = \cosh \theta_Q/\sinh \theta_Q$, or

$$\begin{aligned} (a_Q + a_{-Q}^*)/(a_Q - a_{-Q}^*) &= S_{Q\xi}/iS_{Q\eta} \\ &= (\cosh \theta_Q + \sinh \theta_Q)/(\cosh \theta_Q - \sinh \theta_Q) \\ &= \cosh 2\theta_Q + \sinh 2\theta_Q \\ &= [A_Q + A_{-Q} - 2B_Q]^{1/2}/[A_Q + A_{-Q} + 2B_Q]^{1/2}. \end{aligned} \quad (13.16)$$

For ω_{-Q} ($\alpha_Q = \alpha_Q^* = 0$), $\cosh \theta_Q$ and $\sinh \theta_Q$ are interchanged, and one has the negative of the ratio (13.16) for $S_{Q\xi}/iS_{Q\eta}$. Since $S_{n\xi}$ and $S_{n\eta}$ for the $+Q$ mode vary as $|S_{Q\xi}| \cos(\mathbf{Q} \cdot \mathbf{R}_n - \omega_Q t)$ and $|S_{Q\eta}| \sin(\mathbf{Q} \cdot \mathbf{R}_n - \omega_Q t)$,

^{19c} The writer is indebted for this explanation to Dr. Elliott, who has pointed out (privately) that there is always one mode with zero frequency. This is confirmed in Section 14 by an independent direct calculation for a special case. Cooper and Elliott,¹⁹ by an erroneous calculation, originally obtained a nonzero frequency for this case. See also the footnote 19g on page 361.

respectively,^{19d} each spin vector describes an ellipse in the right-handed sense when one sees it opposite to its direction. The whole cone performs a right-handed rocking rotational motion about the z axis; in space, the wave propagates with wave vector \mathbf{Q} . This mode is, therefore, active to a right-handed rotating electromagnetic field. For $\omega_{-\mathbf{Q}}$, the motion is just reverse in time, and it is active to a left-handed rotating electromagnetic field.

14. THE FAN WITH A FIELD PARALLEL TO x

In the fan, one has φ_n oscillating with position \mathbf{R}_n , with wave vector \mathbf{Q} ; θ is $\pi/2$. We may put, for convenience,¹⁴

$$\sin(\varphi_n/2) = 2\delta \sin(\mathbf{Q} \cdot \mathbf{R}_n), \quad (14.1)$$

where δ is assumed to be small; it varies with field as $(H_0 - H)^{1/2}$, where H_0 is the fan-ferro transition field. In the following, we shall keep terms up to $O(\delta^2)$.

For the exchange energy, the following quadratic form in spin-wave amplitudes is obtained:

$$\begin{aligned} 2S \sum_{\mathbf{q}} & \{ [[J(0) - J(\mathbf{q})] + 4\delta^2 [2J(\mathbf{Q}) - 2J(0) + C(\mathbf{q})]] a_{\mathbf{q}}^* a_{\mathbf{q}} \\ & + 2\delta^2 C(\mathbf{q}) (a_{\mathbf{q}}^* a_{-\mathbf{q}}^* + a_{\mathbf{q}} a_{-\mathbf{q}}) \\ & - 2\delta^2 [2J(\mathbf{Q}) - J(0) - J(2\mathbf{Q}) - C(\mathbf{q})] (a_{\mathbf{q}+\mathbf{Q}}^* a_{\mathbf{q}-\mathbf{Q}} + a_{\mathbf{q}-\mathbf{Q}}^* a_{\mathbf{q}+\mathbf{Q}}) \\ & + \delta^2 C(\mathbf{q}) (a_{\mathbf{q}+\mathbf{Q}}^* a_{-\mathbf{q}+\mathbf{Q}}^* + a_{\mathbf{q}-\mathbf{Q}}^* a_{-\mathbf{q}-\mathbf{Q}}^* + a_{\mathbf{q}+\mathbf{Q}} a_{-\mathbf{q}+\mathbf{Q}} + a_{\mathbf{q}-\mathbf{Q}} a_{-\mathbf{q}-\mathbf{Q}}) \}, \end{aligned} \quad (14.2)$$

where

$$C(\mathbf{q}) = J(\mathbf{q}) - \frac{1}{2}J(\mathbf{Q} - \mathbf{q}) - \frac{1}{2}J(\mathbf{Q} + \mathbf{q}). \quad (14.3)$$

We consider an anisotropy energy of p -fold symmetry about the z axis

^{19d} The classical picture is presented here. It follows from (13.8) that, for $\omega_{\mathbf{Q}}$,

$$a_{\mathbf{Q}} = \alpha_{\mathbf{Q}} \cosh \theta_{\mathbf{Q}}, \quad a_{-\mathbf{Q}}^* = \alpha_{\mathbf{Q}} \sinh \theta_{\mathbf{Q}}, \quad a_{-\mathbf{Q}} = \alpha_{\mathbf{Q}}^* \sinh \theta_{\mathbf{Q}}, \quad a_{\mathbf{Q}}^* = \alpha_{\mathbf{Q}}^* \cosh \theta_{\mathbf{Q}}$$

so that

$$\begin{aligned} S_{n\xi} &= (S/2)^{1/2} (a_n^* + a_n) \\ &= (S/2N)^{1/2} [(a_{\mathbf{Q}} + a_{-\mathbf{Q}}^*) \exp(i\mathbf{Q} \cdot \mathbf{R}_n) + (a_{-\mathbf{Q}} + a_{\mathbf{Q}}^*) \exp(-i\mathbf{Q} \cdot \mathbf{R}_n)] \\ &= (S/2N)^{1/2} (\cosh \theta_{\mathbf{Q}} + \sinh \theta_{\mathbf{Q}}) [\alpha_{\mathbf{Q}} \exp(i\mathbf{Q} \cdot \mathbf{R}_n) + \alpha_{\mathbf{Q}}^* \exp(-i\mathbf{Q} \cdot \mathbf{R}_n)] \\ &\propto \cos(\mathbf{Q} \cdot \mathbf{R}_n - \omega_{\mathbf{Q}} t + \text{const.}) \end{aligned}$$

Similarly, $S_{n\eta}$ varies as $\sin(\mathbf{Q} \cdot \mathbf{R}_n - \omega_{\mathbf{Q}} t + \text{const.})$.

($p = 2, 4, 6$) and assume it to have the simplest form^{19e}

$$\sum_n \{DS_{nz}^2 - p^{-2}S^{-p+2}G[(S_{nx} + iS_{ny})^p + (S_{nx} - iS_{ny})^p]\}. \quad (14.4)$$

This can be written in a_n^* and a_n as

$$\begin{aligned} \sum_n & \{ -\frac{1}{2}DS(a_n^* - a_n)^2 + GS[(2/p)a_n^*a_n + [(p-1)/2p](a_n^* + a_n)^2] \\ & \cdot [1 - 4p^2\delta^2(1 - \cos 2\mathbf{Q} \cdot \mathbf{R}_n)] \}, \end{aligned} \quad (14.5)$$

and in spin-wave amplitudes, a_q^* and a_q , as

$$\begin{aligned} DS \sum_q & (a_q^*a_q - \frac{1}{2}a_q^*a_{-q}^* - \frac{1}{2}a_qa_{-q}) \\ & + GS \sum_q \{ [(1 + p^{-1})a_q^*a_q + \frac{1}{2}(1 - p^{-1})(a_q^*a_{-q}^* + a_qa_{-q})] (1 - 4p^2\delta^2) \\ & + 2p(p+1)\delta^2(a_{q+Q}^*a_{q-Q} + a_{q-Q}^*a_{q+Q}) \\ & + p(p-1)\delta^2(a_{q+Q}^*a_{-q+Q}^* + a_{q-Q}^*a_{-q-Q}^* + a_{q+Q}a_{-q+Q} + a_{q-Q}a_{-q-Q}) \}. \end{aligned} \quad (14.6)$$

^{19e} From the second term of (14.4) arises an additional anisotropy energy that prevents the spin from deviating from the xy plane. To see it, let us assume that the spin points almost in the x direction. Dropping the suffix n for the sake of brevity, we can write this term as

$$-\sum_{\pm} p^{-2}S^{-p+2}G[S - (S - S_z) \pm iS_y]^p. \quad (a)$$

Expanding this in powers of $(S - S_z)$ and S_y , and writing

$$\begin{aligned} S - S_z &= S - [S(S+1) - S_y^2 - S_z^2]^{1/2} \\ &\doteq (1/2S)(S_y^2 + S_z^2) \end{aligned} \quad (b)$$

under the assumption of large S , we can calculate (a), up to the second order of S_y and S_z , to be

$$GS_y^2 + (1/p)GS_z^2. \quad (c)$$

Thus, the total anisotropy energy has an effective coefficient $D + (1/p)G$ in the z direction and G in the y direction. This situation is also manifested in (14.5). Namely, the second term in the first line of (14.5), which has a factor in the second line arising from the fan distribution, can be written as

$$GS[\frac{1}{2}(a_n^* + a_n)^2 - (1/2p)(a_n^* - a_n)^2], \quad (d)$$

and by (V.1) this can be expressed as

$$GS_{n\xi}^2 + (1/p)GS_{n\eta}^2, \quad (e)$$

where ξ is the same as z in the present case. In Section 5, after Eq. (5.3), we assumed an anisotropy energy of the form $DS_y^2 + GS_z^2$ and calculated spin-wave frequencies for the ferromagnetic alignment in the x -direction. In the present section, $D + (1/p)G$ appears in place of D .

Furthermore, we have the Zeeman energy:

$$-H \sum_n S_{nx} = H \sum_{\mathbf{q}} \{(1 - 4\delta^2) a_{\mathbf{q}}^* a_{\mathbf{q}} + 2\delta^2 (a_{\mathbf{q}+\mathbf{Q}}^* a_{\mathbf{q}-\mathbf{Q}} + a_{\mathbf{q}-\mathbf{Q}}^* a_{\mathbf{q}+\mathbf{Q}})\}, \quad (14.7)$$

where we understand by H the field that includes the static demagnetizing field. The dynamical part of the demagnetizing energy can be written for an ellipsoidal sample, whose principal axes coincide with the coordinate axes, as follows:

$$\begin{aligned} & -SN_x \delta^2 (a_{\mathbf{Q}}^* - a_{\mathbf{Q}} - a_{-\mathbf{Q}}^* + a_{-\mathbf{Q}})^2 + \frac{1}{4} SN_y (1 - 8\delta^2) (a_0^* + a_0)^2 \\ & + SN_y \delta^2 (a_0^* + a_0) (a_{\mathbf{Q}}^* + a_{2\mathbf{Q}} + a_{-2\mathbf{Q}}^* + a_{-2\mathbf{Q}}) \\ & - \frac{1}{4} SN_z (a_0^* - a_0)^2. \end{aligned} \quad (14.8)$$

As before, this part has been considered only for uniform modes. In (14.8), there is a coupling term between mode 0 and modes $\pm 2\mathbf{Q}$, but we shall disregard it, since this off-diagonal term will contribute to the frequency of mode 0 a correction of the order of δ^4 .

The mathematical problem we have to solve is to diagonalize the sum of (14.2), (14.6), (14.7), and (14.8). We neglect all terms $a_{\mathbf{q}}^* a_{\mathbf{q}'}$, $a_{\mathbf{q}}^* a_{-\mathbf{q}'}$, and $a_{\mathbf{q}} a_{-\mathbf{q}'}$ with $\mathbf{q} \neq \mathbf{q}'$, since these off-diagonal terms will also contribute a correction of $O(\delta^4)$ to the spin-wave frequencies. Then, except for $\mathbf{q} = 0$ and $\mathbf{q} = \pm\mathbf{Q}$, the total Hamiltonian can be written in the following form:

$$2S \sum_{\mathbf{q}} [A_{\mathbf{q}} a_{\mathbf{q}}^* a_{\mathbf{q}} + \frac{1}{2} B_{\mathbf{q}} (a_{\mathbf{q}}^* a_{-\mathbf{q}}^* + a_{\mathbf{q}} a_{-\mathbf{q}})], \quad (14.9)$$

where

$$\begin{aligned} A_{\mathbf{q}} = & J(0) - J(\mathbf{q}) + 4\delta^2 [2J(\mathbf{Q}) - 2J(0) + C(\mathbf{q}) - \frac{1}{2}p(p+1)G] \\ & + \frac{1}{2}[D + (1/p)G] + \frac{1}{2}G + (H/2S)(1 - 4\delta^2), \end{aligned} \quad (14.10)$$

$$B_{\mathbf{q}} = -\frac{1}{2}[D + (1/p)G] + \frac{1}{2}G + 4\delta^2 [C(\mathbf{q}) - \frac{1}{2}p(p-1)G]. \quad (14.11)$$

Here $A_{\mathbf{q}} = A_{-\mathbf{q}}$ and $B_{\mathbf{q}} = B_{-\mathbf{q}}$. Using H_0 defined by (9.1), $A_{\mathbf{q}}$ can be written as

$$\begin{aligned} A_{\mathbf{q}} = & J(\mathbf{Q}) - J(\mathbf{q}) + \frac{1}{2}[D + (1/p)G] - \frac{1}{2}G \\ & + 4\delta^2 [J(\mathbf{Q}) - J(0) + C(\mathbf{q}) - \frac{1}{2}(p-1)(p+2)G] - [(H_0 - H)/2S]. \end{aligned} \quad (14.12)$$

The second line of this quantity is proportional to $H_0 - H$, since δ^2 is proportional to $H_0 - H$.

As before, we have

$$\hbar\omega_q = 2S[(A_q + B_q)(A_q - B_q)]^{1/2}. \quad (14.13)$$

This formula is not applicable to the cases of $q = 0$ and $q = \pm Q$ for which we have to take account of demagnetizing effect. The additional terms due to this effect can be obtained from (14.8). For $q = 0$, then, we have

$$\hbar\omega_0 = 2S[(A_0 + B_0)(A_0 - B_0)]^{1/2}, \quad (14.14)$$

with

$$\begin{aligned} A_0 + B_0 &= J(Q) - J(0) - 4\delta^2[J(Q) - J(0) + (p^2 - 1)G] \\ &\quad - (2S)^{-1}(H_0 - H) + \frac{1}{2}N_y(1 - 8\delta^2) \end{aligned} \quad (14.15)$$

and

$$\begin{aligned} A_0 - B_0 &= J(Q) - J(0) + [D + (1/p)G] - G \\ &\quad + 4\delta^2[J(Q) - J(0) - (p - 1)G] \\ &\quad - (2S)^{-1}(H_0 - H) + \frac{1}{2}N_z. \end{aligned} \quad (14.16)$$

It can be seen from the functional forms of (14.14)–(14.16) that ω_0 decreases with increasing $H_0 - H$. The mode of this oscillation is such that the spin vectors describe, in unison, ellipses of the *same size* in space whose principal axes are along ξ and z , as one may expect from the demagnetizing coefficients, N_y and N_z , contained in (14.15) and (14.16). This does not mean that the fan oscillates as a rigid body, in which case the ellipses described by the spin vectors on both edges of the fan will have a smaller semiaxial length in the z -direction than the semiaxial length of the ellipse described by the central spin vectors. Thus, the actual motion of the fan includes a bending oscillation of the fan plane. For this reason, N_z appears fully in (14.16), not with a factor $\langle \cos \varphi_n \rangle$, averaged over n . If one had just one moment vector whose magnitude is equal to the average moment of the fan in the field direction, then one would have had a factor $\langle \cos \varphi_n \rangle$ to both N_z in (14.16) and N_y in (14.15). In actuality, N_y appears with a factor $\langle \cos^2 \varphi_n \rangle$, which is equal to $1 - 8\delta^2$. This is because the fan has a finite angular width, and it oscillates in the direction of φ as if it were a rigid body.

In the case of $q = \pm Q$, we have the spin-wave Hamiltonian:

$$\begin{aligned} 2S[A_Q(a_Q^*a_Q + a_{-Q}^*a_{-Q}) + B_Q(a_Q^*a_{-Q}^* + a_Qa_{-Q}) \\ + C_Q(a_Q^*a_{-Q} + a_{-Q}^*a_Q) \\ + \frac{1}{2}D_Q(a_Q^*a_Q^* + a_{-Q}^*a_{-Q}^* + a_Qa_Q + a_{-Q}a_{-Q})], \end{aligned} \quad (14.17)$$

with

$$\begin{aligned} A_Q &= \frac{1}{2}[D + (1/p)G] - \frac{1}{2}G \\ &\quad + 4\delta^2[2J(Q) - \frac{3}{2}J(0) - \frac{1}{2}J(2Q) - \frac{1}{2}(p-1)(p+2)G] \\ &\quad - (2S)^{-1}(H_0 - H) + N_x\delta^2, \end{aligned} \quad (14.18)$$

$$\begin{aligned} B_Q &= -\frac{1}{2}[D + (1/p)G] + \frac{1}{2}G \\ &\quad + 4\delta^2[J(Q) - \frac{1}{2}J(0) - \frac{1}{2}J(2Q) - \frac{1}{2}p(p-1)G] + N_x\delta^2, \end{aligned} \quad (14.19)$$

$$\begin{aligned} C_Q &= -2\delta^2[2J(Q) - J(0) - J(2Q) - \frac{1}{2}(p-1)(p+2)G] - N_x\delta^2, \\ D_Q &= -2\delta^2[J(Q) - J(0) - \frac{1}{2}p(p-1)G] - N_x\delta^2. \end{aligned} \quad (14.20)$$

$$D_Q = -2\delta^2[J(Q) - J(0) - \frac{1}{2}p(p-1)G] - N_x\delta^2. \quad (14.21)$$

To diagonalize (14.17), we put, with Cooper and Elliott,^{19,19f} as follows:

$$\begin{aligned} p_+ &= \frac{1}{2}(a_Q + a_{-Q}^* + a_{-Q} + a_Q^*) = (2S)^{-1/2}(S_{Q,\xi} + S_{-Q,\xi}), \\ p_- &= \frac{1}{2}(a_Q + a_{-Q}^* - a_{-Q} - a_Q^*) = (2S)^{-1/2}(S_{Q,\xi} - S_{-Q,\xi}), \\ -iq_+ &= \frac{1}{2}(a_Q - a_{-Q}^* + a_{-Q} - a_Q^*) = -i(2S)^{-1/2}(S_{Q,z} + S_{-Q,z}), \\ iq_- &= \frac{1}{2}(a_Q - a_{-Q}^* - a_{-Q} + a_Q^*) = +i(2S)^{-1/2}(S_{Q,z} - S_{-Q,z}). \end{aligned} \quad (14.22)$$

These variables satisfy the commutation rules:

$$[q_+, p_+] = [q_-, p_-] = i, \quad (14.23)$$

$$[p_+, p_-] = [q_+, q_-] = [p_+, q_-] = [p_-, q_+] = 0,$$

so that p_+ , q_+ and p_- , q_- are pairs of canonically conjugate variables. Then, the Hamiltonian can be written as

$$\begin{aligned} 2S[\frac{1}{2}(A_Q + B_Q + C_Q + D_Q)p_+^2 + \frac{1}{2}(A_Q - B_Q + C_Q - D_Q)q_+^2 \\ + \frac{1}{2}(A_Q + B_Q - C_Q - D_Q)p_-^2 + \frac{1}{2}(A_Q - B_Q - C_Q + D_Q)q_-^2], \end{aligned} \quad (14.24)$$

and, correspondingly, the frequencies are found to be

$$\hbar\omega_+ = 2S[(A_Q + B_Q + C_Q + D_Q)(A_Q - B_Q + C_Q - D_Q)]^{1/2}, \quad (14.25)$$

$$\hbar\omega_- = 2S[(A_Q + B_Q - C_Q - D_Q)(A_Q - B_Q - C_Q + D_Q)]^{1/2}. \quad (14.26)$$

^{19f} Cooper and Elliott obtained incorrectly $C_Q/D_Q = -4/2$ for $G = 0$ and $N_x = 0$. Their results (see Cooper and Elliott¹⁹) are thus different from ours.

Here we have

$$\begin{aligned} A_Q + B_Q + C_Q + D_Q \\ = 2\delta^2[3J(Q) - 2J(0) - J(2Q) - (p^2 - 1)G] - (2S)^{-1}(H_0 - H), \end{aligned}$$

which vanishes identically for $H_0 > H$ (thus giving δ^2 explicitly as a function of $H_0 - H$). The mathematical proof of this can be made with a calculation similar to that leading to (10.9) for $p = 6$. However, the vanishing of ω_+ [see (14.25)] can be visualized by looking at the corresponding mode. For ω_+ , we have $S_{Qx} = S_{-Qx}$ and $S_{Qz} = S_{-Qz}$, since $p_+ = q_- = 0$, and furthermore $S_{Qz} = S_{-Qz} = 0$, if the coefficient of p_+^2 in (14.24) vanishes, and hence q_+ must vanish; in the analogy of a classical harmonic oscillator, the vanishing of the coefficient of p_+^2 means an infinite mass, and hence vanishing of the displacement q_+ . Thus, the mode of oscillation is such that the central vectors in the fan structure oscillate in the xy plane with a maximum amplitude, and the vectors at the edges are at rest. This is illustrated in Fig. 5a. This mode may, thus, be called the *cosine mode*. When one sees the spin vectors opposite to the x direction, their tops will be on a sinusoidal curve running along the z direction, as shown by a full curve in Fig. 5b, when there is no oscillation. When the oscillation occurs, this curve will shift to a broken curve shown in the same figure. As may be seen, the oscillation merely shifts the full curve up and down in the z -direc-

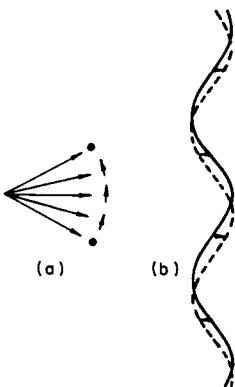


FIG. 5

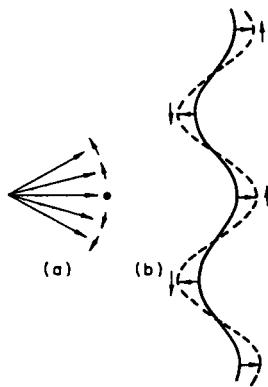


FIG. 6

FIG. 5. Cosine oscillation of the fan vectors. (a) Projection of the vectors on the easy plane. (b) Profile of the mode seen against the field direction, the easy plane being horizontal. The full curve is the locus of the tips of the vectors at rest. This curve moves up and down when the vectors oscillate as shown in (a). The frequency of oscillation is zero.

FIG. 6. Sine oscillation of the fan vectors. (a) and (b) have meanings similar to those of Fig. 5. The full curve in (b) oscillates in amplitude, as shown. The frequency is finite.

tion. No energy change should be associated with this motion, and evidently the frequency must be zero.

For ω_- , we have a *sine mode*, shown in Figs. 6a and 6b. In this case, we have $S_{Q\xi} = -S_{-Q\xi}$ and $S_{Qz} = -S_{-Qz}$, since $p_+ = q_+ = 0$. Correspondingly, the vectors move in the xy plane as shown in Fig. 6a and, with a phase advance of $\pi/2$, also in the z direction. They describe ellipses in space. The ratio between the diameters of the ellipses in the ξ and z directions is

$$(A_Q - B_Q - C_Q + D_Q)^{1/2}/(A_Q + B_Q - C_Q - D_Q)^{1/2}$$

[the square root ratio between the coefficient of q_-^2 and that of p_-^2 in (14.24)]. The profile of the oscillation is as shown in Fig. 6b. The frequency, ω_- , varies as δ , or as $(H_0 - H)^{1/2}$.

In the ferromagnetic range for $H > H_0$, we have just to put $\delta = 0$, leaving $H - H_0$ as it stands. Then, for $\pm Q$, we have

$$\hbar\omega_+ = \hbar\omega_- = \{(H - H_0)[2S(D + (1/p)G - G) + (H - H_0)]\}^{1/2}. \quad (14.27)$$

This vanishes at $H = H_0$.

15. SPIN WAVES IN A HELIX SUBJECTED TO A WEAK FIELD

This case presents a situation so complicated that one becomes almost uninterested in the analysis. No calculation has been reported which includes both the weak applied field and a small anisotropy energy in the plane, although the effect of an applied field only has been studied rather in detail (Cooper and Elliott¹⁹ and Watabe *et al.*^{19a}). We shall not enter into detailed calculations and shall discuss only a few points of interest.

If we assume an anisotropy energy of the form (14.4), or

$$\sum_n \{DS_{nz} - p^{-2}S^{-p+2}G[(S_{nx} + iS_{ny})^p + (S_{nx} - iS_{ny})^p]\},$$

where G is small and D is positive (not necessarily small), and assume a weak H in the x -direction, then the azimuthal angle, φ_n , of the n th spin in equilibrium will be given in the form

$$\varphi_n = \mathbf{Q} \cdot \mathbf{R}_n + \epsilon_1 \sin \mathbf{Q} \cdot \mathbf{R}_n + \epsilon_p \sin p\mathbf{Q} \cdot \mathbf{R}_n. \quad (15.1)$$

ϵ_1 is proportional to H , and ϵ_p is proportional to G . To second order in H and G , the classical total energy (exchange, Zeeman, and anisotropy energies) per atom can easily be calculated to be the following:

$$S^2 \{-J(\mathbf{Q}) + \frac{1}{4}\epsilon_1^2[2J(\mathbf{Q}) - J(\mathbf{0}) - J(2\mathbf{Q})] + \frac{1}{4}\epsilon_p^2[2J(\mathbf{Q}) - J((p-1)\mathbf{Q}) - J((p+1)\mathbf{Q})] + (2S)^{-1}H\epsilon_1 + p^{-1}G\epsilon_p\}. \quad (15.2)$$

Minimizing this expression with respect to ϵ_1 and ϵ_p , we obtain

$$\epsilon_1 = -H/\{S[2J(Q) - J(0) - J(2Q)]\}, \quad (15.3)$$

$$\epsilon_p = -2G/\{p[2J(Q) - J((p-1)Q) - J((p+1)Q)]\}. \quad (15.4)$$

The spin-wave Hamiltonian to second order in H and G can be obtained without difficulty; the Zeeman and anisotropy terms are rather simple, but the exchange terms are fairly complicated. This Hamiltonian involves not only momentum-conserving terms, $a_q^*a_q$, $a_q^*a_{-q}^*$, $a_{-q}a_q$, but also momentum-nonconserving terms such as $a_q^*a_{q'}$, $a_q^*a_{-q'}$, $a_{-q}a_{q'}$ with $q' = q + Q$, $q \pm 2Q$, $q \pm (p-1)Q$, $q \pm (p+1)Q$, $q \pm pQ$, $q \pm 2pQ$. This means that Bragg reflections occur at $q = \pm \frac{1}{2}Q$, $q = \pm Q$, etc., because of the modulation in the helix, of the form of (15.1), caused by the external and anisotropy fields. The spin-wave frequencies are drastically modified at and near these Bragg points in the q -space. Not only near the Bragg points but also at points far apart from them, the frequency formula cannot be obtained in a simple way, because, although the coefficients of the momentum-conserving terms depend only linearly on H^2 and G^2 , those of the nonconserving terms contain linear (as well as quadratic) forms of H and G , so that one has to include the nonconserving terms in the second-order perturbation. Thus, the spin-wave frequency generally depends quadratically on H and G in a very complicated way.

Like the $q = 0$ mode in the case of a conical arrangement and the cosine mode with $q = \pm Q$ in a fan arrangement, here we have also a mode with zero frequency, if the period of the spin pattern is irrational in units of the lattice spacing along the helix. The reason is similar to that mentioned in Chapter V, Section 13 in connection with the $q = 0$ mode and in Chapter V, Section 14 in connection with the cosine mode. Another way of reasoning the existence of a zero frequency mode in any case of an irrational period is the following.^{19a} The spin pattern in the existence of an applied field and an anisotropy energy can be pictured by specifying the direction of the spin vector at R_n as a function of $Q \cdot R_n$ with a period of 2π . Since the period of the spin pattern was assumed to be irrational, $Q \cdot R_n$ covers uniformly the whole angular range of 0 to 2π when R_n runs over all the atoms in the infinite crystal (which should have a uniform, infinite cross section perpendicular to the direction of Q). In such a case, the choice of the origin of R_n 's, whether on a net plane or between net planes, is immaterial for the spin pattern; the same function of $Q \cdot R_n$ with different choices of the origin gives spin patterns that are translated along Q with

^{19a} Elliott and Lange^{19b} give a mathematical proof of the existence of a zero frequency and the continuity of the frequency spectrum near zero.

^{19b} R. J. Elliott and R. V. Lange, *Phys. Rev.* **152**, 235 (1966).

respect to each other. In other words, we can add an arbitrary phase constant, α , to $\mathbf{Q} \cdot \mathbf{R}_n$. A change in α just shifts the spin pattern in space, not always a small shift even if this change is small. However, no energy change should accompany it. The spin wave mode corresponding to a small change in α has, therefore, zero frequency. It may be noted that this spin-wave mode does not always correspond to a single \mathbf{q} -value, namely, 0 or \mathbf{Q} , but it consists generally of many harmonics.

VI. Complex Spin Configurations

So far, we have confined ourselves to a simple helical spin arrangement and its modifications caused by anisotropy energy and an external magnetic field. It may be recalled that we have assumed lattices in which there is only one magnetic atom per unit cell. If we now have lattices whose unit cell contains several magnetic atoms, we may expect more complex spin arrangements. Yafet and Kittel²⁰ predicted for the spinel structure a *triangular* spin arrangement, which was subsequently observed by a neutron diffraction experiment in CuCr_2O_4 .²¹ Kaplan *et al.*^{22a-f} predicted, in 1961, for the same crystalline structure a *ferrimagnetic multiple cone* spin arrangement, which was then observed in MnCr_2O_4 .²³ We shall study such problems in this and the next parts. Our approach will be somewhat different from that followed by the authors mentioned, although it will depend upon their treatment. Throughout (except in Part VIII), we shall confine ourselves to absolute zero.^{23a}

Let $\mathbf{S}_{n\nu}$ be the spin vector of the ν th atom in the n th unit cell and $\mathbf{R}_{n\nu} = \mathbf{R}_n + \mathbf{R}_\nu$ its position, where we understand by \mathbf{R}_n a translational lattice vector. The exchange energy will be written as

$$E = - \sum_{m,\mu} \sum_{n,\nu} J_{m\mu,n\nu} \mathbf{S}_{m\mu} \cdot \mathbf{S}_{n\nu}. \quad (\text{VI.1})$$

²⁰ Y. Yafet and C. Kittel, *Phys. Rev.* **87**, 290 (1952).

²¹ E. Prince, *Acta Cryst.* **10**, 554 (1957); R. Nathans, S. J. Pickart, and A. Miller, *Bull. Am. Phys. Soc. [2]* **6**, 54 (1961).

^{22a} T. A. Kaplan, *Phys. Rev.* **119**, 1460 (1960).

^{22b} D. H. Lyons and T. A. Kaplan, *Phys. Rev.* **120**, 1580 (1960).

^{22c} T. A. Kaplan, K. Dwight, D. H. Lyons, and N. Menyuk, *J. Appl. Phys.* **32**, 13S (1961).

^{22d} D. H. Lyons, T. A. Kaplan, K. Dwight, and N. Menyuk, *Phys. Rev.* **126**, 540 (1962).

^{22e} N. Menyuk, K. Dwight, D. Lyons, and T. A. Kaplan, *Phys. Rev.* **127**, 1983 (1962).

^{22f} T. A. Kaplan, H. E. Stanley, K. Dwight, and N. Menyuk, *J. Appl. Phys.* **36**, 1129 (1965).

²³ J. M. Hastings and L. M. Corliss, *Phys. Rev.* **126**, 556 (1962).

^{23a} A concise presentation of Parts VI and VII, together with a few other materials, will be published by T. Nagamiya in *J. Appl. Phys. Suppl.* (1968) (*Proc. Intern. Congr. Magnetism, Boston, 1967*).

We make Fourier transformations of the spins:

$$\mathbf{S}_{n\nu} = S_\nu \sum_{\mathbf{q}} \delta_{\mathbf{q}\nu} \exp(i\mathbf{q} \cdot \mathbf{R}_n) \quad (\delta_{-\mathbf{q}\nu} = \delta_{\mathbf{q}\nu}^*, \nu = 1, 2, \dots, k). \quad (\text{VI.2})$$

Here k is the number of atoms per unit cell. Then (VI.1) can be written

$$E = - \sum_{\mathbf{q}} \sum_{m,\mu} \sum_{n,\nu} S_\mu S_\nu J_{m\mu,n\nu} \exp(i\mathbf{q} \cdot \mathbf{R}_{mn}) \delta_{\mathbf{q}\mu} \cdot \delta_{\mathbf{q}\nu}^*, \quad (\text{VI.3})$$

where $\mathbf{R}_{mn} = \mathbf{R}_m - \mathbf{R}_n$. This can be written also as

$$E/N = \sum_{\mathbf{q}} \sum_{\mu} \sum_{\nu} A_{\mu\nu}(\mathbf{q}) \delta_{\mathbf{q}\mu} \cdot \delta_{\mathbf{q}\nu}^*, \quad (\text{VI.4})$$

where N is the number of unit cells and

$$\begin{aligned} A_{\mu\nu}(\mathbf{q}) &= -S_\mu S_\nu \sum_m J_{m\mu,n\nu} \exp(i\mathbf{q} \cdot \mathbf{R}_{mn}) \\ &= A_{\mu\nu}^*(-\mathbf{q}) = A_{\nu\mu}(-\mathbf{q}). \end{aligned} \quad (\text{VI.5})$$

We want to minimize (VI.4) under the condition that $\mathbf{S}_{n\nu}^2 = S_\nu^2$ for all n , which can be written as

$$\left[\sum_{\mathbf{q}} \delta_{\mathbf{q}\nu} \exp(i\mathbf{q} \cdot \mathbf{R}_n) \right]^2 = 1 \quad \text{for all } n \text{ and } \nu. \quad (\text{VI.6})$$

We shall study separately a number of cases.

16. CASE 1: THERE IS A SINGLE \mathbf{q} THAT IS EQUIVALENT TO $-\mathbf{q}$

Such a \mathbf{q} is either zero or equal to half a reciprocal lattice vector; in other words, \mathbf{q} is at the origin of the reciprocal lattice space or at a symmetry point on the Brillouin zone boundary. If such a \mathbf{q} minimizes the energy, then the problem is simplified to

$$E/N = \sum_{\mu} \sum_{\nu} A_{\mu\nu}(\mathbf{q}) \delta_{\mathbf{q}\mu} \cdot \delta_{\mathbf{q}\nu}^* = \min, \quad (16.1)$$

$$\delta_{\mathbf{q}\nu}^2 = 1 \quad \text{for all } \nu. \quad (16.2)$$

Here all quantities are real. If there are two atoms per unit cell, one will have

$$E/N = A_{11}(\mathbf{q}) + A_{22}(\mathbf{q}) + 2A_{12}(\mathbf{q}) \delta_{\mathbf{q}1} \cdot \delta_{\mathbf{q}2} = \min,$$

from which will follow that $\delta_{\mathbf{q}1}$ and $\delta_{\mathbf{q}2}$ are parallel or antiparallel according as $A_{12}(\mathbf{q})$ is negative or positive. The \mathbf{q} -vector must be such that it corresponds to the lowest value of $A_{11}(\mathbf{q}) + A_{22}(\mathbf{q}) - 2|A_{12}(\mathbf{q})|$. If $\mathbf{q} = 0$, then $\mathbf{S}_{n\nu} = S_\nu \delta_{\mathbf{q}\nu}$ ($\nu = 1, 2$), so that one has a ferromagnetic or ferrimagnetic (antiferromagnetic when $S_1 = S_2$) spin arrangement according

as $A_{12}(\mathbf{q})$ is negative or positive. When \mathbf{q} is half a reciprocal lattice vector, one has $\exp(i\mathbf{q}\cdot\mathbf{R}_n) = +1$ or -1 according as $\mathbf{q}\cdot\mathbf{R}_n$ is an even multiple of π or an odd multiple of π , so that both the two sublattices are anti-ferromagnetic.

When there are three atoms per unit cell, we can show that the three vectors $\delta_{\mathbf{q}1}, \delta_{\mathbf{q}2}, \delta_{\mathbf{q}3}$ must be coplanar for the minimum of the energy. If, in particular, the three sublattices are symmetrically related in such a way that $A_{23}(\mathbf{q}) = A_{31}(\mathbf{q}) = A_{12}(\mathbf{q})$, the three vectors are determined from

$$A_{23}(\mathbf{q})(\delta_{\mathbf{q}2}\cdot\delta_{\mathbf{q}3} + \delta_{\mathbf{q}3}\cdot\delta_{\mathbf{q}1} + \delta_{\mathbf{q}1}\cdot\delta_{\mathbf{q}2}) = \min,$$

which is equivalent to

$$A_{23}(\mathbf{q})(\delta_{\mathbf{q}1} + \delta_{\mathbf{q}2} + \delta_{\mathbf{q}3})^2 = \min.$$

Thus, when $A_{23}(\mathbf{q})$ is negative the three vectors are parallel, and when $A_{23}(\mathbf{q})$ is positive, they add up to zero. In the case of $\mathbf{q} = 0$, one has a ferromagnetic or a *triangular* spin arrangement corresponding to the negative or positive sign of $A_{23}(\mathbf{q})$. In the case of \mathbf{q} equal to half a basic reciprocal lattice vector, one has three antiferromagnetic sublattices or an *antiferromagnetic triangular* arrangement.

17. CASE 2: THERE IS A SINGLE \mathbf{q} THAT IS NOT EQUIVALENT TO $-\mathbf{q}$

In this case, we have the following problem:

$$E/N = \sum_{\mu} \sum_{\nu} [A_{\mu\nu}(\mathbf{q}) \delta_{\mathbf{q}\mu} \cdot \delta_{\mathbf{q}\nu}^* + \text{complex conjugate}] = \min, \quad (17.1)$$

$$[\delta_{\mathbf{q}\nu} \exp(i\mathbf{q}\cdot\mathbf{R}_n) + \delta_{\mathbf{q}\nu}^* \exp(-i\mathbf{q}\cdot\mathbf{R}_n)]^2 = 1. \quad (17.2)$$

The complex vector $\delta_{\mathbf{q}\mu}$ may be written as a combination of two real vectors, $\mathbf{u}_{\mathbf{q}\nu}$ and $\mathbf{v}_{\mathbf{q}\nu}$:

$$\delta_{\mathbf{q}\nu} = \frac{1}{2}(\mathbf{u}_{\mathbf{q}\nu} - i\mathbf{v}_{\mathbf{q}\nu}). \quad (17.3)$$

Then we can write the condition (17.2) as

$$\begin{aligned} & [\mathbf{u}_{\mathbf{q}\nu} \cos \mathbf{q}\cdot\mathbf{R}_n + \mathbf{v}_{\mathbf{q}\nu} \sin \mathbf{q}\cdot\mathbf{R}_n]^2 \\ &= \frac{1}{2}\mathbf{u}_{\mathbf{q}\nu}^2(1 + \cos 2\mathbf{q}\cdot\mathbf{R}_n) + \frac{1}{2}\mathbf{v}_{\mathbf{q}\nu}^2(1 - \cos 2\mathbf{q}\cdot\mathbf{R}_n) + \mathbf{u}_{\mathbf{q}\nu} \cdot \mathbf{v}_{\mathbf{q}\nu} \sin 2\mathbf{q}\cdot\mathbf{R}_n = 1. \end{aligned}$$

From this equation follows that $\mathbf{u}_{\mathbf{q}\nu}$ and $\mathbf{v}_{\mathbf{q}\nu}$ are orthogonal unit vectors, except when $2\mathbf{q}\cdot\mathbf{R}_n$ is an integral multiple of π , namely, \mathbf{q} is a quarter of a basic reciprocal lattice vector ($\mathbf{q} = \mathbf{K}/4$). In this exceptional case, $\mathbf{u}_{\mathbf{q}\nu}$ and $\mathbf{v}_{\mathbf{q}\nu}$ need not be orthogonal, since $\sin 2\mathbf{q}\cdot\mathbf{R}_n = 0$, but they have to be unit vectors. Since

$$\mathbf{S}_{\mathbf{n}\nu} = S_{\nu}(\mathbf{u}_{\mathbf{q}\nu} \cos \mathbf{q}\cdot\mathbf{R}_n + \mathbf{v}_{\mathbf{q}\nu} \sin \mathbf{q}\cdot\mathbf{R}_n), \quad (17.4)$$

each sublattice has a helical spin arrangement of wave vector \mathbf{q} . In the

case of $\mathbf{q} = \mathbf{K}/4$, it can be seen from this equation that each sublattice splits into two uncorrelated antiferromagnetic sublattices having $\mathbf{u}_{\mathbf{q}\nu}$ and $\mathbf{v}_{\mathbf{q}\nu}$ as magnetization axes.

The energy (17.1) can be written as

$$\begin{aligned} E/N &= \sum_{\mu} \sum_{\nu} \frac{1}{4} [A_{\mu\nu}(\mathbf{q}) (\mathbf{u}_{\mathbf{q}\mu} \cdot \mathbf{u}_{\mathbf{q}\nu} + \mathbf{v}_{\mathbf{q}\mu} \cdot \mathbf{v}_{\mathbf{q}\nu} - i\mathbf{v}_{\mathbf{q}\mu} \cdot \mathbf{u}_{\mathbf{q}\nu} + i\mathbf{u}_{\mathbf{q}\mu} \cdot \mathbf{v}_{\mathbf{q}\nu}) \\ &\quad + \text{complex conjugate}] \\ &= \sum_{\nu} A_{\nu\nu}(\mathbf{q}) + \sum_{\mu < \nu} [\text{Re } A_{\mu\nu}(\mathbf{q}) (\mathbf{u}_{\mathbf{q}\mu} \cdot \mathbf{u}_{\mathbf{q}\nu} + \mathbf{v}_{\mathbf{q}\mu} \cdot \mathbf{v}_{\mathbf{q}\nu}) \\ &\quad + \text{Im } A_{\mu\nu}(\mathbf{q}) (\mathbf{v}_{\mathbf{q}\mu} \cdot \mathbf{u}_{\mathbf{q}\nu} - \mathbf{u}_{\mathbf{q}\mu} \cdot \mathbf{v}_{\mathbf{q}\nu})]. \end{aligned} \quad (17.5)$$

It is not easy to solve generally the problem of minimizing this energy. However, in the case of two atoms per unit cell, it can be shown easily that the two sets of orthogonal unit vectors, $\mathbf{u}_{\mathbf{q}1}$, $\mathbf{v}_{\mathbf{q}1}$ and $\mathbf{u}_{\mathbf{q}2}$, $\mathbf{v}_{\mathbf{q}2}$, lie in the same plane, and the sense of rotation in going from $\mathbf{u}_{\mathbf{q}1}$ to $\mathbf{v}_{\mathbf{q}1}$ is the same as that from $\mathbf{u}_{\mathbf{q}2}$ to $\mathbf{v}_{\mathbf{q}2}$, that is, the helical rotation has the same sense in the two sublattices. Actually, as we shall see in Section 19, these facts are valid in a general lattice. The relative phase angle, ϕ_{12} , between the two helices in the case of two sublattices is determined from

$\text{Re } A_{12}(\mathbf{q}) \cos \phi_{12} + \text{Im } A_{12}(\mathbf{q}) \sin \phi_{12} = \min$,
namely, as

$$\phi_{12} = \alpha + \pi, \quad \text{where } A_{12}(\mathbf{q}) = |A_{12}(\mathbf{q})| \exp(i\alpha). \quad (17.6)$$

The corresponding energy is

$$E/N = A_{11}(\mathbf{q}) + A_{22}(\mathbf{q}) - 2 |A_{12}(\mathbf{q})|. \quad (17.7)$$

The value of \mathbf{q} corresponds to the lowest value of this energy. The value of \mathbf{q} in case 1 also corresponded to the lowest value of the same expression.

When \mathbf{q} is a quarter of a reciprocal lattice vector ($\mathbf{q} = \mathbf{K}/4$), the foregoing argument fails partly. In this case, each of the two sublattices splits into two uncorrelated antiferromagnetic sublattices, as mentioned before. $\mathbf{u}_{\mathbf{q}1}$ and $\mathbf{v}_{\mathbf{q}1}$ point arbitrarily, but $\mathbf{u}_{\mathbf{q}2}$ and $\mathbf{v}_{\mathbf{q}2}$ are related to $\mathbf{u}_{\mathbf{q}1}$ and $\mathbf{v}_{\mathbf{q}1}$. If $|\text{Re } A_{12}(\mathbf{q})| > |\text{Im } A_{12}(\mathbf{q})|$, then $\mathbf{u}_{\mathbf{q}2}$ and $\mathbf{v}_{\mathbf{q}2}$ are parallel and anti-parallel to $\mathbf{u}_{\mathbf{q}1}$ and $\mathbf{v}_{\mathbf{q}1}$, respectively, according as $\text{Re } A_{12}(\mathbf{q})$ is negative or positive; if $|\text{Re } A_{12}(\mathbf{q})| < |\text{Im } A_{12}(\mathbf{q})|$, $\mathbf{u}_{\mathbf{q}2}$ is parallel to $\mathbf{v}_{\mathbf{q}1}$ and $\mathbf{v}_{\mathbf{q}2}$ anti-parallel to $\mathbf{u}_{\mathbf{q}1}$ when $\text{Im } A_{12}(\mathbf{q})$ is negative, whereas antiparallel and parallel are reversed when $\text{Im } A_{12}(\mathbf{q})$ is positive.

In the case where there are three sublattices symmetrically related to each other, so that $A_{23}(\mathbf{q}) = A_{31}(\mathbf{q}) = A_{12}(\mathbf{q})$ and $A_{ij}(\mathbf{q}) = A_{ji}(\mathbf{q})$ hold,

we have a problem:

$$A_{23}(\mathbf{q}) (\delta_{q2} \cdot \delta_{q3}^* + \delta_{q3} \cdot \delta_{q1}^* + \delta_{q1} \cdot \delta_{q2}^*) + \text{c. c.} = \min,$$

or

$$A_{23}(\mathbf{q}) |\delta_{q1} + \delta_{q2} + \delta_{q3}|^2 = \min.$$

It follows that, if $A_{23}(\mathbf{q})$ is negative, δ_{q1} , δ_{q2} , δ_{q3} point parallel to each other, so that there is a triple helical spin arrangement. If $A_{23}(\mathbf{q})$ is positive, $\delta_{q1} + \delta_{q2} + \delta_{q3}$ must vanish, so that these three vectors form a triangle; the corresponding spin arrangement is such that this triangle rotates along the direction of \mathbf{q} . An example of such a triangular helix has been observed in Mn_3Sn at low temperatures by neutron diffraction.²⁴ This crystal is hexagonal, and the \mathbf{q} vector points along the hexagonal axis. At temperatures above 270°K , the spin arrangement is triangular with $\mathbf{q} = 0$, the transition from a finite \mathbf{q} to $\mathbf{q} = 0$ being abrupt. In an isomorphous crystal, Mn_3Ge , and a cubic crystal, Mn_3Rh , a triangular spin arrangement with $\mathbf{q} = 0$ has been observed in the temperature range of spin ordering (below 77°K in Mn_3Ge and below 600°K in Mn_3Rh). Σ

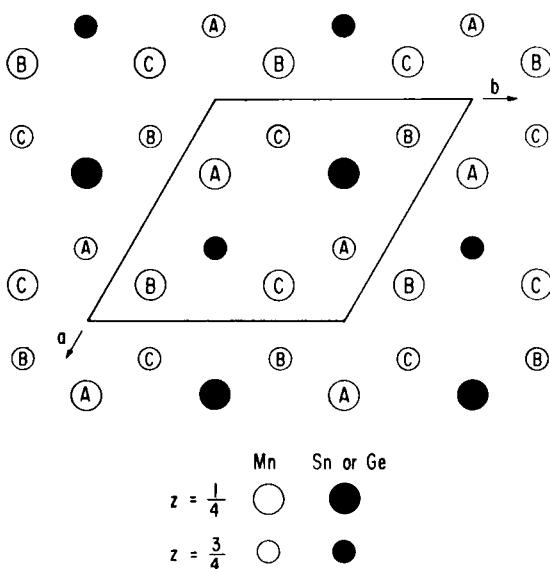


FIG. 7. Basal plane projection of the crystal structure of Mn_3Sn and Mn_3Ge . Magnetic sublattices are labeled A , B , C . Moments on them form an equilateral triangle whose plane is perpendicular to the plane of the figure. In Mn_3Sn , this triangle rotates as the position advances in the direction perpendicular to the plane of the figure (after Kouvel and Kasper²⁴).

²⁴ J. S. Kouvel and J. S. Kasper, *Proc. Intern. Conf. Magnetism, Nottingham, 1964*, p. 169. Inst. Phys. Phys. Soc., London, 1965.

The value of \mathbf{q} corresponds to the lowest value of the energy, which is
 $E/N = A_{11}(\mathbf{q}) + A_{22}(\mathbf{q}) + A_{33}(\mathbf{q}) + 6A_{23}(\mathbf{q}) \quad \text{when } A_{23}(\mathbf{q}) < 0,$
 $E/N = A_{11}(\mathbf{q}) + A_{22}(\mathbf{q}) + A_{33}(\mathbf{q}) - 3A_{23}(\mathbf{q}) \quad \text{when } A_{23}(\mathbf{q}) > 0.$

18. CASE 3: THERE ARE TWO WAVE VECTORS, \mathbf{q} AND \mathbf{q}'

If we assume two wave vectors, \mathbf{q} and \mathbf{q}' , to coexist in the spin arrangement, condition (VI.6) becomes

$$[\delta_{\mathbf{q},\nu} \exp(i\mathbf{q}\cdot\mathbf{R}_n) + \delta_{\mathbf{q}',\nu} \exp(i\mathbf{q}'\cdot\mathbf{R}_n) + \text{c. c.}]^2 = 1.$$

Writing $\delta_{\mathbf{q},\nu}$ and $\delta_{\mathbf{q}',\nu}$ in the form of (17.3), we can rewrite this equation as
 $[\mathbf{u}_{\mathbf{q},\nu} \cos \mathbf{q} \cdot \mathbf{R}_n + \mathbf{v}_{\mathbf{q},\nu} \sin \mathbf{q} \cdot \mathbf{R}_n + \mathbf{u}_{\mathbf{q}',\nu} \cos \mathbf{q}' \cdot \mathbf{R}_n + \mathbf{v}_{\mathbf{q}',\nu} \sin \mathbf{q}' \cdot \mathbf{R}_n]^2$

$$\begin{aligned} &= \frac{1}{2}\mathbf{u}_{\mathbf{q},\nu}^2(1 + \cos 2\mathbf{q} \cdot \mathbf{R}_n) + \frac{1}{2}\mathbf{v}_{\mathbf{q},\nu}^2(1 - \cos 2\mathbf{q} \cdot \mathbf{R}_n) + \mathbf{u}_{\mathbf{q},\nu} \cdot \mathbf{v}_{\mathbf{q},\nu} \sin 2\mathbf{q} \cdot \mathbf{R}_n \\ &\quad + \frac{1}{2}\mathbf{u}_{\mathbf{q}',\nu}^2(1 + \cos 2\mathbf{q}' \cdot \mathbf{R}_n) + \frac{1}{2}\mathbf{v}_{\mathbf{q}',\nu}^2(1 - \cos 2\mathbf{q}' \cdot \mathbf{R}_n) \\ &\quad + \mathbf{u}_{\mathbf{q},\nu} \cdot \mathbf{v}_{\mathbf{q},\nu} \sin 2\mathbf{q}' \cdot \mathbf{R}_n + \mathbf{u}_{\mathbf{q},\nu} \cdot \mathbf{u}_{\mathbf{q}',\nu} [\cos(\mathbf{q} - \mathbf{q}') \cdot \mathbf{R}_n \\ &\quad + \cos(\mathbf{q} + \mathbf{q}') \cdot \mathbf{R}_n] + \mathbf{v}_{\mathbf{q},\nu} \cdot \mathbf{v}_{\mathbf{q}',\nu} [\cos(\mathbf{q} - \mathbf{q}') \cdot \mathbf{R}_n - \cos(\mathbf{q} + \mathbf{q}') \cdot \mathbf{R}_n] \\ &\quad + \mathbf{u}_{\mathbf{q},\nu} \cdot \mathbf{v}_{\mathbf{q}',\nu} [-\sin(\mathbf{q} - \mathbf{q}') \cdot \mathbf{R}_n + \sin(\mathbf{q} + \mathbf{q}') \cdot \mathbf{R}_n] \\ &\quad + \mathbf{v}_{\mathbf{q},\nu} \cdot \mathbf{u}_{\mathbf{q}',\nu} [\sin(\mathbf{q} - \mathbf{q}') \cdot \mathbf{R}_n + \sin(\mathbf{q} + \mathbf{q}') \cdot \mathbf{R}_n] = 1. \end{aligned} \quad (18.1)$$

In order that this equation be satisfied for all \mathbf{R}_n when all the sines and cosines are different, the two real vectors $\mathbf{u}_{\mathbf{q},\nu}$ and $\mathbf{v}_{\mathbf{q},\nu}$ must be orthogonal and have the same length; also, $\mathbf{u}_{\mathbf{q}',\nu}$ and $\mathbf{v}_{\mathbf{q}',\nu}$ must be orthogonal and have the same length, and, as can be seen from the last four terms of the equation, $\mathbf{u}_{\mathbf{q},\nu}$ and $\mathbf{v}_{\mathbf{q},\nu}$ must be orthogonal to both $\mathbf{u}_{\mathbf{q}',\nu}$ and $\mathbf{v}_{\mathbf{q}',\nu}$, which is impossible. We are thus led to a restriction to be imposed on \mathbf{q} and \mathbf{q}' . This restriction is such that there must be equivalent vectors among the nine vectors $0, \pm 2\mathbf{q}, \pm 2\mathbf{q}', \pm(\mathbf{q} - \mathbf{q}'),$ and $\pm(\mathbf{q} + \mathbf{q}')$. If we assume \mathbf{q} to be a general vector, we have the possibilities in the accompanying table.

	0	$2\mathbf{q}$	$2\mathbf{q}'$	$\mathbf{q} - \mathbf{q}'$	$\mathbf{q} + \mathbf{q}'$
(a) $\mathbf{q}' \equiv 0$	0	$2\mathbf{q}$	0	\mathbf{q}	\mathbf{q}
(b) $\mathbf{q}' \equiv -\mathbf{q}' (= \frac{1}{2}\mathbf{K})$	0	$2\mathbf{q}$	0	$\mathbf{q} - \frac{1}{2}\mathbf{K}$	$\mathbf{q} - \frac{1}{2}\mathbf{K}$
(c) $2\mathbf{q}' \equiv -2\mathbf{q}' (\mathbf{q}' = \frac{1}{2}\mathbf{K})$	0	$2\mathbf{q}$	$\frac{1}{2}\mathbf{K}$	$\mathbf{q} - \frac{1}{2}\mathbf{K}$	$\mathbf{q} + \frac{1}{2}\mathbf{K}$
(d) $2\mathbf{q}' \equiv -2\mathbf{q} (\mathbf{q}' = -\mathbf{q} + \frac{1}{2}\mathbf{K})$	0	$2\mathbf{q}$	$-2\mathbf{q}$	$2\mathbf{q} - \frac{1}{2}\mathbf{K}$	$\frac{1}{2}\mathbf{K}$
(e) $\mathbf{q}' \equiv 3\mathbf{q}$ or $\mathbf{q}' \equiv -3\mathbf{q}$	0	$2\mathbf{q}$	$\pm 6\mathbf{q}$	$-2\mathbf{q}$ or $4\mathbf{q}$	$4\mathbf{q}$ or $-2\mathbf{q}$

Here \mathbf{K} is a basic reciprocal lattice vector; in the table, on the right, are listed the corresponding five vectors 0 , $2\mathbf{q}$, $2\mathbf{q}'$, $\mathbf{q} - \mathbf{q}'$, and $\mathbf{q} + \mathbf{q}'$.

In cases a and b, $\delta_{\mathbf{q}'}$, is a real vector; it will be written as \mathbf{u}' in the following. By (18.1), it must be orthogonal to both \mathbf{u}_v and \mathbf{v}_v (the subscript \mathbf{q} being omitted for brevity), whereas \mathbf{u}_v and \mathbf{v}_v are orthogonal to each other and have the same length. It follows further from the same equation that

$$\mathbf{u}_v^2 + \mathbf{u}'_v^2 = 1. \quad (18.2)$$

If \mathbf{u}_v and \mathbf{u}'_v are both nonvanishing, the spin arrangement in the v th sublattice is such that the \mathbf{q} -component and \mathbf{q}' -component of the spin at \mathbf{R}_n are, respectively, expressed by

$$S_\mu u_v (\mathbf{i} \cos \mathbf{q} \cdot \mathbf{R}_n + \mathbf{j} \sin \mathbf{q} \cdot \mathbf{R}_n), \quad (18.3)$$

$$S_\nu u'_v \mathbf{k} \exp(i\mathbf{q}' \cdot \mathbf{R}_n), \quad (18.4)$$

where u_v and u'_v are now scalars and \mathbf{i} , \mathbf{j} , and \mathbf{k} are unit orthogonal vectors. Hence, in case a, where $\mathbf{q}' = 0$, the spin vectors rotate on a circular cone with a wave vector \mathbf{q} , and in case b, where $\mathbf{q}' = \mathbf{K}/2$, they rotate alternately on an up-cone and a down-cone. The spin arrangement in the whole lattice in such cases may be called a *multiple cone structure*. Kaplan's ferrimagnetic cone structure (ferrimagnetic spiral, as he calls it) is an example of this.

In case c, \mathbf{u}'_v and \mathbf{v}'_v need not be orthogonal to each other, since $\sin 2\mathbf{q}' \cdot \mathbf{R}_n = 0$ [see (18.1)], but they must have the same magnitude and be orthogonal to both \mathbf{u}_v and \mathbf{v}_v , so that they are parallel to each other. Hence we may put

$$\delta_{\mathbf{q}'_v} = \frac{1}{2}(1 - i)u'_v \mathbf{k} \quad \text{or} \quad \frac{1}{2}(1 + i)u'_v \mathbf{k}, \quad (18.5)$$

where u'_v is a real number. Equation (18.2) must hold in this case, too. The \mathbf{q}' -component of the spin vector at \mathbf{R}_n is

$$\begin{aligned} S_\nu [\frac{1}{2}(1 \mp i)u'_v \exp(i\mathbf{q}' \cdot \mathbf{R}_n) + \text{c. c.}] \\ = S_\nu u'_v (\cos \mathbf{q}' \cdot \mathbf{R}_n \pm \sin \mathbf{q}' \cdot \mathbf{R}_n), \end{aligned} \quad (18.6)$$

which varies as $(++--)$ for the upper sign and $(+-+-)$ for the lower sign for $\mathbf{q}' \cdot \mathbf{R}_n = 0, \pi/2, \pi, 3\pi/2 \pmod{2\pi}$. Thus, the spin vectors rotate on two opposite cones, alternating in every two steps.

In case d, we have a double helix, one helix running over those sites for which $\mathbf{K} \cdot \mathbf{R}_n$ is an even multiple of 2π and the other over those of odd multiples. They have a common \mathbf{q} but are uncorrelated. Case e corresponds to a mixing of the third harmonic. We shall not be concerned with these cases.

Now we consider the interaction between different sublattices. To simplify the problem, we shall henceforth be concerned only with the

case of two sublattices. The total energy is the sum of (17.5) for $\mathbf{q} = \mathbf{q}$ and $\mathbf{q} = \mathbf{q}'$. The \mathbf{q} -part can be treated as in case 2, and we know that the rotation in sublattice 1 has the same axis and the same sense as in sublattice 2. As for the \mathbf{q}' -part, we can see that, in cases a and b, $\mathbf{u}_1' \cdot \mathbf{u}_2'$ appearing in (17.5) is replaced by $2\mathbf{u}_1' \cdot \mathbf{u}_2'$, since \mathbf{v}_1' and \mathbf{v}_2' do not appear and $A_{12}(\mathbf{q}')$ is real; in case c, \mathbf{v}_1' and \mathbf{v}_2' are either parallel or antiparallel to \mathbf{u}_1' and \mathbf{u}_2' , respectively, as can be seen from (18.5). Using scalars u_1 , u_2 and u_1' , u_2' defined by (18.3)–(18.5), we can write the energy as

$$\begin{aligned} E/N = & A_{11}(\mathbf{q})u_1^2 + A_{22}(\mathbf{q})u_2^2 - 2|A_{12}(\mathbf{q})|u_1u_2 + A_{11}(\mathbf{q}')u_1'^2 \\ & + A_{22}(\mathbf{q}')u_2'^2 - 2|\operatorname{Re} \text{ or } \operatorname{Im} A_{12}(\mathbf{q}')|u_1'u_2'. \end{aligned} \quad (18.7)$$

Here u_1 , u_2 and u_1' , u_2' satisfy

$$u_1^2 + u_1'^2 = 1, \quad u_2^2 + u_2'^2 = 1. \quad (18.8)$$

The choice of Re or Im in the last term of (18.7) is made as follows: Re in cases a and b, where $A_{12}(\mathbf{q}')$ itself is real; Re in case c if the first (or second) choice in (18.5) is made for both sublattices; Im if opposite choices are made for the two sublattices.

The spin configuration that results from the minimization of the energy (18.7) is a double cone configuration. By (18.8), we can put

$$u_1 = \cos \phi_1, \quad u_1' = \sin \phi_1, \quad u_2 = \cos \phi_2, \quad u_2' = \sin \phi_2. \quad (18.9)$$

Then, abbreviating $A_{11}(\mathbf{q}) - A_{11}(\mathbf{q}') = a_1$, $A_{22}(\mathbf{q}) - A_{22}(\mathbf{q}') = a_2$, $|A_{12}(\mathbf{q})| = b$, and $|\operatorname{Re} \text{ or } \operatorname{Im} A_{12}(\mathbf{q}')| = b'$, we can write the equations to determine ϕ_1 and ϕ_2 as

$$\begin{aligned} a_1 \sin 2\phi_1 - 2b \sin \phi_1 \cos \phi_2 + 2b' \cos \phi_1 \sin \phi_2 &= 0, \\ a_2 \sin 2\phi_2 - 2b \cos \phi_1 \sin \phi_2 + 2b' \sin \phi_1 \cos \phi_2 &= 0. \end{aligned}$$

Denoting

$$\cos \phi_2 / \cos \phi_1 = c, \quad \sin \phi_2 / \sin \phi_1 = s, \quad (18.10)$$

we have

$$a_1 - bc + b's = 0, \quad a_2 - b/c + b'/s = 0,$$

so that

$$c = (ba_1 + b'a_2cs)/(b^2 - b'^2), \quad s = (b'a_1 + ba_2cs)/(b^2 - b'^2). \quad (18.11)$$

Multiplying these equations side by side, we obtain a quadratic equation for cs . Solving it and substituting the result into (18.11), we obtain

$$\begin{aligned} c &= (1/2a_2b)\{b^2 - b'^2 + a_1a_2 \pm [((b + b')^2 - a_1a_2)((b - b')^2 - a_1a_2)]^{1/2}\}, \\ s &= (1/2a_2b')\{b^2 - b'^2 - a_1a_2 \pm [((b + b')^2 - a_1a_2)((b - b')^2 - a_1a_2)]^{1/2}\}. \end{aligned} \quad (18.12)$$

From (18.10), we have

$$c^2 \cos \phi_1 + s^2 \sin^2 \phi_1 = 1, \quad (18.13)$$

from which we can determine ϕ_1 . This equation is equivalent to two equations:

$$c^2 x^2 + s^2 y^2 = 1, \quad x^2 + y^2 = 1.$$

The crosspoint of these ellipse and circle gives the value of ϕ_1 . We may assume that both ϕ_1 and ϕ_2 occur in the first quadrant. Then we have

$$c > 1 > s > 0 \quad \text{or} \quad s > 1 > c > 0. \quad (18.14)$$

This inequality imposes certain restrictions on the exchange constants, which we shall discuss briefly at the end of this section. It can be shown that there are cases where the inequality (18.14) is satisfied. If ϕ_1 and ϕ_2 turn out to be zero or $\pi/2$, only \mathbf{q} or \mathbf{q}' will survive.

It can be shown that the minimum energy is expressible in terms of c and s as

$$\begin{aligned} E/N &= A_{11}(\mathbf{q}) + A_{22}(\mathbf{q}) - 2 |A_{12}(\mathbf{q})| - |A_{12}(\mathbf{q})|(c^{1/2} - c^{-1/2})^2 \\ &= A_{11}(\mathbf{q}') + A_{22}(\mathbf{q}') - 2 |\operatorname{Re} \operatorname{Im} A_{12}(\mathbf{q}')| \\ &\quad - |\operatorname{Re} \operatorname{Im} A_{12}(\mathbf{q}')|(s^{1/2} - s^{-1/2})^2. \end{aligned} \quad (18.15)$$

Comparing (18.15) with (18.7), we see that the energy of the configuration in which \mathbf{q} and \mathbf{q}' coexist is lower than the energy of the configuration in which \mathbf{q} or \mathbf{q}' alone exists (i.e., $u_1 = u_2 = 1$ or $u_1' = u_2' = 1$). Our mathematical problem will then be to minimize (18.15) with respect to \mathbf{q} , but in this process, one of the components, that having \mathbf{q} or \mathbf{q}' , could vanish.

Either of the inequalities (8.14) imposes certain restrictions on a_1 , a_2 , b , and b' . To study these restrictions, we assume that $a_2 > 0$; if otherwise, the primed and unprimed A 's may be interchanged [and, at the same time, c and s must be interchanged because of the definitions (18.9) and (18.10)]. Some elaborate analysis is necessary to find the conditions under which c and s are ensured to be real positive and either of the inequalities (18.14) is ensured to hold. Here the results only will be written.

For simplicity, we write

$$b/b' = \beta, \quad a_1/b' = \alpha_1, \quad a_2/b' = \alpha_2.$$

The condition $s > 1 > c > 0$ requires the upper sign in (18.12). It requires further that one of the two sets of inequalities, (A) and (B) given below, must be satisfied:

$$(A) \quad \alpha_2 > \beta - 1 > \alpha_1, \quad 1 > (\beta - \alpha_1)(\beta - \alpha_2), \quad \beta^2 - 1 > \alpha_2(\alpha_1 + 2).$$

$$(B) \quad \alpha_2 > \beta - 1 > \alpha_1, \quad 1 > (\beta - \alpha_1)(\beta - \alpha_2), \quad \alpha_2(\alpha_1 + 2) > \beta^2 - 1,$$

$$\beta^2 > (\alpha_1 + 1)(\alpha_2 + 1).$$

In order that $c > 1 > s > 0$ be satisfied, one must take the lower sign in (18.12), and the following set of inequalities must hold:

$$(C) \quad \alpha_1 > \beta - 1 > \alpha_2 > 0, \quad 1 > (\beta - \alpha_1)(\beta - \alpha_2), \\ \beta^2 - 1 > \alpha_2(2\beta - \alpha_1), \quad \beta^2 > (\alpha_1 + 1)(\alpha_2 + 1).$$

One can verify that within each of the groups, (A), (B), and (C), none of the inequalities is superfluous, nor is it conflicting with other inequalities. Hence, there may be cases where one of these groups of inequalities holds. If α_1 is positive in (A) or (B), one can prove that $(\beta - 1)^2 > \alpha_1\alpha_2$, so that c and s given by (18.12) are real and positive. In particular, if $\alpha_1 = \alpha_2$, it follows that $\beta = 1$, and hence $\alpha_1 = \alpha_2 = 0$; in other words, if the two sublattices are equivalent to each other and the Fourier transform of the exchange constants within each sublattice is nonvanishing, coexistence of two wave vectors is impossible.

19. LYONS-KAPLAN THEORY

We have studied spin configurations under the assumption that there is a single pair of wave vectors $(\mathbf{q}, -\mathbf{q})$ or there are two pairs $(\mathbf{q}, -\mathbf{q})$ and $(\mathbf{q}', -\mathbf{q}')$ in the whole crystalline structure. We are, however, not sure whether or not the spin configuration of the lowest energy to be obtained under such an assumption is the configuration of the lowest energy of all possible configurations. A mathematical theory developed by Kaplan and his co-workers^{22b-f} helps us to elucidate this problem. We shall describe it below in a little different form.

We have the condition (VI.6) imposed on each spin: $\mathbf{s}_{n\nu}^2 = S_\nu^2$. It can be written in another form:

$$\sum_{\mathbf{q}''} \left(\sum_{\mathbf{q}} \mathbf{d}_{\mathbf{q}\nu} \cdot \mathbf{d}_{\mathbf{q}-\mathbf{q}''\nu}^* \right) \exp(i\mathbf{q}'' \cdot \mathbf{R}_n) = 1.$$

Note that $\mathbf{d}_{\mathbf{q}\nu} = \mathbf{d}_{-\mathbf{q}\nu}^*$. In order that this equation be satisfied for all \mathbf{R}_n , the following equations must hold:

$$\sum_{\mathbf{q}} \mathbf{d}_{\mathbf{q}\nu} \cdot \mathbf{d}_{\mathbf{q}\nu}^* = 1, \tag{19.1}$$

$$\sum_{\mathbf{q}} \mathbf{d}_{\mathbf{q}\nu} \cdot \mathbf{d}_{\mathbf{q}-\mathbf{q}''\nu}^* = 0 \quad (\mathbf{q}'' \neq 0). \tag{19.2}$$

For a single pair of $(\mathbf{q}, -\mathbf{q})$, these equations reduce to^{24a}

$$2\mathbf{d}_{\mathbf{q}\nu} \cdot \mathbf{d}_{\mathbf{q}\nu}^* = 1, \tag{19.3}$$

$$\mathbf{d}_{\mathbf{q}\nu}^2 = \mathbf{d}_{\mathbf{q}\nu}^{*2} = 0. \tag{19.4}$$

^{24a} When \mathbf{q} and $-\mathbf{q}$ are equivalent vectors, the factor 2 in (19.3) drops; (19.5) is then replaced by $\mathbf{d}_{\mathbf{q}\nu} = u_{\mathbf{q}\nu} \mathbf{i}$.

(19.4) can be satisfied if we assume for $\delta_{q\nu}$ the form

$$\delta_{q\nu} = \frac{1}{2}u_{q\nu}(i - ij), \quad (19.5)$$

where i and j are orthogonal unit vectors (that may depend on ν) and $u_{q\nu}$ is a complex number. To satisfy (19.3), we must take $|u_{q\nu}| = 1$. For two pairs, $(q, -q)$ and $(q', -q')$, where q' may be zero or $K/2$, or even $K/4$, according to the preceding section, we may also assume (19.5) and have to take

$$\delta_{q'\nu} = u_{q'\nu}k \quad \text{when } q' = 0 \text{ or } K/2, \quad (19.6)$$

$$\delta_{q'\nu} = \frac{1}{2}(1 - i)u_{q'\nu}k \quad \text{or} \quad \frac{1}{2}(1 + i)u_{q'\nu}k \quad \text{when } q' = K/4, \quad (19.7)$$

where $u_{q'\nu}$ is a real number and k a unit vector perpendicular to both i and j . Equation (19.2) is then satisfied. Equation (19.1) is written as

$$|u_{q\nu}|^2 + |u_{q'\nu}|^2 = 1. \quad (19.8)$$

We see by these examples that (19.2) can be satisfied by appropriate forms of $\delta_{q\nu}$, whereas (19.1) puts relations between the magnitudes of these vectors.

Assuming that (19.2) can somehow be satisfied, let us confine ourselves to condition (19.1) and look for the lowest value of the exchange energy (VI.4). We may introduce a set of Lagrange multipliers $\{\lambda_\nu\}$, $\nu = 1, 2, \dots, k$, and consider the problem

$$\sum_q \sum_\mu \sum_\nu A_{\mu\nu}(q) \delta_{q\mu} \cdot \delta_{q\nu}^* - \sum_\nu \lambda_\nu \sum_q \delta_{q\nu} \cdot \delta_{q\nu}^* = \min. \quad (19.9)$$

Since this expression is diagonal with respect to q , we may first confine ourselves to a single pair of q and $-q$. Then our minimum problem will be reduced to solving the equations

$$\sum_\mu A_{\mu\nu}(q) \delta_{q\mu} = \lambda_\nu \delta_{q\nu}, \quad \nu = 1, 2, \dots, k. \quad (19.10)$$

The negative of the left-hand side of this equation is the exchange field acting on the ν th sublattice when there is a helical spin arrangement of wave vector q .

$$\sum_\mu A_{\mu\nu}(q) \delta_{q\mu} \cdot \delta_{q\nu}^* + \text{complex conjugate}$$

is the energy associated with the ν th sublattice in this configuration. (The complex conjugate arises from $-q$.) It consists of the energy of interaction within the ν th sublattice and half the energy of interaction with other sublattices, since when summed over ν it gives the total energy (per unit cell). Hence, twice this energy is the energy of interaction between

each spin and the Weiss molecular field in the ν th sublattice. It must be negative when a pair of \mathbf{q} and $-\mathbf{q}$ gives the stable spin configuration; if it were positive that would mean that the spins are aligned opposite to the Weiss field, and hence the configuration is not stable. Multiplying Eq. (19.10) by $\delta_{\mathbf{q}\nu}^*$ and adding the complex conjugate, we obtain on the left-hand side the energy considered and on the right-hand side the quantity $2\lambda, \delta_{\mathbf{q}\nu} \cdot \delta_{\mathbf{q}\nu}^*$. Hence, λ , must be negative. Thus, we put

$$\lambda_\nu = \lambda/\beta_\nu^2, \quad (19.11)$$

where λ is negative real and β_ν real. Then (19.10) can be written as

$$\sum_\mu A_{\mu\nu}(\mathbf{q}) \delta_{\mathbf{q}\mu} = (\lambda/\beta_\nu^2) \delta_{\mathbf{q}\nu}, \quad \nu = 1, 2, \dots, k. \quad (19.12)$$

Rewriting this equation in the form

$$\sum_\mu \beta_\mu \beta_\nu A_{\mu\nu}(\mathbf{q}) (\delta_{\mathbf{q}\mu}/\beta_\mu) = \lambda (\delta_{\mathbf{q}\nu}/\beta_\nu), \quad (19.12a)$$

one will see that the problem here concerned is an eigenvalue problem. The eigenvalues are obtained from

$$\det |A_{\mu\nu}(\mathbf{q}) - \lambda/\beta_\nu^2| = 0 \quad (19.13)$$

or

$$\det |\beta_\mu \beta_\nu A_{\mu\nu}(\mathbf{q}) - \lambda| = 0. \quad (19.13a)$$

For each eigenvalue, we can determine the eigenvector $(\delta_{\mathbf{q}1}, \delta_{\mathbf{q}2}, \dots, \delta_{\mathbf{q}k})$ from Eq. (19.12) or (19.12a). Since each $\delta_{\mathbf{q}\nu}$ is a vector (complex vector) and Eq. (19.12) or (19.12a) implies that each of its three Cartesian components satisfies the same equation, we obtain the same ratio of $\sigma_{\mathbf{q}1i}, \sigma_{\mathbf{q}2i}, \dots, \sigma_{\mathbf{q}ki}$ for $i = x, y, z$. This means that we can assume one of the forms (19.5)–(19.7) for the eigenvector, with \mathbf{i}, \mathbf{j} , and \mathbf{k} independent of ν . Then, $u_{\mathbf{q}\nu}$ is determined from equations similar to (19.12):

$$\sum_\mu A_{\mu\nu}(\mathbf{q}) u_{\mathbf{q}\mu} = (\lambda/\beta_\nu^2) u_{\mathbf{q}\nu}, \quad \nu = 1, 2, \dots, k. \quad (19.14)$$

The eigenvalues are functions of \mathbf{q} . We vary \mathbf{q} and look for the lowest eigenvalue. This lowest eigenvalue will be denoted by λ_0 and the corresponding value of \mathbf{q} by \mathbf{Q} . The associated eigenvector will be denoted by $(\delta_{\mathbf{Q}1}^0, \delta_{\mathbf{Q}2}^0, \dots, \delta_{\mathbf{Q}k}^0)$ or $(u_{\mathbf{Q}1}^0, u_{\mathbf{Q}2}^0, \dots, u_{\mathbf{Q}k}^0)$. \mathbf{Q}, λ_0 , and the eigenvector depend still on parameters β_ν . To determine these parameters and simultaneously the value of \mathbf{Q} , we try to satisfy condition (19.3) by assuming (19.5) when \mathbf{Q} is a general vector. When \mathbf{Q} is zero or $\mathbf{K}/2$, we assume (19.6) and try to satisfy $|\delta_{\mathbf{Q}\nu}^0|^2 = 1$. For $\mathbf{Q} = \mathbf{K}/4$, we may assume either (19.5) or (19.7) and consider (19.3). In all these cases, we are led to the condition $|u_{\mathbf{Q}\nu}^0|^2 = 1$. If we are able to satisfy this condition by taking

appropriate values of β_ν , and to determine simultaneously the value of \mathbf{Q} , and if the corresponding λ_0 is ensured to be the lowest *nondegenerate* eigenvalue among all the eigenvalues including those for different \mathbf{q} values, then the spin configuration represented by $(u_{Q1}^0, u_{Q2}^0, \dots, u_{Qk}^0)$ is the configuration of the lowest energy, as we shall prove below explicitly. In this comparison of the eigenvalues, β_ν 's are fixed at the adopted values.

From (19.12) follows that the energy of the spin configuration under consideration is given by

$$\begin{aligned} E_0/N &= \sum_{\mu} \sum_{\nu} A_{\mu\nu}(\mathbf{Q}) \delta_{Q\mu}^0 \cdot \delta_{Q\nu}^{*0} + \text{complex conjugate} \\ &= \lambda_0 \sum_{\nu} \beta_\nu^{-2} 2 |\delta_{Q\nu}^0|^2 = \lambda_0 \sum_{\nu} \beta_\nu^{-2}. \end{aligned} \quad (19.15)$$

(In the case of $\mathbf{Q} = 0$ or $\mathbf{Q} = \mathbf{K}/2$, the term "complex conjugate" and the factor 2 before $|\delta_{Q\nu}^0|^2$ do not appear.) Now consider any other real or unreal spin configuration represented by a set of Fourier coefficients $\{\delta_{\mathbf{q}\nu}\}$ that satisfy Eq. (19.1). These coefficients can be expressed as a linear combination of eigenvectors $\{\delta_{\mathbf{q}\nu}^\alpha\}$ in the form

$$\sigma_{\mathbf{q}\nu i} = \sum_{\alpha} a_{\mathbf{q}\nu i}^\alpha \sigma_{\mathbf{q}\nu i}^\alpha \quad (i = x, y, z),$$

where i denotes cartesian component and α specifies different eigenvectors ($\alpha = 1, 2, \dots, k$). The replacement of \mathbf{q} by $-\mathbf{q}$ always means the change of quantities into their complex conjugates. For convenience, $\sigma_{\mathbf{q}\nu i}^\alpha$ may here be normalized according to

$$\sum_{\nu} \sigma_{\mathbf{q}\nu i}^\alpha (\sigma_{\mathbf{q}\nu i}^\alpha)^* / \beta_\nu^2 = \sum_{\nu} 1 / \beta_\nu^2.$$

We denote by $\lambda_\alpha(\mathbf{q}) [= \lambda_\alpha(-\mathbf{q})]$ the eigenvalue corresponding to $\{\delta_{\mathbf{q}\nu}^\alpha\}$. Then using (19.12), we can calculate the energy of the spin configuration $\{\delta_{\mathbf{q}\nu}\}$ as follows:

$$\begin{aligned} E/N &= \sum_{\mathbf{q}} \sum_{\mu} \sum_{\nu} A_{\mu\nu}(\mathbf{q}) \delta_{\mathbf{q}\mu} \cdot \delta_{\mathbf{q}\nu}^* \\ &= \sum_{\mathbf{q}} \sum_{\mu} \sum_{\nu} A_{\mu\nu}(\mathbf{q}) \sum_i \left(\sum_{\alpha} a_{\mathbf{q}\nu i}^\alpha \sigma_{\mathbf{q}\nu i}^\alpha \right) \left(\sum_{\alpha'} (a_{\mathbf{q}\nu i}^{\alpha'})^* (\sigma_{\mathbf{q}\nu i}^{\alpha'})^* \right) \\ &= \sum_{\mathbf{q}} \sum_{\nu} \sum_i \sum_{\alpha} \sum_{\alpha'} a_{\mathbf{q}\nu i}^\alpha (a_{\mathbf{q}\nu i}^{\alpha'})^* [\lambda_\alpha(\mathbf{q}) / \beta_\nu^2] \sigma_{\mathbf{q}\nu i}^\alpha (\sigma_{\mathbf{q}\nu i}^{\alpha'})^*. \end{aligned}$$

In the last line, the sum over ν vanishes except when $\alpha = \alpha'$, since different eigenvectors are orthogonal:

$$\sum_{\nu} (\sigma_{\mathbf{q}\nu i}^\alpha / \beta_\nu) (\sigma_{\mathbf{q}\nu i}^{\alpha'} / \beta_\nu)^* = 0 \quad (\alpha \neq \alpha').$$

Hence, utilizing the normalization condition for $\alpha = \alpha'$, we obtain

$$E/N = \sum_{\mathbf{q}} \sum_i \sum_{\alpha} |a_{\mathbf{q}i}^{\alpha}|^2 \lambda_{\alpha}(\mathbf{q}) \sum_{\nu} 1/\beta_{\nu}^2. \quad (19.16)$$

On the other hand, from (19.1) we have

$$\sum_{\mathbf{q}} \sum_{\nu} \delta_{\mathbf{q}\nu} \cdot \delta_{\mathbf{q}\nu}^* / \beta_{\nu}^2 = \sum_{\nu} 1/\beta_{\nu}^2,$$

which can be transformed by a similar calculation into

$$\sum_{\mathbf{q}} \sum_i \sum_{\alpha} |a_{\mathbf{q}i}^{\alpha}|^2 \sum_{\nu} 1/\beta_{\nu}^2 = \sum_{\nu} 1/\beta_{\nu}^2,$$

so that

$$\sum_{\mathbf{q}} \sum_i \sum_{\alpha} |a_{\mathbf{q}i}^{\alpha}|^2 = 1. \quad (19.17)$$

Thus, the energy (19.16) is a weighted mean of eigenvalues multiplied by $\sum_{\nu} 1/\beta_{\nu}^2$. Evidently, it is higher than the energy (19.15).

There are k parameters β_{ν} to be determined from the k conditions $|u_{\mathbf{Q}\nu}^0| = 1$. However, one of these parameters can be put equal to 1, since only the ratio of $u_{\mathbf{Q}1}^0, u_{\mathbf{Q}2}^0, \dots, u_{\mathbf{Q}k}^0$ is determined from the eigenvalue equation. Although this determination of the parameters may be possible in some cases, there may be also other cases where real values of the parameters cannot be obtained. In the latter case, we have to look for other spin configurations.^{24b}

We may then examine spin configurations in which two wave vectors

^{24b} If a helical configuration having a wave vector \mathbf{Q} is the configuration of the lowest energy, it must result from the eigenvalue equation (19.12), and thus the determination of β_{ν} for this \mathbf{Q} must be possible. However, it will not immediately follow that the eigenvalue $\lambda_0(\mathbf{Q})$ is the lowest of all eigenvalues for this set of β_{ν} . That this configuration has the lowest energy would mean that $\lambda_0(\mathbf{Q}) \sum_{\nu} 1/\beta_{\nu}^2$ [see (19.15)] is the lowest of (19.16), i.e., by virtue of (19.17), that

$$\sum_{\mathbf{q}} \sum_i \sum_{\alpha} |a_{\mathbf{q}i}^{\alpha}|^2 [\lambda_{\alpha}(\mathbf{q}) - \lambda_0(\mathbf{Q})] \cdot \sum_{\nu} 1/\beta_{\nu}^2 \geq 0.$$

If we were able to choose $a_{\mathbf{q}i}^{\alpha}$ arbitrarily, except that they are subject to condition (19.17), then the absolute minimum of $\lambda_0(\mathbf{Q})$ would follow from the preceding inequality. In actuality, however, $a_{\mathbf{q}i}^{\alpha}$ are subject to further conditions that would result from (19.2), although no explicit use of such conditions was made in deriving (19.16) and (19.17). When the helical configuration having \mathbf{Q} as its wave vector is the lowest in energy compared with other *helical* configurations, what we can say is only that $\lambda_0(\mathbf{Q}, \beta_{\nu}) \cdot \sum_{\nu} 1/\beta_{\nu}^2$, β_{ν} being determined for \mathbf{Q} , is the lowest of all $\lambda_0(\mathbf{q}, \beta_{\nu}) \cdot \sum_{\nu} 1/\beta_{\nu}^2$, β_{ν} being determined for \mathbf{q} (not \mathbf{Q}). It will then not necessarily follow that $\partial \lambda_0(\mathbf{q}, \beta_{\nu}) / \partial \mathbf{q} = 0$ for $\mathbf{q} = \mathbf{Q}$, i.e., that $\lambda_0(\mathbf{Q}, \beta_{\nu})$ is the lowest of all $\lambda_0(\mathbf{q}, \beta_{\nu})$ when β_{ν} 's are determined for \mathbf{Q} . In other words, even if we fail in the procedure mentioned in the text, we cannot exclude the existence of a helical configuration as the stable configuration. A similar situation arises in the case of a configuration having two or more wave vectors.

coexist. One of these two can be a general wave vector, which we shall denote by \mathbf{Q} . The other must be a special wave vector, such as zero or $\mathbf{K}/2$. In this case, the spin configuration is conical, since we must assume (19.5) and (19.6) for the two wave vectors, with \mathbf{i} , \mathbf{j} , and \mathbf{k} perpendicular to each other, as we have shown in Section 18. Also, (19.8) must be satisfied for $\mathbf{q} = \mathbf{Q}$ and $\mathbf{q}' = 0$ or $\mathbf{q}' = \mathbf{K}/2$. In place of (19.9), we now have

$$\sum_{\mu} \sum_{\nu} [A_{\mu\nu}(\mathbf{q}) u_{q\mu} u_{q\nu}^* + A_{\mu\nu}(\mathbf{q}') u_{q'\mu} u_{q'\nu}^*] - \sum_{\nu} \lambda_{\nu} (u_{q\nu} u_{q\nu}^* + u_{q'\nu} u_{q'\nu}^*) = \min.$$

Thus, we are led to the same equation as (19.10) for both $u_{q\nu}$ and $u_{q'\nu}$, and the negativeness of λ , can be seen in the same way as before for the stable spin configuration. Putting λ_{ν} as in (19.11), we have Eq. (19.13) or (19.13a) to determine the same value of λ for $\mathbf{q} = \mathbf{Q}$ and $\mathbf{q}' = 0$ or $\mathbf{K}/2$. The eigenvectors are determined from (19.14) for $\mathbf{q} = \mathbf{Q}$ and $\mathbf{q} = 0$ or $\mathbf{K}/2$.

There are again k parameters β , and, in this case, two amplitudes of the eigenvectors. One of these $k + 2$ constants can be put equal to 1, and the remaining $k + 1$ are to be determined from the k equations (19.8) and the condition that the eigenvalues for the two wave vectors take the same value. The value of \mathbf{Q} is determined simultaneously by the equation $\text{grad}_{\mathbf{Q}}\lambda(\mathbf{Q}) = 0$. The equality of the two eigenvalues has been called "forced degeneracy" by Lyons and Kaplan, since the parameters are so chosen as to make the two eigenvalues degenerate.

After the determinations just mentioned have been made, we have to check if the degenerate eigenvalues are the lowest of all eigenvalues for the adopted values of the β 's and if there is no other degeneracy. When we succeed in all these procedures, we are sure that we have found the stable spin configuration. That this configuration has the lowest energy can be seen from (19.16) and (19.17). When we fail, we may proceed to investigating spin configurations having three or more wave vectors, but the calculations will become more complicated and might be unsuccessful.

VII. Spin Configurations in Spinel Lattice

As an application of the preceding section, we shall here study possible spin configurations in the spinel-type lattice. A complete study of this problem has not yet been made, although detailed calculations have been carried out by Kaplan and his co-workers. We shall here limit our discussion to certain simple features of the problem. A brief account of the crystalline and magnetic structures of the spinel lattice will be given first.

20. CRYSTALLINE AND MAGNETIC STRUCTURES

In the spinel lattice, there are two kinds of cation sites, usually called *A* and *B* sites, which are surrounded tetrahedrally and octahedrally, respectively, by anions. The structure is shown in Fig. 8, which can be derived from the NaCl-type lattice by removing every other cation along each line parallel to each of the principal axes and returning half of the removed cations to fill one-eighth of the vacant tetrahedral sites.

In MnCr_2O_4 , the Mn^{2+} occupy the *A* sites and Cr^{3+} the *B* sites; this type of cation arrangement is called *normal*. In Fe_3O_4 , as is well known, half of the Fe^{3+} occupy *A*, and the other half Fe^{3+} and Fe^{2+} occupy *B*; this type is called *inverse*. Chromites are usually normal, but ferrites are often inverse. Some compounds, such as CuCr_2O_4 and FeCr_2O_4 , are tetragonally distorted below a certain critical temperature by a cooperative Jahn-Teller effect, a theory of which has been worked out by Kanamori.²⁵

In Néel's theory of ferrimagnetism,²⁶ the spins of the *B* sites are assumed to align parallel to each other and antiparallel to the spins of the *A* sites. The *A-B* exchange interaction is assumed to be dominant and antiferromagnetic. Yafet and Kittel²⁰ have shown, however, that the Néel structure is no longer stable when the *B-B* (or *A-A*) exchange interaction becomes

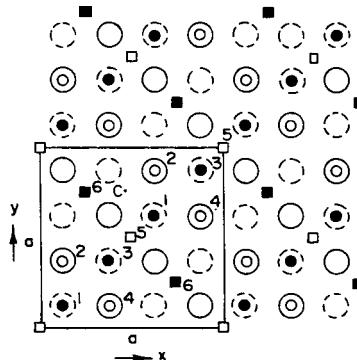


FIG. 8. Structure of spinel lattice. Four (001) layers of atoms projected on the (001) plane are shown. The first layer at zero height consists of cations ● on *B* sites and anions ○. The second layer at a height of $c/8$ ($= a/8$ in cubic case) consists exclusively of cations ■ on *A* sites. The third layer is at a height of $c/4$ and consists of cations ○ on *B* sites and anions (dashed circles). The fourth layer is at $3c/8$ and consists of cations □ on *A* sites. The whole structure is generated by a vertical translation of $c/4$ combined with a rotation by 90° about the vertical line which passes the point *C* in the figure. In actual crystals, the four anions surrounding each *A* site are displaced toward the latter.

²⁵ J. Kanamori, *J. Appl. Phys.* **31**, 14S (1960).

²⁶ L. Néel, *Ann. Phys. (Paris)* [12] **3**, 137 (1948).

large and antiferromagnetic. They have shown that a triangular spin arrangement is more stable in this case; this is an arrangement in which the *B* (or *A*) lattice is split into two equal sublattices having moments at an angle less than 180° and the resultant of these moments is antiparallel to the *A* (or *B*) moment. They have also shown that, when both the *B-B* and *A-A* interactions are large and antiferromagnetic, the *B* and *A* lattices both have antiferromagnetic arrangement (uncorrelated to each other). As mentioned before, copper chromite, CuCr_2O_4 , has a triangular arrangement.²¹ Here the *B* lattice is split into two in such a way that each (001) net plane of the *B* atoms (Cr) is ferromagnetic, and its moment direction alternates from plane to plane. Jacobs²⁷ made magnetic measurements at high fields on CuCr_2O_4 and two other tetragonal spinels, Mn_3O_4 and FeCr_2O_4 , as well as on several ferrite-chromite solid solutions, and observed a linear field-dependence of the moment, from which he expected that triangular spin arrangements exist in these substances. Mn_3O_4 possesses a spin configuration characterized by a wave vector along $[110]$,²⁸ and FeCr_2O_4 has a multiple cone configuration at low temperatures, as will be described below.

The magnetic structure of a cubic spinel MnCr_2O_4 , observed by Hastings and Corliss,²³ is the first example of the conical structure predicted by Kaplan *et al.*^{22c-e} (*ferrimagnetic spiral*, as they call it). The Néel temperature of this substance was observed to be about 43°K , and the neutron magnetic reflection lines down to 18°K corresponded to a Néel-type collinear spin arrangement (there was, however, an additional diffuse peak in the region where satellite lines develop below 18°K), but below 18°K additional sharp lines (satellites) appeared, which could be interpreted as being due to a transition to a conical spin arrangement. The Mn-spins (on sites 5 and 6 in our Fig. 8), the Cr_1 -spins (on sites 1 and 3), and the Cr_2 -spins (on sites 2 and 4) rotate on their respective cones under translation along $[110]$ to equivalent sites in other unit cells. The rotation corresponds to a wave number of 0.98 \AA^{-1} (at 4.2°K , the lattice constant at room temperature being 8.437 \AA). The cone axes are common and parallel to $[1\bar{1}0]$, and the cone angles (measured from $[1\bar{1}0]$) are 24° , 152.5° ($= 180^\circ - 27.5^\circ$), 104° ($= 180^\circ - 76^\circ$) for Mn, Cr_1 , Cr_2 , so that the cones for Cr are opposite to the cone for Mn. These results are in good agreement with the theory. However, there are unexplained features, particularly concerning the magnitudes of the atomic magnetic moments. Also, the line intensities giving the magnitudes of the axial components of the moments did not change in going through the transition at 18°K ; together with a diffuse peak in the region of the satellites, this fact would suggest a disorder or large fluctuations in the transverse components

²⁷ I. S. Jacobs, *Phys. Chem. Solids* **15**, 54 (1960).

²⁸ J. S. Kasper, *Bull. Am. Phys. Soc.* [2] **4**, 178 (1959).

above 18°K. Furthermore, some aspects of the theory have not yet been fully clarified, as will be mentioned later.^{28a}

Attempts to measure the cone angles by NMR have been made.^{29,30} The idea is that by applying a magnetic field the cone axis would become parallel to the field and the internal magnetic field acting on the nucleus of each magnetic atom, whose direction is parallel to the spin moment of the atom, would change proportionally to the strength of the applied field and the cosine of the cone angle. Under this assumption and from the observed linear field-dependence of the resonance frequency, the angles were determined to be 68°, 94°, 97° for Mn, Cr₁, Cr₂, which do not agree with the neutron diffraction results.

In FeCr₂O₄, a conical structure was also observed.^{31,32} This crystal becomes tetragonal below 135°K. (The *c*-axis is only 3% smaller than the *a* axis; *c* = 8.21 Å, and *a* = 8.46 Å.) It has a Néel temperature of about 80°K, and below about 40°K it gives satellites that indicate a conical structure. The propagation vector is parallel to [110] and has a value of 0.063 Å⁻¹ (see Shirane and Cox³¹) or 0.037 Å⁻¹ (see Bacchella and Pinot³²), or parallel to [001] and has a value of 0.026 Å⁻¹ (see Bacchella and Pinot³²), depending on different indexing of the lines. In the range between 80° and 40°K, the spin component perpendicular to the cone axis seems to be disordered or fluctuating.

A cubic spinel CoCr₂O₄ at low temperatures also produces neutron lines that can be interpreted by a conical structure.³³ The propagation vector is again parallel to [110] and has a value of $0.62 \times 2^{1/2}/a$ (*a* = 8.332 Å at room temperature). The cone axis is parallel to [001]. The cone angles have been determined to be 32°, 90°, 150° for Co, Cr₁, Cr₂, and the magnetic moments of these ions to be 3 Bohr magnetons (the spin only value). The Néel temperature is 97°K. The temperature-dependence of magnetic properties reported by Menyuk, Dwight, and Wold³³ is extremely interesting. They performed a detailed calculation of the conical spin configuration over the whole temperature range from 0° to 97°K under the assumption of the molecular field approximation. Taking a parameter value *u* = 2.03 (*u* = $4J_{BB}S_B/3J_{AB}S_A$, which will appear in the next section), they found a very good agreement between the calculated and observed curves of the saturation magnetization versus temperature, except below 27°K. With increasing temperature, the calculated curve increases gradually up to about 20°K, then rises linearly with temperature, and attains a high broad

^{28a} The *u* parameter, which we shall discuss in Part VII, Section 22, is 1.6 in this example, which is too large to ensure the stability of the conical structure.

²⁹ T. W. Houston and A. J. Heeger, *Phys. Letters* **10**, 29 (1964).

³⁰ H. Nagasawa and T. Tsushima, *Phys. Letters* **15**, 205 (1965).

³¹ G. Shirane and D. E. Cox, *J. Appl. Phys.* **35**, 954 (1964).

³² G. L. Bacchella and M. Pinot, *J. Phys.* **25**, 537 (1964).

³³ N. Menyuk, K. Dwight, and A. Wold, *J. Phys.* **25**, 528 (1964).

maximum at 77°K, which is then followed by a steep decrease going to zero at 97°K. On the other hand, the observed curve decreases gradually up to 27°K, shows a break in slope at this temperature, and then follows closely the calculated curve. The calculation showed that a transition occurs at 86°K from the conical configuration to the Néel colinear arrangement. The calculation was very sensitive to the choice of u ($\pm 2.5\%$ changes in the u value resulted in conspicuous changes in the magnetization curve). Despite the discrepancy below 27°K, the neutron line positions and intensities at 4.2°K can be accounted for very well by the theoretical model. But, despite the agreement above 27°K, the satellite lines corresponding to the rotating component of the spins do not appear as sharp lines but appear only as diffuse peaks, whereas the magnetic contributions to the intensities of the fundamental lines, corresponding to the axially aligned magnetic moments, are in excellent agreement with those predicted theoretically. The authors note that the Lyons-Kaplan-Dwight-Menyuk theory^{22d} of the conical spin configuration at absolute zero predicts local instability (against small spin deviations) for $u > 1.3$ and that this may have a relationship with the low-temperature discrepancy. They note further that the absence of sharp satellites for the rotating component above 27°K may be due to a failure of the molecular field theory, which does not adequately takes account of correlations between the thermal fluctuations of neighboring spins.

In contrast to the complicated spin structures mentioned previously, a very simple helical spin arrangement was observed in $ZnCr_2Se_4$,³⁴ and it was interpreted theoretically.³⁵ In this crystal, there are magnetic atoms only on B sites. The spins in each lattice plane (001) are parallel and lie in the plane, and the helix propagates in the $\langle 001 \rangle$ direction with a turn angle of 42° (at 4.2°K) for adjacent (001) planes. Of course, the second (and further) neighbor interactions are necessary to account for the helical structure.^{35a}

21. THE NÉEL AND YAFET-KITTEL CONFIGURATIONS

It is not our intention to study completely the spin configurations in the spinel lattice. Our main purpose is only to show the main idea of how to find these configurations and how to discuss their stability.

In the cubic or tetragonal unit cell shown in Fig. 8, there are sixteen B sites and eight A sites, but if we take a regular or distorted rhombohedral unit cell defined by basic vectors $(0, \frac{1}{2}, \frac{1}{2})$, $(\frac{1}{2}, 0, \frac{1}{2})$, and $(\frac{1}{2}, \frac{1}{2}, 0)$, there

³⁴ R. Plumier, *Compt. Rend.* **260**, 3348 (1965).

³⁵ F. K. Lotgering, *Solid State Commun.* **3**, 347 (1965).

^{35a} $ZnCr_2O_4$ becomes tetragonal below 20°K ($= T_N$), with a decrease of about 0.05% in c/a . Precise measurements of the temperature variation of c/a were made by R. Kleinberger and R. de Kouchkovsky, *Compt. Rend.* **262**, 628 (1966).

are four B sites and two A sites. Taking the origin at one of these four B sites, we can write the regular or distorted rhombohedral coordinates of the six sites as

$$\begin{array}{cccc} 1 & 2 & 3 & 4 \\ B: & (0, 0, 0), (\frac{1}{2}, 0, 0), (0, 0, \frac{1}{2}), (0, \frac{1}{2}, 0); \\ 5 & 6 \\ A: & (\frac{3}{8}, \frac{3}{8}, \frac{3}{8}), (\frac{5}{8}, \frac{5}{8}, \frac{5}{8}). \end{array}$$

In the original cubic or tetragonal coordinate system, these can be written as

$$\begin{array}{cccc} 1 & 2 & 3 & 5 \\ B: & (0, 0, 0), (0, \frac{1}{4}, \frac{1}{4}), (\frac{1}{4}, \frac{1}{4}, 0), (\frac{1}{4}, 0, \frac{1}{4}); \\ 5 & 6 \\ A: & (\frac{3}{8}, \frac{3}{8}, \frac{3}{8}), (\frac{5}{8}, \frac{5}{8}, \frac{5}{8}). \end{array}$$

These sites change as $1 \rightarrow 2 \rightarrow 3 \rightarrow 4$ and $5 \rightarrow 6 \rightarrow 5 \rightarrow 6$ by successive operations of rotation by 90° about the vertical line passing through the point C in Fig. 8 and translation by $\frac{1}{4}$ parallel to this axis.^{35b} This property is useful in calculating the exchange matrix. As in Kaplan *et al.*,²² we shall take nearest-neighbor exchange interactions only.

There are six B sites surrounding each B site. Two of these six are in the same (001) plane as the central site, and we shall denote the exchange constant between each of these two and the central site by J_{BB} . The remaining four are not in the same (001) plane as the central site, two being below and two above, and we shall denote the corresponding exchange constant by J'_{BB} . There are also six A sites surrounding each B site, two at a level higher by $\frac{1}{8}$ than the central site, two at a level $-\frac{1}{8}$, and one each at levels $\frac{3}{8}$ and $-\frac{3}{8}$. We denote the exchange constants corresponding to these by J_{AB} , J'_{AB} , and J''_{AB} . (Conversely, each A site is surrounded by 12 B sites.) The A lattice is of the diamond type, so that we have only one kind of nearest-neighbor exchange constant, J_{AA} . For convenience, we write^{35c}

$$\begin{aligned} \frac{2J_{BB}S_B^2}{(2J_{AB} + J'_{AB})S_AS_B} &= s, & \frac{2J'_{BB}S_B^2}{(2J_{AB} + J'_{AB})S_AS_B} &= s', \\ \frac{J_{AA}S_A^2}{(2J_{AB} + J'_{AB})S_AS_B} &= t, & s + s' &= u, \\ \frac{s'}{s + s'} &= v, & \frac{J'_{AB}}{2J_{AB} + J'_{AB}} &= w. \end{aligned} \quad (21.1)$$

^{35b} Our 1, 2, 3, 4, 5, 6 correspond to 5, 4, 6, 3, 1, 2 of Kaplan *et al.*²²

^{35c} t , u , v , w are the same as those appearing in Menyuk *et al.*^{22e} Here we shall restrict ourselves to $t = 0$ and $0 \leq w \leq \frac{1}{2}$.

We shall assume $2J_{AB} + J'_{AB} < 0$.

As the exchange matrix, it is convenient to consider $C_{\mu\nu}(\mathbf{q})$ defined by

$$\begin{aligned} C_{\mu\nu}(\mathbf{q}) &= A_{\mu\nu}(\mathbf{q}) \exp [i\mathbf{q} \cdot (\mathbf{R}_\mu - \mathbf{R}_\nu)] / (|2J_{AB} + J'_{AB}| S_A S_B) \\ &= - \sum_n S_\mu S_\nu J_{m\mu, n\nu} \exp [i\mathbf{q} \cdot (\mathbf{R}_{m\mu} - \mathbf{R}_{n\nu})] / (|2J_{AB} + J'_{AB}| S_A S_B). \end{aligned} \quad (21.2)$$

The summation with respect to n is taken over those points on the ν th sublattice which are neighbors of a point on the μ th sublattice. Using this $C_{\mu\nu}(\mathbf{q})$ and writing $\delta_{\mathbf{q}\nu}$ for the previous $\delta_{\mathbf{q}\nu}$ multiplied by $\exp(-i\mathbf{q} \cdot \mathbf{R}_\nu)$, the energy can be expressed as

$$\epsilon = E/(N |2J_{AB} + J'_{AB}| S_A S_B) = \sum_{\mathbf{q}} \sum_{\mu} \sum_{\nu} C_{\mu\nu}(\mathbf{q}) \delta_{\mathbf{q}\mu} \delta_{\mathbf{q}\nu}^*. \quad (21.3)$$

Furthermore, using $u_{\mathbf{q}\nu}$ defined by (19.5) for a general \mathbf{q} and (19.6) or (19.7) for the special vectors written there, we can write a single wave vector term of ϵ (\mathbf{q} or $-\mathbf{q}$ term for a general \mathbf{q}) as^{35d}

$$\epsilon_{\mathbf{q}} = \frac{1}{2} \sum_{\mu} \sum_{\nu} C_{\mu\nu}(\mathbf{q}) u_{\mathbf{q}\mu} u_{\mathbf{q}\nu}^*. \quad (21.4)$$

Our mathematical problem is to find the lowest eigenvalue of this expression by the procedure described in Part VI, Section 19 (the Lyons-Kaplan theory). For this purpose, we must calculate $C_{\mu\nu}(\mathbf{q})$ by (21.2) using the exchange constants appearing in (21.1) and the coordinates of the B and A sites. In this calculation, it is convenient to use cubic or tetragonal coordinates. The results are as follows, where $k_x = aq_x/4$, $k_y = aq_y/4$, and $k_z = cq_z/4$:

$$\begin{aligned} C_{12} &= s' \cos(k_y + k_z), & C_{13} &= s \cos(k_x + k_y), & C_{14} &= s' \cos(k_x + k_z), \\ C_{23} &= s' \cos(k_x - k_z), & C_{24} &= s \cos(k_x - k_y), \\ C_{34} &= s' \cos(k_y - k_z), \end{aligned}$$

$$\begin{aligned} C_{15} &= \frac{1}{2}(1-w) \{ \exp[(i/2)(-3k_x + k_y + k_z)] \\ &\quad + \exp[(i/2)(k_x - 3k_y + k_z)] \} + w \exp[(i/2)(k_x + k_y - 3k_z)] \\ &= C_{16}^*, \end{aligned}$$

$$\begin{aligned} C_{26} &= \frac{1}{2}(1-w) \{ \exp[(i/2)(-k_x - 3k_y + k_z)] \\ &\quad + \exp[(i/2)(3k_x + k_y + k_z)] \} + w \exp[(i/2)(-k_x + k_y - 3k_z)] \\ &= C_{25}^*, \end{aligned}$$

^{35d} This is half of a single wave vector term for \mathbf{q} equal to zero or half a reciprocal lattice vector, and represents half of the energy associated with \mathbf{q} and $-\mathbf{q}$ for any \mathbf{q} .

$$\begin{aligned}
C_{35} &= \frac{1}{2}(1-w)\{\exp[(i/2)(3k_x - k_y + k_z)] \\
&\quad + \exp[(i/2)(-k_x + 3k_y + k_z)]\} + w \exp[(i/2)(-k_x - k_y - 3k_z)] \\
&= C_{36}^*, \\
C_{46} &= \frac{1}{2}(1-w)\{\exp[(i/2)(k_x + 3k_y + k_z)] \\
&\quad + \exp[(i/2)(-3k_x - k_y + k_z)]\} + w \exp[(i/2)(k_x - k_y - 3k_z)] \\
&= C_{45}^*, \\
C_{56} &= 2t[\cos(k_x - k_y) \exp(ik_z) + \cos(k_x + k_y) \exp(-ik_z)]. \quad (21.5)
\end{aligned}$$

We shall first investigate two cases completely, i.e., the case that \mathbf{k} is parallel to [001] and the case that \mathbf{k} is parallel to [100], the lattice being assumed to be tetragonal. Then we shall consider the case of $\mathbf{k} \parallel [110]$. For simplicity, we shall put $t = 0$ and assume $0 < w < \frac{1}{2}$.

(1) $\mathbf{k} = (0, 0, k)$. In this case, we can write the energy (21.4) as

$$\begin{aligned}
\epsilon_{\mathbf{k}} &= \frac{1}{2}\{s' \cos k(u_1u_2^* + u_1u_4^* + u_3u_2^* + u_3u_4^*) + s(u_1u_3^* + u_2u_4^*) \\
&\quad + [(1-w) \exp(ik/2) + w \exp(-3ik/2)] \\
&\quad \times (u_1u_5^* + u_3u_5^* + u_2u_6^* + u_4u_6^*) + [(1-w) \exp(-ik/2) \\
&\quad + w \exp(3ik/2)](u_1u_6^* + u_3u_6^* + u_2u_5^* + u_4u_5^*)\} \\
&\quad + \text{complex conjugate.} \quad (21.6)
\end{aligned}$$

Under a unitary transformation,

$$\begin{aligned}
x_1 &= \frac{1}{2}(u_1 + u_2 + u_3 + u_4), & x_2 &= \frac{1}{2}(u_1 + u_2 - u_3 - u_4), \\
x_3 &= \frac{1}{2}(u_1 - u_2 + u_3 - u_4), & x_4 &= \frac{1}{2}(u_1 - u_2 - u_3 + u_4), \\
x_5 &= 2^{-1/2}(u_5 + u_6), & x_6 &= 2^{-1/2}(u_5 - u_6), \quad (21.7)
\end{aligned}$$

(21.6) becomes

$$\begin{aligned}
\epsilon_{\mathbf{k}} &= s' \cos k(x_1x_1^* - x_3x_3^*) + \frac{1}{2}s(x_1x_1^* - x_2x_2^* + x_3x_3^* - x_4x_4^*) \\
&\quad + 2^{1/2}[(1-w) \cos(k/2) + w \cos(3k/2)](x_1x_5^* + x_5x_1^*) \\
&\quad + i2^{1/2}[(1-w) \sin(k/2) - w \sin(3k/2)](x_8x_6^* - x_6x_8^*). \quad (21.8)
\end{aligned}$$

Thus, the variables are separated into four sets: (x_1, x_5) , x_2 , (x_3, x_6) , x_4 . However, the three variables x_3 , x_4 , x_6 are redundant, since, by replacing k by $k + \pi$, u_2 and u_4 change sign, and u_5 and u_6 change into iu_5 and $-iu_6$, respectively, so that x_1 , x_2 , x_5 change into x_3 , x_4 , ix_6 , and by these changes the terms of (21.8) quadratic in x_1 , x_2 , x_5 go over into those quadratic in x_3 , x_4 , x_6 . Thus, we may put $x_3 = x_4 = x_6 = 0$, or $u_1 = u_2$, $u_3 = u_4$,

$u_5 = u_6$. Hence,

$$x_1 = u_1 + u_3, \quad x_2 = u_1 - u_3, \quad x_5 = 2^{1/2}u_5. \quad (21.9)$$

From (21.8), we have a matrix for x_1 and x_5 :

$$\begin{vmatrix} s' \cos k + \frac{1}{2}s & 2^{1/2} \left[(1-w) \cos \left(\frac{k}{2} \right) + w \cos \left(\frac{3k}{2} \right) \right] \\ 2^{1/2} \left[(1-w) \cos \left(\frac{k}{2} \right) + w \cos \left(\frac{3k}{2} \right) \right] & 0 \end{vmatrix} \quad (21.10)$$

and a one-dimensional matrix for x_2 :

$$-\frac{1}{2}s. \quad (21.11)$$

Solving the equation

$$\begin{vmatrix} s' \cos k + \frac{1}{2}s - \lambda & 2^{1/2} \left[(1-w) \cos \left(\frac{k}{2} \right) + w \cos \left(\frac{3k}{2} \right) \right] \\ 2^{1/2} \left[(1-w) \cos \left(\frac{k}{2} \right) + w \cos \left(\frac{3k}{2} \right) \right] & -\lambda/\beta^2 \end{vmatrix} = 0, \quad (21.12)$$

we obtain two eigenvalues, of which the lower one is

$$\begin{aligned} \lambda_1(k, \beta) = \frac{1}{2} \{ s' \cos k + \frac{1}{2}s - & [(s' \cos k + \frac{1}{2}s)^2 \\ & + 4\beta^2(1 + \cos k)(1 - 2w + 2w \cos k)^2]^{1/2} \}. \end{aligned} \quad (21.13)$$

From (21.11) we obtain another eigenvalue

$$\lambda_2 = -\frac{1}{2}s. \quad (21.14)$$

If we put $k = 0$ and determine β in such a way that u_1 becomes equal to $-u_5$, so that $x_1 = -2^{1/2}x_5$, then we obtain the Néel configuration as represented by the eigenvector x_1, x_5 ($x_2 = 0$). Namely, in the first row of (21.12) we put $k = 0$ and set the ratio of the two elements to be 1 to $2^{1/2}$. Then we have

$$\beta^2 = \frac{1}{2}(1 - \frac{1}{2}s - s') \equiv \beta_N^2 \quad (1 > \frac{1}{2}s + s') \quad (21.15)$$

and, correspondingly, from (21.13)

$$\lambda_1(0, \beta_N) = -1 + \frac{1}{2}s + s' \equiv \lambda_N. \quad (21.16)$$

The energy of this configuration is calculated from

$$\epsilon = (\sum \beta_v^{-2})\lambda = (4 + 2\beta^{-2})\lambda \quad (21.17)$$

to be

$$\epsilon_N = -8 + 2s + 4s'. \quad (21.18)$$

The condition for the stability of the Néel configuration, within the restriction $\mathbf{k} = (0, 0, k)$, is that λ_N is the lowest of $\lambda_1(k, \beta_N)$ and lower than λ_2 . That λ_N is the lowest of $\lambda_1(k, \beta_N)$ is equivalent to the requirement that the determinant appearing in (21.12) is positive for $\lambda = \lambda_N$ when $k \neq 0$ and $\beta = \beta_N$. This gives a condition $s' < \frac{1}{2}$, provided $0 < w < \frac{1}{2}$, as one will see by a simple calculation. From $\lambda_N < \lambda_2$, follows that $s + s' < 1$, which is stronger than the inequality written in parentheses in (21.15). Hence, the stability region of the Néel configuration becomes as shown in Fig. 9.

There are two Yafet-Kittel configurations (for $t = 0$). In one of them, spins on sites 1 and 3 are parallel, and spins on 2 and 4 are also parallel, but the former spins make an angle with the latter spins, and their resultant is antiparallel to the spins on sites 5 and 6. This configuration, which we shall denote as YK1, has been observed in CuCr_2O_4 . The other configuration, YK2, is obtained from YK1 by the interchange of 3 and 2; in this configuration, spins within each (100) net plane of the B lattice are parallel to each other, but their direction alternates from plane to plane. If 3 and 4 are interchanged, this alternation will take place for (010) net planes, but

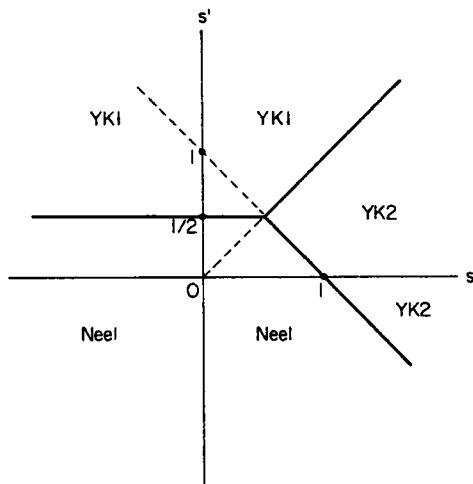


FIG. 9. Stability regions for the three spin configurations, Néel, Yafet-Kittel 1, and Yafet-Kittel 2, obtained from the study of the cases $\mathbf{q} \parallel [001]$ and $\mathbf{q} \parallel [100]$.

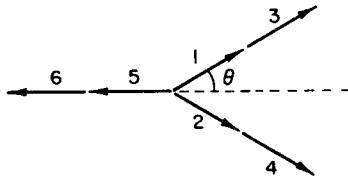


FIG. 10. Sublattice magnetizations in the Yafet-Kittel 1 configuration; $\tan \theta = [(2s')^2 - 1]^{1/2}$.

this configuration is equivalent to that for (100) net planes in tetragonal crystals. In cubic crystals, YK1 and YK2 are equivalent.

To obtain YK1, we put

$$\lambda_1(0, \beta) = \lambda_1(\pi, \beta). \quad (21.19)$$

Since each spin vector is given by $S_{n\nu} = S_s u_\nu \mathbf{k}_q \exp(i\mathbf{q} \cdot \mathbf{R}_{n\nu})$ for a single \mathbf{q} that is zero or a reciprocal lattice vector, where \mathbf{k}_q is an arbitrary unit vector that appeared in (19.6),^{35e} the relations $u_1 = u_2$, $u_3 = u_4$, $u_5 = u_6$ mean that for zero wave vector the spins on 1 and 2 are parallel, the spins on 3 and 4 are parallel, and the spins on 5 and 6 are also parallel, whereas for $\mathbf{k} = (0, 0, \pi)$, or $\mathbf{q} = (0, 0, 4\pi/c)$, each of these pairs consists of anti-parallel spins (spins on 1 are antiparallel to spins on 2, etc.). We note further that the eigenvectors associated with $\lambda_1(0, \beta)$ and $\lambda_1(\pi, \beta)$ have vanishing x_2 ($= u_1 - u_3$), so that the spins on 1 and 3 are parallel in the configuration represented by each of these eigenvectors. Hence, a superposition of these eigenvectors, with $\mathbf{k}_{(0,0,0)}$ and $\mathbf{k}_{(0,0,\pi)}$ at right angles, will yield a YK1 configuration. Now, (21.19) implies

$$\beta^2 = \frac{1}{2}s'(2s' - s) \equiv (\beta_{YK1})^2 \quad \text{if } 2s' > s, s' > 0, \quad (21.20)$$

and $\beta^2 = 0$ if $2s' < s$. We are not interested in the latter, since in this case the eigenvalue vanishes. For the former, we have a negative eigenvalue:

$$\lambda_1(0, \beta_{YK1}) = \lambda_1(\pi, \beta_{YK1}) = \frac{1}{2}s - s' \equiv \lambda_{YK1}. \quad (21.21)$$

The corresponding energy is

$$\epsilon_{YK1} = 2s - 4s' - 2/s'. \quad (21.22)$$

The requirement that λ_{YK1} is the lowest eigenvalue gives an inequality $s' > s$. From the eigenvalue equations, one obtains $u_1/u_5 (= x_1/2^{1/2}x_5) = -1/2s'$ for $k = 0$, whereas for $k = \pi$, u_1 is arbitrary and u_5 vanishes; thus, the two eigenvectors must be superposed with an amplitude ratio of 1 to $[1 - (1/2s')^2]^{1/2}$, so that the spins on 1 have the same magnitude as the

^{35e} In the present case, \mathbf{q} is zero or a reciprocal lattice vector, not half a reciprocal lattice vector; a reciprocal lattice vector appears, since we disregarded x_3 , x_4 , x_6 .

spins on 5 (Fig. 10). The stability region for the YK1 configuration is therefore characterized by $s' > s$ and $2s' > 1$, as shown in Fig. 9.

The YK2 configuration (see Fig. 11) can be obtained from $\lambda_1(0, \beta) = \lambda_2$, with $k = 0$ for λ_2 . This equation gives

$$\beta^2 = \frac{1}{4}s(s + s') \equiv (\beta_{YK2})^2, \quad \lambda_{YK2} = -s/2. \quad (21.23)$$

The corresponding energy is calculated to be

$$\epsilon_{YK2} = -2s - 4/(s + s'). \quad (21.24)$$

A similar consideration to that made for YK1 yields a stability region for YK2 as characterized by $s' < s$ and $s' + s > 1$ and shown also in Fig. 9. Since the three regions cover the whole s, s' plane, no other spin configurations are conceivable as long as \mathbf{k} is considered parallel to [001].

(2) $\mathbf{k} = (k, 0, 0)$. Similar calculations can be carried out for this case. Under the same transformation (21.7), the variables separate into (x_1, x_5) , x_4 , (x_2, x_6) , x_3 , and we can put $x_2 = x_6 = x_3 = 0$, since the quadratic form of the first three variables in the energy expression ϵ_k goes over to that of the last three variables when k is replaced by $k + \pi$. The lower eigenvalue for (x_1, x_5) is calculated to be

$$\begin{aligned} \lambda_1(k, \beta) = \frac{1}{4}\{s' + (s + s') \cos k - [(s' + (s + s') \cos k)^2 \\ + 16\beta^2(1 + \cos k)(w + (1 - w) \cos k)^2]^{1/2}\} \end{aligned} \quad (21.25)$$

and the eigenvalue for x_4 to be

$$\lambda_2(k) = -\frac{1}{2}[s' + (s - s') \cos k]. \quad (21.26)$$

These eigenvalues are connected with the previous $\lambda_1(k, \beta)$ and λ_2 , respectively, at $k = 0$. It can be shown that λ_N , λ_{YK1} , and λ_{YK2} are the lowest eigenvalue in the respective regions of Fig. 9 when compared with (21.25) and (21.26), β being put equal to β_N , β_{YK1} , and β_{YK2} , respectively. The study of the present case thus adds no new results.

(3) $\mathbf{k} = (h, h, 0)$, $h = k/2^{1/2}$. This case cannot be treated easily.

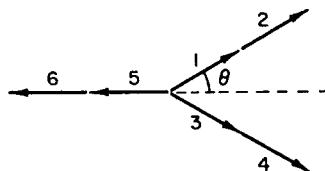


FIG. 11. Sublattice magnetizations in the Yafet-Kittel 2 configuration; $\tan \theta = [(s + s')^2 - 1]^{1/2}$.

Here we will mention only a simple conclusion. Then in Section 22, we shall discuss the results obtained by Kaplan and his co-workers by extensive calculations. The fact to be mentioned is that the stability regions shown in Fig. 9 for the Néel and Yafet-Kittel spin configurations become narrowed by some energetically favorable states having propagation vectors included within the present case.

In the case at hand, the variables x_i ($i = 1, 2, \dots, 6$) separate into two sets, (x_1, x_3, x_5) and (x_2, x_4, x_6) . A further transformation,

$$\begin{aligned} y_1 &= (x_1 + x_3)/2^{1/2} = (u_1 + u_3)/2^{1/2}, & y_2 &= (x_2 + x_4)/2^{1/2} = (u_1 - u_3)/2^{1/2}, \\ y_3 &= (x_1 - x_3)/2^{1/2} = (u_2 + u_4)/2^{1/2}, & y_4 &= (x_2 - x_4)/2^{1/2} = (u_2 - u_4)/2^{1/2}, \end{aligned} \quad (21.27)$$

separates the (x_2, x_4, x_6) -space into a two-dimensional space and a one-dimensional space; also, it simplifies the energy expression for (x_1, x_3, x_5) . The calculated energy matrices are as follows. For (y_1, y_3, x_5) one has

$$\begin{bmatrix} \frac{1}{2}s \cos 2h & s' \cos h & \cos h \\ s' \cos h & \frac{1}{2}s & w + (1-w) \cos 2h \\ \cos h & w + (1-w) \cos 2h & 0 \end{bmatrix}; \quad (21.28)$$

for y_4 , $(-\frac{1}{2}s)$; and for (y_2, x_6) ,

$$\begin{bmatrix} -\frac{1}{2}s \cos 2h & i(1-2w) \sin h \\ -i(1-2w) \sin h & 0 \end{bmatrix}. \quad (21.29)$$

From the one-dimensional matrix, we obtain an eigenvalue $\lambda_2 = -\frac{1}{2}s$, which is identical with (21.14). For the two-dimensional matrix, we may introduce β^2 as in (21.12), and we have

$$\begin{vmatrix} -\frac{1}{2}s \cos 2h - \lambda & i(1-2w) \sin h \\ -i(1-2w) \sin h & -\lambda/\beta^2 \end{vmatrix} = 0, \quad (21.30)$$

$$\lambda_3 = \frac{1}{4}\{-s \cos 2h - [s^2 \cos^2 2h + 16\beta^2(1-2w)^2 \sin^2 h]^{1/2}\}. \quad (21.31)$$

This λ_3 becomes equal to λ_2 at $h = 0$ when s is positive. It can be shown again that for $h \neq 0$ this eigenvalue is higher than the eigenvalues for the Néel, YK1, and YK2 configurations in the respective regions of Fig. 9, provided β takes its respective values.

We have thus to study the three-dimensional matrix (21.28). If we add $-\lambda$, $-\lambda$, and $-\lambda/\beta^2$ to the diagonal elements of (21.28)—since β_i 's for x_i 's ($i = 1, 2, \dots, 6$) have been assumed hitherto to be $1, 1, 1, 1, \beta, \beta$ —

and calculate the determinant by putting λ and β equal to λ_N , β_N or λ_{YK1} , β_{YK1} or λ_{YK2} , β_{YK2} , and see if the determinant is positive or not, then we shall be able to see whether the stability regions of Fig. 9 become narrowed or not. For simplicity, we confine ourselves to $h = \pi/4$. The point $(\pi/4, \pi/4, 0)$ is the projection of the center of a hexagon of the Brillouin zone boundary on the (001) plane. It turns out that the determinant is no longer positive in the shaded region of Fig. 12, which is bounded by a hyperbola

$$ss' + s'^2 - (1 - \frac{1}{2}w^2)s - (\frac{7}{2} - w - w^2)s' + (\frac{3}{2} - w^2) = 0$$

and two straight lines

$$(1 + w)s - 2ws' = 0, \quad (1 + w)s - 2s' = 0.$$

We may therefore have to look for some other structure in this shaded region.

Menyuk *et al.*^{22e} investigated in detail the stability regions for the Néel, YK1, and YK2 configurations by examining many more \mathbf{k} vectors, in particular those parallel to the symmetry directions $\langle 001 \rangle$, $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 101 \rangle$, and a special \mathbf{k} vector at the edge of the Brillouin zone in the $\langle 201 \rangle$ direction. Those \mathbf{k} vectors which are perpendicular to the c axis and are in the neighborhood of the origin were also examined. Furthermore, for certain ranges of the parameters, all \mathbf{k} vectors in the first Brillouin zone were examined. These authors also investigated the "destabilizing wave vector" on the boundary of the stability region, namely, the wave vector for which the determinant vanishes and which is thus suggestive of a spin configuration outside that region. Much computer work was

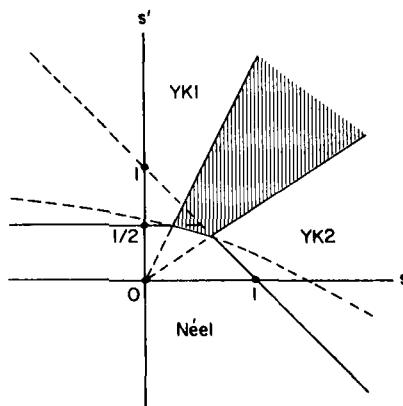


FIG. 12. Stability regions for the Néel, YK1, and YK2 configurations, narrowed by the consideration of a wave vector $(\pi/4, \pi/4, 0)$. In the shaded region, one may have some other spin configurations. The figure was drawn for $w = \frac{1}{2}$.

necessary for this study, and the results are not so simple as to be described here briefly. The reader is referred to the original paper.

22. MULTIPLE CONE STRUCTURE

We have seen that in the shaded region of Fig. 12 (and possibly in a wider region) the stability of the three spin configurations studied is not ensured. Along the *cubic* line $s = s'$, a multiple cone structure possessing a zero wave vector and a nonzero wave vector \mathbf{Q} parallel to $\langle 110 \rangle$ seems to exist as the stable configuration, as discussed by Kaplan and co-workers.^{22e} They showed that the Néel structure is stable for $u (=2s) \gtrless \frac{8}{3}$, and a multiple cone structure is more stable than the previous three spin configurations and in fact more stable than any other spin configurations that possess "equal relative angles," i.e., where $\mathbf{S}_{m\mu} \cdot \mathbf{S}_{n\nu}$ is invariant under lattice translations. Also, a multiple cone structure was shown to be locally stable (stable against small spin deviations) for $\frac{8}{3} < u < u'' = 1.298$ and locally unstable for $u > u''$. Extension of the calculation beyond u'' showed that the structure was coplanar for $u > 3.817$. Kaplan and co-workers report, however, that for all $u > \frac{8}{3}$ there are some wave vectors \mathbf{q} in the Brillouin zone for which the inequality $\lambda_0(\mathbf{Q}) < \lambda_\alpha(\mathbf{q})$ is *not* satisfied. This would mean that the Lyons-Kaplan stability criterion cannot be applied to the multiple cone structure and helical structure studied, so that one does not know whether these structures are stable or not. In this study, nearest-neighbor interaction and the $A-A$ interaction were neglected.

The multiple cone structure can be expressed by

$$\mathbf{S}_{n\nu} = S_\nu \{ [i \cos (\mathbf{Q} \cdot \mathbf{R}_{n\nu} + \gamma_\nu) + j \sin (\mathbf{Q} \cdot \mathbf{R}_{n\nu} + \gamma_\nu)] \sin \phi_\nu + k \cos \phi_\nu \}, \quad (22.1)$$

where i, j, k are orthogonal unit vectors. Corresponding to (22.1), we have

$$u_{Q\nu} = \sin \phi_\nu \exp(i\gamma_\nu), \quad u_{0\nu} = \cos \phi_\nu. \quad (22.2)$$

(Our numbering of 1, 2, ..., 6 is different from that of Kaplan and co-workers; see footnote 35b on page 381.) The three-dimensional matrix of (21.28) can be written in the present cubic case as

$$\begin{bmatrix} \frac{1}{2}s \cos 2h & s \cos h & \cos h \\ s \cos h & s/2 & \frac{2}{3} \cos 2h + \frac{1}{3} \\ \cos h & \frac{2}{3} \cos 2h + \frac{1}{3} & 0 \end{bmatrix}. \quad (22.3)$$

The corresponding secular equation, obtained by adding $-\lambda$, $-\lambda/\alpha^2$, and

$-\lambda/\beta^2$ along the diagonal, will yield an eigenvalue $\lambda_1(h, \alpha, \beta)$. The condition for a minimum, $\partial\lambda_1(h, \alpha, \beta)/\partial h = 0$, the degeneracy condition, $\lambda_1(0, \alpha, \beta) = \lambda_1(h, \alpha, \beta)$, and three more relations, $|u_{h\nu}|^2 + |u_{0\nu}|^2 = 1$ ($\nu = 1, 2, 5$) will determine h , α , β , and the amplitudes of the two eigenvectors, and, thus, ϕ , and γ_ν . [It is noted that this procedure is equivalent to minimizing the quadratic form of the energy, constructed from the preceding matrix with variables y_1 , y_3 , and x_5 and summed over $h = h$ and $h = 0$, under the conditions (19.8) in which \mathbf{q} and \mathbf{q}' are replaced by h and 0, respectively.] If we restrict γ_ν 's within $(0, \pi)$, we can see quite easily from the energy expression that $\gamma_2 - \gamma_1 = \gamma_5 - \gamma_1 = \pi$, provided s (or $u = 2s$) is positive, so that the helical component in the multiple cone structure rotates by an angle of $\pi - h$ in going from one (110) atomic plane to the next. To visualize this situation, one may refer to Fig. 8.

According to the computation carried out by Kaplan and co-workers, h (their ρ) stays nearly at a constant value of about 0.92 for $\frac{8}{9} < u < 3.817$ and then increases slightly for $u > 3.817$. They also give ϕ_ν and γ_ν as functions of u (Fig. 2 in Menyuk *et al.*^{22e}). The axial components of the three sublattices, specified by (1, 3), (2, 4), and (5, 6), are arranged more or less as in the Néel configuration.

An interesting point reported is that at $u = 2$ the eigenvalue $\lambda_2 = -s/2$, given before (21.29) [see also (21.14)], becomes degenerate with $\lambda_1(0, \alpha, \beta)$ and $\lambda_1(h, \alpha, \beta)$, and for $u > 2$ no real values of α and β are found. In order to solve this situation, Kaplan and co-workers chose α and β so as to retain the threefold degeneracy $\lambda_1(0, \alpha, \beta) = \lambda_1(h, \alpha, \beta) = \lambda_2$ for $u \geq 2$, together with the condition $\partial\lambda_1(h, \alpha, \beta)/\partial h = 0$. A superposition of the three corresponding eigenvectors was made to construct the spin configuration. At $u = 3.817$, however, the coefficient of the eigenvector associated with $\lambda_1(0, \alpha, \beta)$ vanished, so that only $\lambda_1(h, \alpha, \beta) = \lambda_2$ and $\partial\lambda_1(h, \alpha, \beta)/\partial h = 0$ were retained for $u \geq 3.817$.^{35f}

^{35f} The mixing of the eigenvector associated with λ_2 would result in the splitting of the (2, 4) sublattice into the (2) sublattice and the (4) sublattice. This is because the eigenvector associated with λ_2 consists only of y_4 which means that the only non-vanishing u_ν 's are u_2 and u_4 , related to each other by $u_2 = -u_4$, where as the eigenvector associated with $\lambda_1(h, \alpha, \beta)$, with either $h = 0$ or $h \neq 0$, consists of y_1 , y_3 , and y_5 giving nonvanishing $u_1 = u_3$, $u_2 = u_4$, and $u_6 = u_5$. If the wave number associated with λ_2 is assumed to be zero, the superposition would give a difference in the axial component of the spins on the (2) sublattice and that on the (4) sublattices, but then the geometrical condition stating that the spin length should be the same on the two sublattices would be violated. If the wave number associated with λ_2 is assumed to be h , and if u_2 of $\lambda_1(h, \alpha, \beta)$ and u_2 of λ_2 are assumed to have a phase difference of $\pi/2$, then the spins on the two sublattices would describe similar cones having the same half-cone angle, and the geometrical condition mentioned would be satisfied. For $u > 3.817$, in particular, one would have similar helices (the half-cone angle being $\pi/2$), since the eigenvector associated with $\lambda_1(0, \alpha, \beta)$ is lacking.

VIII. Néel Temperature and Spin Ordering for Complex Lattices

The Néel temperature, T_N , and the mode of spin ordering immediately below T_N have been investigated by Kaplan and co-workers^{22d,f} for a general complex lattice and, in particular, for normal cubic spinels.

Let $\mathbf{H}_{n\nu}$ be the exchange field acting on spin $\mathbf{S}_{n\nu}$. $\mathbf{H}_{n\nu}$ may be assumed to be small immediately below T_N . Then for the thermal average of $\mathbf{S}_{n\nu}$ one should have (in the molecular field approximation)

$$\langle \mathbf{S}_{n\nu} \rangle = [S_\nu(S_\nu + 1)/3kT_N] \mathbf{H}_{n\nu}. \quad (\text{VIII.1})$$

On the other hand, one has

$$\mathbf{H}_{n\nu} = 2 \sum_{m\mu} J_{m\mu, n\nu} \langle \mathbf{S}_{m\mu} \rangle. \quad (\text{VIII.2})$$

By Fourier transformations,

$$\langle \mathbf{S}_{n\nu} \rangle = S_\nu \sum_{\mathbf{q}} \langle \mathbf{d}_{\mathbf{q}\nu} \rangle \exp(i\mathbf{q} \cdot \mathbf{R}_n), \quad (\text{VIII.3})$$

$$\mathbf{H}_{n\nu} = \sum_{\mathbf{q}} \mathbf{H}_{\mathbf{q}\nu} \exp(i\mathbf{q} \cdot \mathbf{R}_n), \quad (\text{VIII.4})$$

Eq. (VIII.2) changes into

$$\mathbf{H}_{\mathbf{q}\nu} = -(2/S_\nu) \sum_{\mu} A_{\mu\nu}(\mathbf{q}) \langle \mathbf{d}_{\mathbf{q}\mu} \rangle, \quad (\text{VIII.5})$$

where $A_{\mu\nu}(\mathbf{q})$ is defined by (VI.5). Hence, (VIII.1) becomes

$$\langle \mathbf{d}_{\mathbf{q}\nu} \rangle = -[2S_\nu(S_\nu + 1)/3kT_N S_\nu^2] \sum_{\mu} A_{\mu\nu}(\mathbf{q}) \langle \mathbf{d}_{\mathbf{q}\mu} \rangle, \quad (\text{VIII.6})$$

or, written in a symmetrical form,

$$\left[\frac{S_\nu}{S_\nu + 1} \right]^{1/2} \langle \mathbf{d}_{\mathbf{q}\nu} \rangle = -\frac{2}{3kT_N} \sum_{\mu} \left[\frac{S_\mu + 1}{S_\mu} \frac{S_\nu + 1}{S_\nu} \right]^{1/2} A_{\mu\nu}(\mathbf{q}) \left[\frac{S_\mu}{S_\mu + 1} \right]^{1/2} \langle \mathbf{d}_{\mathbf{q}\mu} \rangle.$$

Thus, $\lambda = -(\frac{3}{2})kT_N$ must be an eigenvalue of the matrix

$$B_{\mu\nu}(\mathbf{q}) = \left[\frac{S_\mu + 1}{S_\mu} \frac{S_\nu + 1}{S_\nu} \right]^{1/2} A_{\mu\nu}(\mathbf{q}), \quad (\text{VIII.7})$$

and in fact the lowest eigenvalue determines T_N and simultaneously the value of \mathbf{q} . Equation (VIII.6) will then determine the mode of spin ordering immediately below T_N .

In the case of normal cubic spinels, Kaplan and co-workers^{22d} find that for $u \leq 2.177$ the eigenvalue giving rise to the Néel-type spin configuration is the absolute minimum over all α (which specifies different eigenvalues) and \mathbf{q} . For $u > 2.177$, the minimum eigenvalue of the matrix (VIII.7)

yields a nonzero wave vector along [110], the corresponding spin configuration being a [110] helix, and this eigenvalue is the absolute minimum over all α and \mathbf{q} . Since the Néel configuration is stable at $T = 0$ only for $u < \frac{8}{9}$ and a multiple cone structure has a lower energy for $\frac{8}{9} < u < 1.298$ and is probably the stable configuration at $T = 0$ as mentioned in Section 22, it appears that for the latter range of u there will be a phase transition between T_N and $T = 0$. For $1.298 < u < 3.817$, the multiple cone structure has no local stability and hence is unstable at $T = 0$, and a certain deviated structure should be the stable configuration. Since neither the Néel structure nor a helix is stable at $T = 0$ in this range of u , there will also be a phase transition between T_N and $T = 0$.

Equation (VIII.1) is based on the molecular field approximation. A rigorous equation to determine the Néel temperature and the mode of spin ordering immediately below it may be derived in the following way. Let $\mathbf{H}_v(\mathbf{r})$ be a fictitious magnetic field acting only on the v th sublattice and varying with position \mathbf{r} , and $\mathbf{H}_{qv} \exp(i\mathbf{q} \cdot \mathbf{r})$ be a Fourier component of it. For convenience, one may assume that all \mathbf{H}_{qv} ($v = 1, 2, \dots, k$) point in the same direction, since one is dealing with a spin system having isotropic exchange interactions. The interaction between the set of Fourier components $\mathbf{H}_{qv} \exp(i\mathbf{q} \cdot \mathbf{r})$ ($v = 1, 2, \dots, k$) and the spin system will be

$$-\sum_v \sum_n \mathbf{S}_{nv} \cdot \mathbf{H}_{qv} \exp(i\mathbf{q} \cdot \mathbf{R}_{nv}) = -N \sum_v \mathbf{S}_{qv}^* \cdot \mathbf{H}_{qv} \equiv \mathcal{K}', \quad (\text{VIII.8})$$

where \mathbf{S}_{qv} is the Fourier transform of \mathbf{S}_{nv} and N the number of unit cells. The thermal average of the component of \mathbf{S}_{nv} in the direction of \mathbf{H}_{qv} will be

$$\left\langle \frac{\mathbf{S}_{qv} \cdot \mathbf{H}_{qv}}{H_{qv}} \right\rangle = \frac{\text{tr} (\mathbf{S}_{qv} \cdot \mathbf{H}_{qv} / H_{qv}) \exp[-\beta(\mathcal{K}_0 + \mathcal{K}')] }{\text{tr} \exp[-\beta(\mathcal{K}_0 + \mathcal{K}')]}, \quad (\text{VIII.9})$$

where \mathcal{K}_0 is the exchange energy. In the temperature range of vanishing spin order, one has $\text{tr} \mathbf{S}_{qv} \exp[-\beta\mathcal{K}_0] = 0$, so that to the first order in \mathcal{K}' ,

$$\begin{aligned} \langle \mathbf{S}_{qv} \cdot \mathbf{H}_{qv} / H_{qv} \rangle &= -\text{tr} \exp(-\beta\mathcal{K}_0) \int_0^\beta \exp(\lambda\mathcal{K}_0) \mathcal{K}' \\ &\quad \times \exp(-\lambda\mathcal{K}_0) (\mathbf{S}_{qv} \cdot \mathbf{H}_{qv} / H_{qv}) d\lambda / [\text{tr} \exp(-\beta\mathcal{K}_0)]. \end{aligned}$$

If m and n are eigenstates of \mathcal{K}_0 which are connected by \mathcal{K}' and E_m and E_n are eigenvalues of \mathcal{K}_0 , and if one assumes $|E_m - E_n| \ll \beta^{-1} = kT$, then the preceding expression will become

$$\begin{aligned} &-\sum_m \sum_n \exp(-\beta E_m) \frac{\exp[\beta(E_m - E_n)] - 1}{E_m - E_n} \\ &\quad \times (m | \mathcal{K}' | n) (n | \mathbf{S}_{qv} \cdot \mathbf{H}_{qv} / H_{qv} | m) / [\sum_m \exp(-\beta E_m)], \end{aligned}$$

or

$$-\beta \sum_m \sum_n \exp(-\beta E_m) \\ \times (m | \mathcal{H}' | n) (n | \mathbf{S}_{q\nu} \cdot \mathbf{H}_{q\nu} / H_{q\nu} | m) / [\sum_m \exp(-\beta E_m)].$$

By virtue of (VIII.8), this can be written as

$$N\beta \sum_\mu \langle (\mathbf{S}_{q\mu}^* \cdot \mathbf{H}_{q\mu}) (\mathbf{S}_{q\nu} \cdot \mathbf{H}_{q\nu}) \rangle / H_{q\nu},$$

or

$$\frac{1}{3} N\beta \sum_\mu \langle \mathbf{S}_{q\mu}^* \cdot \mathbf{S}_{q\nu} \rangle H_{q\mu},$$

where the thermal average is taken over the canonical ensemble for \mathcal{H}_0 . Thus, $\frac{1}{3} N \langle \mathbf{S}_{q\mu}^* \cdot \mathbf{S}_{q\nu} \rangle$ is the \mathbf{q} -dependent susceptibility matrix and may be written as $\chi_{\mu\nu}(\mathbf{q})$, and the left-hand side of Eq. (VIII.9), i.e., the induced moment, may be written as

$$M_{q\nu} = \sum_\mu \chi_{\mu\nu}(\mathbf{q}) H_{q\mu}, \quad \text{where } \chi_{\mu\nu}(\mathbf{q}) = \frac{1}{3} N \beta \langle \mathbf{S}_{q\mu}^* \cdot \mathbf{S}_{q\nu} \rangle. \quad (\text{VIII.10})$$

If we denote by $\chi_{\mu\nu}(\mathbf{q})^{-1}$ the reciprocal susceptibility matrix, (VIII.10) can be written also as

$$\sum_\nu M_{q\nu} \chi_{\nu\mu}(\mathbf{q})^{-1} = H_{q\mu}. \quad (\text{VIII.11})$$

The Néel temperature is the temperature at which infinitesimal magnetization sets in for vanishing field. Hence it is determined from

$$\det |\chi_{\mu\nu}(\mathbf{q})^{-1}| = 0 \quad \text{or} \quad \det |\chi_{\mu\nu}(\mathbf{q})| = \infty. \quad (\text{VIII.12})$$

This mode of spin ordering setting in at T_N is determined from (VIII.10) with $H_{q\mu} = 0$. (VIII.11) means that the largest eigenvalue of the matrix $\chi_{\mu\nu}(\mathbf{q})$ becomes infinite at T_N , and of course the value of \mathbf{q} which gives the highest T_N should be adopted. This value of \mathbf{q} will be the wave vector of the spin ordering setting in at T_N . If there are degenerate eigenvalues associated with different values of \mathbf{q} , one will have a conical or some other spin ordering.

Since

$$\mathbf{S}_{q\nu} = N^{-1} \sum_n \mathbf{S}_{n\nu} \exp(-i\mathbf{q} \cdot \mathbf{R}_{n\nu})$$

($\mathbf{R}_{n\nu} = \mathbf{R}_n + \mathbf{R}_\nu$ is used here instead of \mathbf{R}_n for the sake of convenience), the expression for the susceptibility given in (VIII.9) can be rewritten as

$$\chi_{\mu\nu}(\mathbf{q}) = (\beta/3N) \sum_m \sum_n \langle \mathbf{S}_{m\mu} \cdot \mathbf{S}_{n\nu} \rangle \exp(i\mathbf{q} \cdot \mathbf{R}_{m\mu, n\nu})$$

$$= \frac{S_\nu(S_\nu + 1)}{3kT} \delta_{\mu\nu} + \frac{1}{3kT} \sum_m' \langle \mathbf{S}_{m\mu} \cdot \mathbf{S}_{n\nu} \rangle \exp(i\mathbf{q} \cdot \mathbf{R}_{m\mu, n\nu}). \quad (\text{VIII.13})$$

The first term represents the Curie law. The second term arises from correlations between different spins, and a prime attached to the summation symbol means that $m = n$ is excluded when $\mu = \nu$, i.e., the correlation in the same atom has been separated as the first term. The second term may be calculated approximately by an expansion in powers of β . The calculation to first order in β with the use of (VI.1) (which is \mathcal{C}_0) and (VI.5) yields the following result:

$$\chi_{\mu\nu}(\mathbf{q}) = \frac{S_\nu(S_\nu + 1)}{3kT} \delta_{\mu\nu} - \frac{2(S_\mu + 1)(S_\nu + 1)}{(3kT)^2} C_{\mu\nu}(\mathbf{q}), \quad (\text{VIII.14})$$

where

$$C_{\mu\nu}(\mathbf{q}) = A_{\mu\nu}(\mathbf{q}) \exp[i\mathbf{q} \cdot (\mathbf{R}_\mu - \mathbf{R}_\nu)]. \quad (\text{VIII.15})$$

The determinant of $\chi_{\mu\nu}(\mathbf{q})$ does not become infinite. The usual mathematical trick in such a case is to calculate the reciprocal susceptibility to first order in β and to derive the Néel temperature from the vanishing of it. (VIII.14) can be written in a more symmetrical form:

$$\chi_{\mu\nu}(\mathbf{q}) = \{[S_\mu(S_\mu + 1)S_\nu(S_\nu + 1)]^{1/2}/3kT\}\{\delta_{\mu\nu} - (2/3kT)D_{\mu\nu}(\mathbf{q})\},$$

where

$$D_{\mu\nu}(\mathbf{q}) = \left[\frac{S_\mu + 1}{S_\mu} \frac{S_\nu + 1}{S_\nu} \right]^{1/2} C_{\mu\nu}(\mathbf{q}). \quad (\text{VIII.16})$$

Hence the reciprocal susceptibility is obtained to be

$$\chi_{\mu\nu}(\mathbf{q})^{-1} = \{3kT/[S_\mu(S_\mu + 1)S_\nu(S_\nu + 1)]^{1/2}\}\{\delta_{\mu\nu} + (2/3kT)D_{\mu\nu}(\mathbf{q})\}. \quad (\text{VIII.17})$$

The Néel temperature is obtained from

$$\det |\delta_{\mu\nu} + (2/3kT)D_{\mu\nu}(\mathbf{q})| = 0. \quad (\text{VIII.18})$$

Since $D_{\mu\nu}(\mathbf{q})$ is a matrix equivalent to $B_{\mu\nu}(\mathbf{q})$ defined by (VIII.7), Eq. (VIII.18) is the same as that we derived earlier in the molecular field approximation.

Kaplan *et al.*^{22f} carried out a calculation up to β^3 and obtained results that suggest that one can expect to have the Néel ordering for values of u much greater than that predicted by the molecular-field approximation.^{35g}

IX. Neutron Diffraction: Theory and Examples

Neutron scattering experiments provide the most powerful means of determining an ordered spin arrangement in a crystal, as well as of ob-

^{35g} Details have not yet been published.

serving the spin-wave spectrum and correlations between neighboring spins. In the present part, we wish to give a brief discussion of diffraction lines from an ordered spin arrangement, in particular a helical order, and to mention a few interesting observations reported.

23. GENERAL THEORY OF ELASTIC NEUTRON SCATTERING

According to the standard theory,^{36,37} the amplitude of the neutron wave elastically scattered from an atom at the origin by the interaction between the magnetic moment of the neutron and that of the atom is given, apart from a universal constant, by

$$(1/r) \exp(ik'r)[\mathbf{P}_{\text{spin}} - \mathbf{e}(\mathbf{P}_{\text{spin}} \cdot \mathbf{e}) + (i/\kappa)\mathbf{e} \times \mathbf{P}_{\text{orb}}] \cdot \mathbf{s}\chi, \quad (23.1)$$

with

$$\mathbf{e} = \kappa/\kappa, \quad \kappa = \mathbf{k} - \mathbf{k}', \quad |\mathbf{k}| = |\mathbf{k}'| = k',$$

$$\mathbf{P}_{\text{spin}} = (\psi^*, \sum_j \mathbf{s}_j \exp(i\kappa \cdot \mathbf{r}_j)\psi),$$

$$\mathbf{P}_{\text{orb}} = (1/2i)[(\psi^*, \sum_j \exp(i\kappa \cdot \mathbf{r}_j)\nabla_j\psi) - (\psi, \sum_j \exp(i\kappa \cdot \mathbf{r}_j)\nabla_j\psi^*)],$$

where \mathbf{s} is the spin vector of the neutron, χ the spin function of the *incident* neutron, \mathbf{s}_j and \mathbf{r}_j the spin and position vectors of the j th electron in the atom, and ψ the electronic wave function of the atom. From (23.1), it is seen that only the components of \mathbf{P}_{spin} and of \mathbf{P}_{orb} perpendicular to κ are effective in the scattering, κ being the difference between the wave vector \mathbf{k} of the incident neutron and the wave vector \mathbf{k}' of the scattered neutron. When the scattering atom is at position \mathbf{R}_{nv} , a factor $\exp(i\kappa \cdot \mathbf{R}_{nv})$ will separate from \mathbf{P}_{spin} and \mathbf{P}_{orb} , so that one will have, for the amplitude of the scattered wave from a crystal,

$$\sum_{n,v} \exp(i\kappa \cdot \mathbf{R}_{nv}) \mathbf{P}_{nv} \cdot \mathbf{s}\chi, \quad (23.2)$$

where \mathbf{P}_{nv} is the value of $\mathbf{P}_{\text{spin}} - \mathbf{e}(\mathbf{P}_{\text{spin}} \cdot \mathbf{e}) + (i/\kappa)\mathbf{e} \times \mathbf{P}_{\text{orb}}$ associated with the atom at \mathbf{R}_{nv} , the origin of the electron positions being taken at the nucleus of that atom. This \mathbf{P}_{nv} is in essence (apart from the magnetic form factor) the projection of the magnetic moment of the atom at \mathbf{R}_{nv} on the plane perpendicular to the scattering vector κ .

Now, consider that \mathbf{P}_{nv} varies sinusoidally with position \mathbf{R}_{nv} as

$$\mathbf{P}_{nv} = [P_{nx} \cos(Q \cdot \mathbf{R}_{nv} + \alpha_v), P_{ny} \sin(Q \cdot \mathbf{R}_{nv} + \alpha_v)], \quad (23.3)$$

where the x and y axes are taken perpendicular to κ . Then (23.2) becomes

$$\sum_{n,v} \exp(i\kappa \cdot \mathbf{R}_{nv}) [P_{nx}s_x \cos(Q \cdot \mathbf{R}_{nv} + \alpha_v) + P_{ny}s_y \sin(Q \cdot \mathbf{R}_{nv} + \alpha_v)]\chi,$$

³⁶ O. Halpern and M. H. Johnson, *Phys. Rev.* **55**, 898 (1939).

³⁷ G. T. Trammell, *Phys. Rev.* **92**, 1387 (1953).

or

$$\begin{aligned} \frac{1}{2} \sum_{\nu} \sum_n \exp [i(\kappa + Q) \cdot R_{\nu\nu} + i\alpha_{\nu}] (P_{\nu x} s_x - i P_{\nu y} s_y) \chi \\ + \frac{1}{2} \sum_{\nu} \sum_n \exp [i(\kappa - Q) \cdot R_{\nu\nu} - i\alpha_{\nu}] (P_{\nu x} s_x + i P_{\nu y} s_y) \chi. \end{aligned} \quad (23.4)$$

The first term of (IX.4) gives an interference condition

$$\kappa + Q = K \quad \text{or} \quad \mathbf{k}' = \mathbf{k} + \mathbf{Q} - \mathbf{K} \quad (23.5)$$

and the second term another interference condition

$$\kappa - Q = K \quad \text{or} \quad \mathbf{k}' = \mathbf{k} - \mathbf{Q} - \mathbf{K}, \quad (23.6)$$

where \mathbf{K} is a reciprocal lattice vector. These conditions were first derived by Yoshimori.¹ Thus, for $Q \neq 0$, magnetic reflections will appear at positions different from those of nuclear reflections, and in fact the former appear as satellites of the latter. If there are a number of wave vectors coexisting in the spin arrangement, one should of course have as many satellites as there are wave vectors.

Taking the quantization axis of the neutron spin to be parallel to the scattering vector κ , we write the spin function of the incident neutron as

$$\chi = a\alpha + b\beta, \quad (23.7)$$

with

$$a = \cos (\theta/2) \exp (-i\phi/2), \quad b = \sin (\theta/2) \exp (i\phi/2), \quad (23.8)$$

where θ and ϕ are the polar and azimuthal angles of the incident polarization. Then (23.4) becomes, by virtue of (23.5) or (23.6), either

$$\frac{1}{4} N \sum_{\nu} [P_{\nu x}(a\beta + b\alpha) + P_{\nu y}(a\beta - b\alpha)] \exp (i\mathbf{K} \cdot \mathbf{R}_{\nu} + i\alpha_{\nu}) \quad (23.9)$$

or

$$\frac{1}{4} N \sum_{\nu} [P_{\nu x}(a\beta + b\alpha) - P_{\nu y}(a\beta - b\alpha)] \exp (i\mathbf{K} \cdot \mathbf{R}_{\nu} - i\alpha_{\nu}). \quad (23.10)$$

\mathbf{R}_{ν} is the position of the ν th atom in the zeroth unit cell. The squares of the absolute values of these quantities, summed over the spin variable, give the intensities of the satellites. For unpolarized beam, $|a|^2$ and $|b|^2$ are equal on the average, so that (23.9) and (23.10) give intensities that are proportional to

$$|\sum_{\nu} P_{\nu x} \exp (i\mathbf{K} \cdot \mathbf{R}_{\nu} \pm i\alpha_{\nu})|^2 + |\sum_{\nu} P_{\nu y} \exp (i\mathbf{K} \cdot \mathbf{R}_{\nu} \pm i\alpha_{\nu})|^2. \quad (23.11)$$

When α_{ν} is independent of ν , the two satellites corresponding to the plus and minus signs of (23.11) have the same intensity, aside from a minute difference in the form factor due to the different values of κ .

If the incident beam is polarized parallel to the x -axis, the x -component $P_{\nu x}$ will not alter the polarization, but the y -component $P_{\nu y}$ will reverse the polarization direction, as can be seen directly from (23.4). If the incident polarization is parallel to the scattering vector, i.e., $a = 1$ and $b = 0$, the polarization will be reversed, as can be seen from (23.9) and (23.10), and one will have from (23.9) and (23.10) the intensities proportional to

$$\left| \sum_{\nu} (P_{\nu x} \pm P_{\nu y}) \exp(i\mathbf{k} \cdot \mathbf{R}_{\nu} \pm i\alpha_{\nu}) \right|^2, \quad (23.12)$$

respectively. If, in particular, $P_{\nu x} = P_{\nu y}$ or $P_{\nu x} = -P_{\nu y}$, the ν th sublattice will not contribute to the intensity of $\mathbf{k}' = \mathbf{k} - \mathbf{Q} - \mathbf{K}$ or that of $\mathbf{k}' = \mathbf{k} + \mathbf{Q} - \mathbf{K}$, which means that one can, in principle, observe the sense of the spin rotation in a helical configuration.

Theories similar to that just described have been published in a number of papers. The readers will find references in a paper by Blume.³⁸

24. EXAMPLES OF HELICAL SPIN CONFIGURATION

1. Chromium is known to have a magnetically ordered phase below 310°K which is interpreted as being a "spin density wave" state.^{38a} The spin density varies sinusoidally with atomic positions, and this persists down to absolute zero. The wave vector is parallel to one of the cubic axes (the crystalline structure being bcc) and has a magnitude of $2\pi(1 - \frac{1}{25})/a$ (a : the lattice constant) at 20°C, for instance. Between 310° and 121°K, the spin vectors are perpendicular to the wave vector, whereas below 121°K they are parallel to the wave vector. With an unpolarized neutron beam, it is not possible to decide whether the spin configuration in the high-temperature phase is helical or linear-sinusoidal. Brown *et al.*³⁹ used a neutron beam polarized along one of the cubic axes and observed depolarization in the diffracted lines. When, for example, the incident polarization was parallel to [010] and the line $(0, 0, 1 - \frac{1}{25})$ was observed, the ratio R of the intensities of the line with unchanged and reversed polarizations was 1.20 ± 0.03 , which ruled out the simple helical model and suggested the existence of domains with linear-sinusoidal spins parallel to [010] and [100], in the ratio of 1.2 to 1.

2. Examples of helical or related spin order observed up to the present time (1966) are not many. MnO_2 was the first example observed or, rather, interpreted, as mentioned in Section 1. MnAu_2 , referred to in

³⁸ M. Blume, *Phys. Rev.* **130**, 1670 (1963).

^{38a} A discussion on the magnetic properties of Cr and the spin density wave will be given in the forthcoming Part 2.

³⁹ P. J. Brown, C. Wilkinson, J. B. Forsyth, and R. Nathans, *Proc. Phys. Soc. (London)* **85**, 1185 (1965).

Section 12, and a number of rare-earth metals, referred to in many of the preceding sections, will be discussed in some detail in the forthcoming Part 2. These systems provide most interesting examples. Helical and more complicated spin configurations in spinel lattices have been discussed in Chapters VII and VIII. MnI_2 , FeCl_3 , and solid solutions of Cr_2O_3 and Fe_2O_3 also present helical spin arrangements, which will be described below.

3. MnI_2 crystallizes in a hexagonal layer structure of the CdI_2 type, i.e., the iodine ions form a hexagonal close-packed $ABAB$ lattice, and the manganese ions penetrate into every other interlayer space to form hexagonal layers of the C type. Cable *et al.*⁴⁰ observed by neutron diffraction the magnetic order in this crystal. The Néel temperature is 3.40°K , and the magnetic order at 1.3°K is helical with a propagation vector pointing along the $\langle 307 \rangle$ direction, referred to the hexagonal unit cell, and a turn angle of $2\pi/16$. When the plane containing the moments was assumed normal to the helical axis, and a moment of $4.6\mu_B/\text{Mn}^{2+}$ was assumed, the calculated intensities of the Bragg lines were in good agreement with those observed. An interesting observation was that there was an apparent threefold symmetry about the c -axis in the reflections, which could be interpreted as being due to magnetic domains, which, in the absence of a magnetic field, grew with equal probability along three equivalent $\langle 307 \rangle$ axes. Application of a magnetic field favored that domain which had moments most nearly perpendicular to the field direction, and this domain grew at the expense of the other two until at saturation the entire crystal was transformed into a single domain. An attempt to interpret the observed magnetic order in terms of exchange interactions and anisotropy energies has been made by Moriya and the present writer,⁴¹ but it is not easy to attain an unambiguous conclusion.

4. FeCl_3 is another compound that shows a helical order at low temperatures. It crystallizes in a hexagonal layer structure of the BiI_3 type. The chlorine ions form a hexagonal close-packed $ABAB$ lattice, as iodine ions do in MnI_2 , and the ferric ions penetrate into every other interlayer space to form, in this case, honeycomb layers. The unit cell has a c axis that corresponds to three such honeycomb layers. The same authors⁴² also made neutron diffraction observations of the magnetic order. The Néel temperature is at $15 \pm 2^\circ\text{K}$. The moment ordering is helical with a propagation vector pointing along $\langle 140 \rangle$ and with a turn angle of $2\pi/15$. The moment vectors

⁴⁰ J. W. Cable, M. K. Wilkinson, E. O. Wollan, and W. C. Koehler, *Phys. Rev.* **125**, 1860 (1962).

⁴¹ K. Moriya and T. Nagamiya, *J. Phys. Soc. Japan* (1968) (to be published).

⁴² J. W. Cable, M. K. Wilkinson, E. O. Wollan, and W. C. Koehler, *Phys. Rev.* **127**, 714 (1962).

are normal to the propagation vector and have a magnitude of $4.3\mu_B/\text{Fe}^{3+}$. The formation of magnetic domains similar to that in MnI_2 was observed. 5. Cr_2O_3 and $\alpha\text{-Fe}_2\text{O}_3$ possess isomorphous crystalline structures of the rhombohedral corundum type. The oxygen ions form a deviated hexagonal close-packed lattice and the metallic ions, imbedded in it, form deviated honeycomb layers. Each metallic ion is surrounded by six oxygen ions, of which three in the same c plane form a smaller equilateral triangle than that in the regular hexagonal close-packed lattice, and the remaining three, in the opposite c plane, form a larger triangle. The metallic ion at the center is shifted along the c axis toward the center of the larger triangle. Since the larger triangle and smaller triangle are alternately arranged in the same c plane, each honeycomb layer is uneven. In the rhombohedral unit cell, there are four cations situated on the threefold axis, alternately apart and close, as $A_1 A_2 B_1 B_2$. The ordered magnetic structure in Cr_2O_3 below $T_N = 310^\circ\text{K}$ is such that these four cations have spins $(+-+-)$, the spin axis being parallel to the threefold axis. In this spin arrangement, each honeycomb layer is antiferromagnetic. In $\alpha\text{-Fe}_2\text{O}_3$, the ordered spin arrangement below $T_N \simeq 950^\circ\text{K}$ is of the type $(++--)$, which means that each honeycomb layer is ferromagnetic, and adjacent layers have opposite spins. The spin axis in Fe_2O_3 is perpendicular to the threefold axis down to about -15°C but becomes parallel to this axis below -15°C . As is known well, a weak ferromagnetism, due to the Dzyaloshinsky-Moriya interaction, is observed in the temperature range above -15°C .

Cr_2O_3 and $\alpha\text{-Fe}_2\text{O}_3$ crystallize as solid solutions over the entire range of composition. Cox *et al.*⁴³ prepared samples of $(1-x)\text{Cr}_2\text{O}_3 \cdot x\text{Fe}_2\text{O}_3$ for several values of x and made magnetic measurements, crystal parameter determination by X-rays, and spin structure determination by means of neutron diffraction. Starting from Cr_2O_3 , the addition of Fe_2O_3 creates a helically ordered spin component superposed on the Cr_2O_3 -type antiferromagnetically ordered component. Corresponding to the division of the Cr_2O_3 -type antiferromagnetic order into two ferromagnetically aligned sublattices, which are coupled antiferromagnetically with each other, the helical component is divided into two sets, which have opposite phases in the helical rotation (i.e., the phase constant α that appeared in Section 1 differs by π). The propagation vector is parallel to the c axis, and the helical moment vectors are perpendicular to the c axis. Hence, there is a conical spin arrangement on each of the two sublattices, the cones on one sublattice pointing oppositely to those on the other. The period in the helices decreases with increasing x (starting virtually from infinite period at $x = 0$), the magnitude of the helical component relative to that of the

⁴³ D. E. Cox, W. J. Takei, and G. Shirane, *J. Phys. Chem. Solids* **24**, 405 (1963).

antiferromagnetic component increases with x , and the Néel temperature decreases with x to about half of the initial value at $x = 0.2$.

When x exceeds about 0.2, the cone axis becomes perpendicular to the c axis, and at the same time the axial, antiferromagnetic component takes the Fe_2O_3 -type order. The propagation vector for the helical component remains parallel to the c axis, so that this component constitutes a cycloidal spin arrangement. With increasing x , up to $x = 0.35$ or 0.4, the period remains nearly constant, but the cone angle decreases, until finally, when this becomes zero, the Fe_2O_3 -type antiferromagnetic order is attained. All these are, however, the behavior at sufficiently low temperatures. The Néel temperature increases with increasing x . There appears the second critical temperature in the range $0.2 \leq x \leq 0.4$, below which the helical component develops, but above which there is a simple Fe_2O_3 -type spin arrangement. This critical temperature is also indicated by magnetic measurements. It appears to continue from the Néel temperature of the Cr_2O_3 -rich samples but decreases more slowly with increasing x . For $x > 0.4$, no helical component is detected. Weak ferromagnetism is observed over the whole range of $x > 0.25$ but appears to vanish at a particular temperature, which is the second critical temperature. For $x \leq 0.2$, no weak ferromagnetism is observed.

These interesting observations challenged theoreticians for interpretation. On the assumption of a uniform distribution of Cr^{3+} and Fe^{3+} and the consideration of four exchange constants, corresponding to apparently the most important superexchange paths, the theory^{44,45,45a} predicts only a pure helical configuration, other possibilities being the Cr_2O_3 -type and Fe_2O_3 -type antiferromagnetic configurations and the ferromagnetic alignment. [It may be mentioned in passing that recent measurements of the four exchange constants between Cr^{3+} ions in ruby ($\text{Al}_2\text{O}_3 + \text{Cr}$ impurities) by Mollenauer⁴⁶ with the prizospectroscopic method give values such that predict that Cr_2O_3 would have the Cr_2O_3 -type antiferromagnetic spin arrangement.] The theory is based on the Lyons-Kaplan method introduced in Section 19. The reason why a conical configuration cannot be obtained but only a pure helical configuration is predicted is as follows. When one considers cations and exchange interactions only and neglects oxygen ions, one can take a smaller rhombohedral unit cell containing two cations, say A_1 and A_2 (or B_1 and B_2). Since there is a point of in-

⁴⁴ E. F. Bertaut, *Proc. Intern. Conf. Magnetism, Nottingham, 1964*, p. 516. Inst. Phys. Phys. Soc., London, 1965.

⁴⁵ N. Menyuk and K. Dwight, *J. Phys. Chem. Solids* **25**, 1031 (1964).

^{45a} The present author has made an independent calculation: T. Nagamiya, unpublished report (1965).

⁴⁶ L. F. Mollenauer, Thesis, Stanford University (1965).

version symmetry midway between A_1 and A_2 , the exchange interactions within the A_1 sublattice and those within the A_2 sublattice are essentially the same; in particular, the Fourier transforms of the exchange constants defined in Section 16, $A_{11}(\mathbf{q})$ for the A_1 lattice and $A_{22}(\mathbf{q})$ for the A_2 lattice, are the same. In such a case, a conical spin configuration is impossible, as mentioned at the end of Section 18.

If there is a cation order in the solid solutions that destroys the inversion symmetry, so that the two sublattices are no longer equivalent, then one has the possibility of having a conical spin configuration. It is not possible to observe a cation order by X-rays. Nor was it detected by neutrons, since the calculated intensities are barely greater than the observable limit. Despite these facts, Cox *et al.* report that considerable order could exist. Theoretical consideration in such a case has not yet been given.

6. MnP is another interesting substance whose magnetic properties have become understood rather recently. It is ferromagnetic below 291°K and transforms to a metamagnetic phase at 50°K, as found by Huber and Ridgley⁴⁷ from magnetic measurements. By "metamagnetic" is meant the characteristic that the magnetization increases steeply in a certain range of applied magnetic field, approaching a near-saturation (as in FeCl_2 and MnAu_2). A theory for the electronic state in MnP and the like was proposed by Goodenough⁴⁸ and he predicted a helical spin ordering in the metamagnetic phase of MnP, which is somewhat different from that observed by neutron diffraction experiment to be mentioned below. Hirahara and co-workers⁴⁹ also imagined a helical spin configuration from the peculiar results of their torque measurements.

The crystal structure of MnP is orthorhombic ($a > b > c$ for convention). The atomic arrangement can be derived from that of NiAs by displacements of the atoms; the b and c axes in MnP correspond to the hexagonal c and a axes in NiAs, respectively, and the a axis in MnP corresponds to the direction perpendicular to both the c and a axes in NiAs; the manganese atoms, lined up along the b axis in MnP, are displaced parallel to the a axis in a zigzag way (by $\pm a/20$, and each line is actually displaced parallel to the b axis by $\pm b/200$), and the phosphorus atoms are displaced up and down parallel to the b axis (by $\pm 0.06b$); the orthorhombic unit cell contains four manganese atoms, and the manganese lattice can be subdivided into two similar, approximately body-centered, orthorhombic lattices (exactly body-centered if the displacements by $\pm b/200$ were zero); these sublattices are displaced relative to each other by $(a/10, b/2, 0)$.

⁴⁷ E. E. Huber and D. H. Ridgley, *Phys. Rev.* **135**, A1033 (1964).

⁴⁸ J. B. Goodenough, *J. Appl. Phys. Suppl.* **35**, 1083 (1964); J. B. Goodenough, *MIT Lincoln Lab. Tech. Rep.* 345 (1964).

⁴⁹ T. Komatsubara, K. Kinoshita, and E. Hirahara, *J. Phys. Soc. Japan* **20**, 2036 (1965).

Neutron diffraction experiments^{50,51} revealed that the spin arrangement at 4.2°K is helical, the spin vectors rotating in the bc plane with a propagation vector parallel to the a axis. The period of rotation is about $9a$ and the phase difference in the two body-centered sublattices is such that the spin at a corner site ($a/20, 0, 0$) of one sublattice is parallel to the spin at a body-center site ($9a/20, b/2, c/2$) of the other sublattice. We shall not mention here other observations so far made, but we would like to refer to the following fact (after Hirahara). When a magnetic field is applied in the ac plane at 4.2°K and its component in the c direction is less than 2 kOe, a magnetic moment proportional to the field strength is induced in the field direction (the susceptibility in the a direction being eventually equal to that in the c direction), so that no torque is observed in this case. When the c component of the field exceeds 2 kOe (for total field values not exceeding 20 kOe), a ferromagnetic alignment occurs in the c direction, with a little tilt toward the field direction. Thus, 2 kOe is the value of the critical field for the transition from the helix in the bc plane to the ferromagnetism in the c direction. The anisotropy energy to hold the spin vectors in the bc plane appears to be very large and the c axis seems to be the easiest axis of magnetization. In the torque curves in the ac plane there appears a range of vanishing torque in the neighborhood of the a axis, since there the c component of the field does not exceed 2 kOe. In fact, detailed features of the torque curves, as well as essential features of observed magnetizations curves, in the ac plane can be accounted for very well by the picture described above.

When the field is in the bc plane, the change of the spin structure seems to be more complicated, possibly including the formation of a fan structure for a field along the b axis that exceeds 5.4 kOe.

NOTE ADDED IN PROOF. The magnetostrictive energy is the main driving force for the helix to ferromagnetic transition in Dy and Tb, according to B. R. Cooper (*Phys. Rev. Letters* (1967), to appear). The magnetostrictive energy of cylindrical symmetry about the hexagonal axis in the ferromagnetic state has a magnitude that exceeds by far the magnitude of the anisotropy energy of sixfold symmetry. Our formal analysis for high temperatures should still be valid. In our low temperature analysis, w_{\min} in the right-hand side of (5.3) must include this magnetostrictive energy, whereas the same quantity appearing in Eqs. (5.4) and (5.5) must not. Cooper shows that in Tb the magnetostriction is not frozen in the lattice for the $q = 0$ spin wave but it follows the motion of the magnetization, so that it is not effective for the frequency of this spin wave. It may follow, therefore, that w_{\min} appearing in (5.8) is the same as that appearing in (5.4) and (5.5), and hence is smaller than that in (5.3). The inequality (5.3) then might not ensure the inequality (5.8) for $H = 0$. However, in rare-earth metals, where $p = 6$,

⁵⁰ J. B. Forsyth, S. J. Pickart, and P. J. Brown, *J. Appl. Phys.* **37**, 1053 (1966); *Proc. Phys. Soc. (London)* **88**, 333 (1966).

⁵¹ G. P. Felcher, *J. Appl. Phys.* **37**, 1056 (1966).

it seems that (5.8) still holds, since a direct helix to ferromagnetic transition has been observed.

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Appendix

A1. SUSCEPTIBILITY OF THE FAN STRUCTURE

We shall supplement here the mathematical proof of some facts mentioned in Section 9 and derive the amplitudes of the oscillating x and y spin components in the fan structure, as functions of the applied field (applied in the x -direction). Also the formula for the susceptibility χ_{fan} will be derived. We shall confine ourselves to field values in the vicinity of H_0 , given by (9.13), where a transition occurs between the fan and the ferromagnetic structure.

First we shall consider the quantity

$$\exp \beta [H^* S_x + H'_{nx} S_x + H_{ny} S_y - w(S_z)], \quad (\text{A1.1})$$

where H^* is the effective field in the ferromagnetic state, which is given by

$$H^* = 2J(0)S\sigma_0 + H, \quad (\text{A1.2})$$

σ_0 being defined by

$$S\sigma_0 = \frac{\text{tr } S_x \exp \beta [H^* S_x - w(S_z)]}{\text{tr } \exp \beta [H^* S_x - w(S_z)]}. \quad (\text{A1.3})$$

As in Section 9, H'_{nx} and H_{ny} in (A1.1) are the fluctuating x and y components of the exchange field acting on the n th spin.

We expand (A1.1) in powers of H'_{nx} and H_{ny} . Since S_x and S_y do not commute with S_z , we have to make use of the expansion formula given in the first footnote of Section 4, up to any desired power. Then, there appears, for example, such a term as H'_{nx} times

$$\int_0^\beta d\lambda e^{(\beta-\lambda)K} S_x e^{\lambda K}, \quad \text{where } K = H^* S_x - w(S_z). \quad (\text{A1.4})$$

Also, a term H_{ny}^2 times

$$\int_0^\beta d\lambda \int_0^\lambda d\mu e^{(\beta-\lambda)K} S_y e^{(\lambda-\mu)K} S_y e^{\mu K}, \quad (\text{A1.5})$$

and so on appear. For the sake of brevity, we shall write, for instance, the integrals (A1.4) and (A1.5) as

$$(1, S_x), \quad (1, S_y, S_y),$$

respectively, and S_x times these as

$$(S_x, S_x), \quad (S_x, S_y, S_y),$$

respectively. Then, we can show that the following relations hold:

$$\begin{aligned} \text{tr } (1, S_x) &= \beta \text{tr } S_x e^{\beta K}, \\ \text{tr } (1, S_y, S_y) &= \frac{1}{2}\beta \text{tr } (S_y, S_y), \\ \text{tr } (1, S_x, S_x) &= \frac{1}{2}\beta \text{tr } (S_x, S_x), \\ H^* \text{tr } (S_y, S_y) &= \text{tr } S_x e^{\beta K}, \\ H^{*2} \text{tr } (S_x, S_y, S_y) &= H^{*3} \text{tr } (S_y, S_y, S_y, S_y) \\ &= \frac{1}{2}H^* \text{tr } (S_x, S_x) - \frac{1}{2}\text{tr } S_x e^{\beta K}. \end{aligned} \quad (\text{A1.6})$$

The first three relations can be derived by elementary calculations of changing the order of integrations and with a theorem that, in tr , cyclic permutations of the factors are allowed. The rest follow with the method mentioned in Section 9, (9.8)–(9.11); this method is applied to $\text{tr } S_y \exp \beta K$ and $\text{tr } S_x \exp \beta K$, and the expansion in powers of θ is taken up to θ^3 . By (A1.6), we can express with $\text{tr } S_x \exp \beta K$ and $\text{tr } (S_x, S_x)$ all the other traces. Now,

$$\text{tr } S_x \exp \beta K = S\sigma_0 \text{tr } \exp \beta K, \quad (\text{A1.7})$$

by (A1.3), and $\text{tr } (S_x, S_x)$ can be written in terms of the susceptibility of the ferromagnetic state, χ_f , by the following calculation.

When we vary H^* by δH^* , we have

$$(S\sigma_0)_{H^*+\delta H^*} = \frac{\text{tr } S_x \exp \beta[(H^* + \delta H^*)S_x - w]}{\text{tr } \exp \beta[(H^* + \delta H^*)S_x - w]}.$$

The right-hand side can be written as

$$\frac{\text{tr } S_x \exp \beta[H^*S_x - w] + \delta H^* \text{tr } (S_x, S_x)}{\text{tr } \exp \beta[H^*S_x - w] + \delta H^* \text{tr } (1, S_x)}.$$

Using relations (A1.6), we can write this in terms of $S\sigma_0$ and $\text{tr } (S_x, S_x)$ evaluated at H^* . Namely, we have

$$(S\sigma_0)_{H^*+\delta H^*} = (S\sigma_0)_{H^*} + \delta H^* \left\{ \frac{\text{tr } (S_x, S_x)}{\text{tr } \exp \beta K} - \beta(S\sigma_0)^2 \right\}.$$

Thus,

$$\frac{(S\sigma_0)_{H^*+\delta H^*} - (S\sigma_0)_{H^*}}{\delta H^*} \equiv \frac{d(S\sigma_0)}{dH^*} = \frac{\text{tr } (S_z, S_x)}{\text{tr } \exp \beta K} - \beta(S\sigma_0)^2. \quad (\text{A1.8})$$

On the other hand,

$$\frac{d(S\sigma_0)}{dH} = \frac{d(S\sigma_0)}{dH^*} \frac{dH^*}{dH} = \frac{d(S\sigma_0)}{dH^*} \left\{ 2J(0) \frac{d(S\sigma_0)}{dH} + 1 \right\}, \quad (\text{A1.9})$$

since $H^* = 2J(0)S\sigma_0 + H$. Writing

$$d(S\sigma_0)/dH = \chi_f, \quad (\text{A1.10})$$

which is the *ferromagnetic* susceptibility, we have from (A1.9) the following formula:

$$\frac{d(S\sigma_0)}{dH^*} = \frac{\chi_f}{2\chi_f J(0) + 1}. \quad (\text{A1.11})$$

(A1.11), combined with (A1.8), gives $\text{tr } (S_z, S_x)$ in terms of χ_f and σ_0 .

We are now ready to calculate the quantities

$$S\sigma_{nx} = \frac{\text{tr } S_x \exp \beta[H^*S_x + H_{nx}S_x + H_{ny}S_y - w(S_z)]}{\text{tr } \exp \beta[H^*S_x + H_{nx}S_x + H_{ny}S_y - w(S_z)]} \quad (\text{A1.12a})$$

and

$$S\sigma_{ny} = \frac{\text{tr } S_y \exp \beta[H^*S_x + H_{nx}S_x + H_{ny}S_y - w(S_z)]}{\text{tr } \exp \beta[H^*S_x + H_{nx}S_x + H_{ny}S_y - w(S_z)]}. \quad (\text{A1.12b})$$

The results of the calculation are the following:

$$\begin{aligned} S\sigma'_{nx} (&= S\sigma_{nx} - S\sigma_0) \\ &= \frac{d(S\sigma_0)}{dH^*} H'_{nx} + \left[\frac{d(S\sigma_0)}{dH^*} - \frac{S\sigma_0}{H^*} \right] \frac{H_{ny}^2}{2H^*}, \end{aligned} \quad (\text{A1.13a})$$

$$S\sigma'_{ny} = \frac{S\sigma_0}{H^*} H_{ny} + \left[\frac{d(S\sigma_0)}{dH^*} - \frac{S\sigma_0}{H^*} \right] \left(\frac{H'_{nx}H_{ny}}{H^*} + \frac{H_{ny}^3}{2H^{*2}} \right). \quad (\text{A1.13b})$$

From these follows also a relation

$$\left(\sigma'_{nx} - \sigma_0 \frac{H_{nx}}{H^*} \right) \frac{H_{ny}}{H^*} = \sigma'_{ny} - \sigma_0 \frac{H_{ny}}{H^*}. \quad (\text{A1.14})$$

To solve these equations, we put

$$\begin{aligned} \sigma_{ny} &= \sigma_{y1} \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \sigma_{y2} \sin 3(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots, \\ \sigma'_{nx} &= \sigma'_{x0} + \sigma'_{x2} \cos 2(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots, \end{aligned} \quad (\text{A1.15})$$

so that

$$\begin{aligned} H_{ny} &= 2J(\mathbf{Q})S\sigma_{y1} \sin(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + 2J(3\mathbf{Q})S\sigma_{y3} \sin 3(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots, \\ H'_{nz} &= 2J(0)S\sigma'_{z0} + 2J(2\mathbf{Q})S\sigma'_{z2} \cos 2(\mathbf{Q} \cdot \mathbf{R}_n + \alpha) + \dots. \end{aligned} \quad (\text{A1.16})$$

Substituting (A1.15) and (A1.16) into Eqs. (A1.13a) and (A1.14), we obtain equations to determine the Fourier amplitudes. Confining ourselves to the three amplitudes σ_{y1} , σ'_{z0} , σ'_{z2} , we obtain the following equations:

$$\sigma'_{z0} = \frac{d(S\sigma_0)}{dH^*} 2J(0)\sigma'_{z0} + \frac{S}{H^*} \left[\frac{d(S\sigma_0)}{dH^*} - \frac{S\sigma_0}{H^*} \right] J(\mathbf{Q})^2 \sigma_{y1}^2, \quad (\text{A1.17})$$

$$\sigma'_{z2} = \frac{d(S\sigma_0)}{dH^*} 2J(2\mathbf{Q})\sigma'_{z2} - \frac{S}{H^*} \left[\frac{d(S\sigma_0)}{dH^*} - \frac{S\sigma_0}{H^*} \right] J(\mathbf{Q})^2 \sigma_{y1}^2, \quad (\text{A1.18})$$

$$\begin{aligned} \left[1 - \frac{S\sigma_0}{H^*} 2J(0) \right] 2J(\mathbf{Q})\sigma'_{z0} - \left[1 - \frac{S\sigma_0}{H^*} 2J(2\mathbf{Q}) \right] J(\mathbf{Q})\sigma'_{z2} \\ = \frac{H^*}{S} \left[1 - \frac{S\sigma_0}{H^*} 2J(\mathbf{Q}) \right]. \end{aligned} \quad (\text{A1.19})$$

Near the field $H_0 \{ = 2[J(\mathbf{Q}) - J(0)]S\sigma \}$, we can write

$$\begin{aligned} H^* - 2J(\mathbf{Q})S\sigma_0 &\quad [\text{where } H^* = 2J(0)S\sigma_0 + H] \\ &= -H_0\sigma_0/\sigma + H = -H_0(\sigma_0 - \sigma)/\sigma + (H - H_0) \\ &= [1 - (H_0/\sigma)(d\sigma_0/dH)](H - H_0) \\ &= \{1 - 2[J(\mathbf{Q}) - J(0)]\chi_f\}(H - H_0), \end{aligned}$$

where χ_f , given by (A1.10), is evaluated at H_0 . The quantity calculated above appears in the right-hand side of (A1.19). In other terms of equations (A1.17), (A1.18), and (A1.19) near H_0 , $S\sigma_0/H^*$ can be replaced by $\frac{1}{2}J(\mathbf{Q})$. Furthermore, we can utilize (A1.11) for $d(S\sigma_0)/dH^*$. Then, from (A1.17)–(A1.19) we obtain

$$\sigma'_{z0} = -\{1 - 2[J(\mathbf{Q}) - J(0)]\chi_f\}(\sigma_{y1}^2/4\sigma), \quad (\text{A1.20})$$

$$\sigma'_{z2} = \frac{1 - 2[J(\mathbf{Q}) - J(0)]\chi_f \sigma_{y1}^2}{1 - 2[J(2\mathbf{Q}) - J(0)]\chi_f 4\sigma}, \quad (\text{A1.21})$$

$$\begin{aligned} 2[J(\mathbf{Q}) - J(0)]S\sigma'_{z0} - [J(\mathbf{Q}) - J(2\mathbf{Q})]S\sigma'_{z2} \\ = \{1 - 2[J(\mathbf{Q}) - J(0)]\chi_f\}(H - H_0). \end{aligned} \quad (\text{A1.22})$$

Hence we obtain, for instance, σ'_{x0} as

$$S\sigma'_{x0} = \frac{\{1 - 2[J(Q) - J(0)]\chi_f\}\{1 - 2[J(2Q) - J(0)]\chi_f\}(H - H_0)}{3J(Q) - 2J(0) - J(2Q) - 4[J(Q) - J(0)][J(2Q) - J(0)]\chi_f}. \quad (\text{A1.23})$$

The susceptibility of the fan structure immediately below H_0 is given by

$$\begin{aligned} \chi_{\text{fan}} &= \chi_f + \frac{S\sigma'_{x0}}{H - H_0} \\ &= \frac{1 + [J(Q) + 2J(0) - 3J(2Q)]\chi_f}{3J(Q) - 2J(0) - J(2Q) - 4[J(Q) - J(0)][J(2Q) - J(0)]\chi_f}. \end{aligned} \quad (\text{A1.24})$$

It follows from (A1.23), (A1.20), and (A1.21) that σ'_{x0} is proportional to $H - H_0$, σ_{y1} is proportional to $(H_0 - H)^{1/2}$, and σ'_{x2} is proportional to $H_0 - H$. Other Fourier amplitudes that we have neglected are of higher orders.

The susceptibility of the fan structure just derived and given by (A1.24) can be expressed in another way:

$$\chi_{\text{fan}} = \frac{3\beta_2 + 2\gamma}{3\beta_2 + (2 + \beta_2)\gamma} \frac{1}{2[J(Q) - J(0)]}, \quad (\text{A1.25})$$

where β_2 and γ are defined by (8.9) and (8.10). To derive this, we notice that in the present case σ_0 stands for σ of (8.10). So we have

$$\begin{aligned} 2[J(Q) - J(0)]\gamma &= \frac{\sigma_0}{S} \frac{d}{d\sigma_0} \left(\frac{H^*}{\sigma_0} \right) = -\frac{H^*}{S\sigma_0} + \frac{dH^*}{d(S\sigma_0)} \\ &= -2J(Q) + \frac{2J(0)\chi_f + 1}{\chi_f} \end{aligned} \quad (\text{A1.26})$$

[by (A1.11)], from which follows

$$\chi_f = \{2(\gamma + 1)[J(Q) - J(0)]\}^{-1}. \quad (\text{A1.27})$$

Using this and (8.9), we can write (A1.24) as (A1.25). The formula for χ_{fan} was derived in a simpler way by Kitano and Nagamiya¹⁵ by the method described in Section 8.

At absolute zero, where γ is infinity, χ_f vanishes, and from either (A1.24) or (A1.25) we have

$$\chi_{\text{fan}} = [3J(Q) - 2J(0) - J(2Q)]^{-1}, \quad (\text{A1.28})$$

which was derived earlier by Nagamiya *et al.*¹⁴

The coefficients A and B used in Section 9 (C being disregarded in the following) are the coefficients of H'_{nx} and H'_{ny} , respectively, in (A1.13a) and (A1.13b), namely,

$$A = d(S\sigma_0)/dH^*, \quad B = S\sigma_0/H^*. \quad (\text{A1.29})$$

By the definition of σ_0 , (A1.3), one will be able to imagine easily that σ_0 is an increasing function of H^* , going to saturation, $\sigma_0 \rightarrow 1$, for $H^* \rightarrow \infty$. The curve of σ_0 versus H^* must be a monotonically increasing, upward convex curve. Thus, both A and B are decreasing functions of H^* , and we see from (A1.29) that $A < B$ for nonzero finite H^* .

A2. PARALLEL SUSCEPTIBILITY OF THE HELICAL STATE

With the method developed in Section A1, we can also derive the susceptibility of the helical state for weak field applied parallel to the plane of the spin rotation.

We introduce a local coordinate system ξ, ξ, z such that at \mathbf{R}_n the ξ axis coincides with the direction of the thermally averaged n th spin in no external field, the z axis perpendicular to the plane of the spin rotation, and the ξ axis perpendicular to both. Then we have Eqs. (A1.12a) and (A1.12b), with ξ and ξ replacing x and y . H^* is here related to σ by

$$H^* = 2J(\mathbf{Q})S\sigma, \quad (\text{A2.1})$$

and, in place of (A1.3), we have

$$S\sigma = \frac{\text{tr } S_\xi \exp \beta[H^*S_\xi - w(S_z)]}{\text{tr} \exp \beta[H^*S_\xi - w(S_z)]}. \quad (\text{A2.2})$$

In (A1.13a) and (A1.13b), σ_0 is now replaced by σ , and x and y by ξ and ξ . We shall retain only terms linear in $H'_{n\xi}$ and $H'_{n\xi}$. Then, putting

$$\begin{aligned} H'_{n\xi} &= H \cos(\mathbf{Q} \cdot \mathbf{R}_n) + H''_{n\xi}, \\ H_{n\xi} &= -H \sin(\mathbf{Q} \cdot \mathbf{R}_n) + H'_{n\xi}, \end{aligned} \quad (\text{A2.3})$$

we have simple equations:

$$\begin{aligned} S\sigma'_{n\xi} &= [d(S\sigma)/dH^*][H \cos(\mathbf{Q} \cdot \mathbf{R}_n) + H''_{n\xi}], \\ S\sigma_{n\xi} &= [S\sigma/H^*][-H \sin(\mathbf{Q} \cdot \mathbf{R}_n) + H'_{n\xi}]. \end{aligned} \quad (\text{A2.4})$$

Here $H''_{n\xi}$ and $H'_{n\xi}$ are the components of an exchange field acting on the n th spin such that arises from a small change in the spin configuration due to the applied field H .

In the original coordinate system, this small change in the spin configuration can be specified by $S\sigma'_{nx}$ and $S\sigma'_{ny}$; these are related to $S\sigma'_{n\xi}$

and $S\sigma_{n\xi}$ by

$$\begin{aligned} S\sigma'_{nx} &= S\sigma'_{n\xi} \cos (\mathbf{Q} \cdot \mathbf{R}_n) - S\sigma_{n\xi} \sin (\mathbf{Q} \cdot \mathbf{R}_n), \\ S\sigma'_{ny} &= S\sigma'_{n\xi} \sin (\mathbf{Q} \cdot \mathbf{R}_n) + S\sigma_{n\xi} \cos (\mathbf{Q} \cdot \mathbf{R}_n). \end{aligned}$$

The corresponding exchange-field components are

$$\begin{aligned} H'_{nx} &= 2 \sum_m J(\mathbf{R}_{mn}) S\sigma'_{mx}, \\ H'_{ny} &= 2 \sum_m J(\mathbf{R}_{mn}) S\sigma'_{my}, \end{aligned} \quad (\text{A2.5})$$

which are related to $H''_{n\xi}$ and $H'_{n\xi}$ by similar equations.

Transforming (A2.4) back to the original coordinate system, we have

$$\begin{aligned} S\sigma'_{nx} &= \left[\frac{d(S\sigma)}{dH^*} \cos^2 (\mathbf{Q} \cdot \mathbf{R}_n) + \frac{S\sigma}{H^*} \sin^2 (\mathbf{Q} \cdot \mathbf{R}_n) \right] (H + H'_{nx}) \\ &\quad + \left[\frac{d(S\sigma)}{dH^*} - \frac{S\sigma}{H^*} \cos (\mathbf{Q} \cdot \mathbf{R}_n) \sin (\mathbf{Q} \cdot \mathbf{R}_n) \right] H'_{ny}, \end{aligned} \quad (\text{A2.6a})$$

$$\begin{aligned} S\sigma'_{ny} &= \left[\frac{d(S\sigma)}{dH^*} \sin^2 (\mathbf{Q} \cdot \mathbf{R}_n) + \frac{S\sigma}{H^*} \cos^2 (\mathbf{Q} \cdot \mathbf{R}_n) \right] H'_{ny} \\ &\quad + \left[\frac{d(S\sigma)}{dH^*} - \frac{S\sigma}{H^*} \right] \cos (\mathbf{Q} \cdot \mathbf{R}_n) \sin (\mathbf{Q} \cdot \mathbf{R}_n) \cdot (H + H'_{nx}). \end{aligned} \quad (\text{A2.6b})$$

In order to solve (A2.6a) and (A2.6b), together with (A2.5), we expand σ'_{nx} and σ'_{ny} in Fourier components:

$$\begin{aligned} \sigma'_{nx} &= \sigma'_{x0} + \sigma'_{x2} \cos 2(\mathbf{Q} \cdot \mathbf{R}_n) + \sigma'_{x4} \cos 4(\mathbf{Q} \cdot \mathbf{R}_n) + \dots, \\ \sigma'_{ny} &= \sigma'_{y2} \sin 2(\mathbf{Q} \cdot \mathbf{R}_n) + \sigma'_{y4} \sin 4(\mathbf{Q} \cdot \mathbf{R}_n) + \dots. \end{aligned} \quad (\text{A2.7})$$

(Odd harmonics appear only when we include nonlinear terms.) Substituting these into (A2.5), (A2.6a), and (A2.6b), we obtain the following equations:

$$\begin{aligned} S\sigma'_{x0} &= \frac{1}{2}a[H + 2J(0)S\sigma'_{x0}] + \frac{1}{2}bJ(2\mathbf{Q})S(\sigma'_{x2} + \sigma'_{y2}), \\ S\sigma'_{x2} &= aJ(2\mathbf{Q})S\sigma'_{x2} + \frac{1}{2}b[H + J(4\mathbf{Q})S(\sigma'_{x4} + \sigma'_{y4})], \\ &\vdots \\ S\sigma'_{y2} &= aJ(2\mathbf{Q})S\sigma'_{y2} + \frac{1}{2}b[H - J(4\mathbf{Q})S(\sigma'_{x4} + \sigma'_{y4})], \\ &\vdots \end{aligned} \quad (\text{A2.8})$$

where

$$a = d(S\sigma)/dH^* + (S\sigma/H^*), \quad b = d(S\sigma)/dH^* - (S\sigma/H^*). \quad (\text{A2.9})$$

From these equations, we can easily obtain $S\sigma'_{x0}$. With the use of (A1.11), (A2.1), and (A2.9), we can express the result as

$$\begin{aligned} \chi_h &= \frac{S\sigma'_{x0}}{H} \\ &= \frac{1 + 2[J(\mathbf{Q}) + J(0) - 2J(2\mathbf{Q})]\chi_f}{2\{2J(\mathbf{Q}) - J(2\mathbf{Q}) - J(0) + [J(\mathbf{Q}) - J(0)][J(0) - J(2\mathbf{Q})]\chi_f\}}, \end{aligned} \quad (\text{A2.10})$$

where we understand by χ_f the ferromagnetic susceptibility at $H = H_0$. Using (A1.27), we can also express (A2.10) as

$$\chi_h = \frac{2\beta_2 + \gamma}{2\beta_2 + (1 + \beta_2)\gamma} \frac{1}{2[J(\mathbf{Q}) - J(0)]}. \quad (\text{A2.11})$$

For $T \rightarrow 0$, (A2.10) or (A2.11) reduces to

$$\chi_h = \{2[2J(\mathbf{Q}) - J(2\mathbf{Q}) - J(0)]\}^{-1}. \quad (\text{A2.12})$$

These equations for χ_h were derived by Kitano and Nagamiya¹⁵ and earlier, for a special case, by Yoshimori.¹