Magnetization Process of a Screw Spin System. II

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Magnetization processes and structure changes in helical and modified helical spin arrangements due to application of a magnetic field are studied for finite temperatures and absolute zero in a number of cases of different anisotropy energy. In the case of a strong anisotropy giving rise to a proper helical structure and of no anisotropy within the plane, a field applied within the plane causes structure changes helix to fan and then to parallel alignment as the field is increased, as known already for absolute zero, but it is shown that the first transition is of the second kind when the temperature is close to the Néel temperature, whereas it is of the first kind below a certain critical temperature; the second transition is always of the second kind. The case of p-fold symmetry within the plane is also studied; in particular, the case of twofold symmetry with a field applied either along the easy direction or hard direction is studied in detail for arbitrary temperatures, the results being summarized in Figs. 3 and 4; the case of sixfold symmetry is also studied in some detail for absolute zero and it is shown that, with an applied field along one of the easy directions, the transition from fan to ferromagnetism becomes discontinuous (i.e., of the first kind) when the anisotropy constant increases and that the fan structure disappears with a further increase in the anisotropy constant (Fig. 6). With a field applied along one of the six hard directions, the fan structure does not disappear, and there appears an oblique ferromagnetism at higher values of the anisotropy constant (Fig. 7). Finally, structure changes for longitudinal sinusoidal structure, oblique helix, and conical structure are discussed. Comparison with experiment is made on a number of materials, particularly on Ho, and most, if not all, observed features are accounted for.

§ 1. Introduction

In the last few years, an interesting group of ordered spin arrangements, namely, the screw structure and modified screw structures, was studied both theoretically and experimentally, the first prediction of the screw structure being due to Yoshimori, and then to Villain, and Kaplan. Of particular interest are the various modified structures found in heavier rare-earth metals by neutron diffraction experiments by Koehler, Wilkinson, Cable, and Wollan at Oak Ridge and interpreted by Elliott, Kaplan, and Miwa and Yosida as being due to the combined action of exchange energy and anisotropy energies of hexagonal symmetry. A further advance was made by Yosida and Watabe and also by de Gennes in interpreting the period of the spin arrangement at absolute zero in rare-earth metals, in terms of the exchange interaction between the conduction electrons and localized f electrons.
Structure changes in such spin arrangements due to application of a magnetic field were also investigated. Herpin and Mériel\(^{10}\) studied the changes of the screw structure in MnAu\(_2\) (polycrystalline) by neutron diffraction experiment and also theoretically. Their theory was then supplemented by Enz.\(^{11}\) Nagamiya, Nagata, and Kitano\(^{2}\) studied the same problem and dealt with it most generally and extensively, which will hereafter be referred to as paper I. More recently, Koehler\(^{13}\) made neutron diffraction measurements of the structure changes in Ho (single crystal) by applying a magnetic field at various temperatures, either along the easy axis or along the hard axis in the hexagonal plane, which is the easy plane of magnetization in this crystal in a certain high temperature range. The theories quoted above, except those of Yosida and Watabe, and de Gennes, have been reviewed by Nagamiya.\(^{14}\)

The present paper is an extension of paper I. In paper I the magnetization processes, including structure changes, were studied for the simple screw structure with a field applied in the easy plane of magnetization, no anisotropy in this plane being assumed and the temperature being assumed to be absolute zero. In the present paper, we extend the theory to finite temperatures, include anisotropy within the plane of easy magnetization, and also consider modified structures such as sinusoidal, oblique-screw, and conical structures. The theory is based on the Weiss molecular field approximation and the mathematical technique consists in solving the equation which equates the exchange plus external field to the molecular field that can be obtained from statistical-mechanical consideration. The latter is a quantity which is proportional to the inverse Brillouin function of the thermal average of the magnetic moment of an atom under consideration. To be concerned with the inverse Brillouin function, rather than the Brillouin function itself, is a matter of convenience, but then the mathematics becomes a little more elegant. In actuality, we do not make explicit use of the inverse Brillouin function. When we are concerned with absolute zero, we also make use of the torque equation or the principle of minimum energy.

In §§ 2 and 3 we deal with the case that the magnetic moment vectors of the crystalline layers are confined in the easy plane and a magnetic field is applied in the same plane; § 2 is concerned with the case of no anisotropy within the easy plane and § 3 the case with anisotropy within the plane, particularly that of twofold or sixfold symmetry. Section 4 treats the case of modified screw structures, particularly oblique-screw structure and conical structure. The main results obtained in §§ 2–4 are illustrated by figures and summarized in some detail in § 5. Comparison between theory and existing experimental facts is made only briefly in § 6, because the latter is yet few and the theory is not complete, especially in that the period of the spin arrangement is assumed to be constant throughout the whole range of field strength, which is experimentally not true.
§ 2. Magnetization process of a proper screw system

We shall consider a system consisting of a large number of equivalent layers of spins which are aligned ferromagnetically within each layer. The exchange coupling constant (say per pair of atoms) between the \( m \)-th and \( n \)-th layers will be denoted by \( J_{|m-n|} \) and their Fourier transform, defined by

\[
J(q) = \sum_n J_{|n|} \exp(inq) = J_0 + 2J_1 \cos q + 2J_2 \cos 2q + \cdots,
\]

will be assumed to have its absolute maximum at \( q = q_a \).

In this section, we shall assume that the plane of easy magnetization is parallel to the layers and there is no anisotropy energy within this plane. In the absence of external magnetic field, therefore, a proper screw arrangement of spins with a turn angle \( q_0 \) will be formed. The magnitude of the magnetic moment vector (say per atom) of each layer at a finite temperature will be denoted by \( \mu \sigma \) and its saturation value at absolute zero by \( \mu \). If \( H^{(m)} \) is the magnitude of the molecular field acting on each spin, then \( \sigma \) is given by the Brillouin function of \( \mu H^{(m)}/kT \), provided the anisotropy energy is small compared with the exchange energy, and conversely \( H^{(m)} \) is given by the corresponding inverse Brillouin function of \( \sigma \) multiplied by \( kT/\mu \). As such a function of \( \sigma \) and \( T \), the molecular field will be denoted by \( H^{(m)}(\sigma, T) \). On the other hand, the magnitude of the molecular field on each atom calculated from the exchange interaction with the surrounding spins is given by \( J(q_0) \sigma/\mu \), so that \( \sigma \) must satisfy the equation

\[
\mu H^{(m)}(\sigma, T) = J(q_0) \sigma. \tag{2·1}
\]

Now, when a magnetic field \( H \) is applied along a direction parallel to the layers, the magnetic moment vectors of the layers will be tipped towards the field direction, and they will elongate or contract depending on their direction relative to that of the field. In this section and in the next section, we shall assume that the magnetic moment vectors are always confined within the easy plane. If we denote the magnetic moment of the \( n \)-th layer by \( \mu \sigma_n \) and the molecular field acting on it by \( H_n \), we have in place of (2·1) the following equation:

\[
\mu H_n = \mu H^{(m)}(\sigma_n, T) \sigma_n/\sigma_n = \mu H + \sum_m J_{|m-n|} \sigma_m. \tag{2·2}
\]

In the following, we shall investigate the stable arrangement of the magnetic moment vectors by solving (2·2).

(i) High temperature case

Noting that \( \mu H^{(m)}(\sigma_n, T)/\sigma_n \) is an even function of \( \sigma_n \), we expand it around \( \sigma_n = \sigma (\sigma \) being the value of \( \sigma_n \) for no external field) in powers of \( (\sigma_n^2 - \sigma^2) \) as

\[
\frac{\mu H^{(m)}(\sigma_n, T)}{\sigma_n} = \frac{\mu H^{(m)}(\sigma, T)}{\sigma} + \mu \frac{\partial}{\partial \sigma^2} \left( H^{(m)}(\sigma, T)/\sigma \right) (\sigma_n^2 - \sigma^2) + \cdots. \tag{2·3}
\]
According to (2.1), the first term in this expansion should be equal to \( J(q_0) \).
Near the Neel temperature \( T_N \), \( \sigma \) becomes small, being proportional to \( (T_N - T)^{1/2} \), so that \( \sigma_n \) may also be small as long as the applied field is weak, and the expansion (2.3) may converge well.

Substituting (2.3) in (2.2), while retaining only the first term of (2.3), we obtain a linear equation

\[
J(q_0) \sigma_n = \mu H + \sum_m J_{n-m} \sigma_m,
\]

which has the general solution of the form

\[
\sigma_{nx} = \frac{\mu H}{J(q_0) - J(0)} + \xi \sigma \cos(nq_0 + \alpha),
\]

\[
\sigma_{ny} = \eta \sigma \sin(nq_0 + \alpha),
\]

where the \( x \)-axis is taken along the applied field and the \( y \)-axis perpendicular to it. (More generally, the phase constant \( \alpha \) can be different for \( \sigma_{nx} \) and \( \sigma_{ny} \), but this is taken to be the same for the reason that the vector \( \sigma_n \) should oscillate symmetrically about the field direction). \( \xi \) and \( \eta \) are arbitrary amplitudes but they must be equal to 1 for \( H = 0 \), since the initial assumption is that \( \sigma_n \) is equal to \( \sigma \).

To determine \( \xi \) and \( \eta \), we proceed to the next approximation in which the second term of (2.3) is taken into account. Then there will be an additional term proportional to \( (\sigma_n^2 - \sigma^2) \sigma_n \) on the left-hand side of the above linear equation, which we shall at first regard as a perturbing term. In order that the equation with this term be solved, this term must be orthogonal to the free solutions, \( \cos(nq_0 + \alpha) \) and \( \sin(nq_0 + \alpha) \), of the original linear equation, namely,

\[
\sum_n (\sigma_n^2 - \sigma^2) \sigma_{nx} \cdot \cos(nq_0 + \alpha) = 0,
\]

\[
\sum_n (\sigma_n^2 - \sigma^2) \sigma_{ny} \cdot \sin(nq_0 + \alpha) = 0.
\]

(Similar equations with \( \cos \) and \( \sin \) interchanged are identically satisfied.)

In these equations we substitute (2.4) for \( \sigma_{nx}, \sigma_{ny}, \) and \( \sigma_n^2 = \sigma_{nx}^2 + \sigma_{ny}^2 \). A simple calculation will then lead to

\[
\xi^2 = 1 - \frac{4}{\sigma^2} \left\{ \frac{\mu H}{J(q_0) - J(0)} \right\}^2, \quad \eta = 1,
\]

provided \( \mu H \leq \langle 1/2 \rangle \{ J(q_0) - J(0) \} \sigma \). For higher field, \( \xi \) must be zero, and the first orthogonality condition is satisfied identically, while the second condition leads to

\[
\xi = 0, \quad \eta^2 = \frac{4}{3} \left[ 1 - \frac{1}{\sigma^2} \left\{ \frac{\mu H}{J(q_0) - J(0)} \right\}^2 \right].
\]

(2.6) is valid as long as \( \mu H \) does not exceed \( \{ J(q_0) - J(0) \} \sigma \); when \( \mu H \) exceeds this value, both \( \xi \) and \( \eta \) must vanish.
The results expressed by formulas (2·4), (2·5), and (2·6) can be visualized by a hodograph formed by the magnetic moment vectors. When the applied field is weak, the hodograph is an ellipse with its one axis lying along the field direction and its center being shifted from the origin of the vectors towards the field direction. With increasing field strength, the length of the axis of the ellipse along the field direction reduces, while that of the other axis remains unchanged. At a field $H_t$ given by

$$\mu H_t = \frac{1}{2} \{J(q_0) - J(0)\} \sigma,$$  

(2·7)

the ellipse shrinks into a line, so that the magnetic moment vectors arrange themselves in a sinusoidal way, oscillating about the field direction. Thus, the screw spin structure changes continuously into a sinusoidally oscillating spin structure at $H_t$. When the field is further increased, the amplitude of this oscillation decreases and finally it vanishes at a critical field given by

$$\mu H_o = \{J(q_0) - J(0)\} \sigma.$$  

(2·8)

Above this field, all the magnetic moment vectors are forced to align along the field direction.

It may be remarked in passing that at $H=H_o$, where oscillation ceases, $\sigma_n$ becomes again equal to $\sigma$, since $\xi=\eta=0$, as can be seen from (2·4) and (2·8). This fact can be understood in another way by observing that the molecular field at this external field is the sum of $H_o$ and the exchange field $J(0) \sigma/\mu$ so that it is equal to the molecular field at zero external field, $J(q_0) \sigma/\mu$. Therefore, in the immediate neighborhood of $H_o$, as well as at very small fields, $\sigma_n - \sigma^2$ should be very small and the approximation made above should be good for such cases, even if the temperature is low. In particular, Eq. (2·8) for $H_o$ should be valid at any temperature. This field increases with decreasing temperature and tends to $\{J(q_0) - J(0)\}/\mu$ in the limit of absolute zero, in agreement with the result obtained in paper I, though the second coefficient in (2·3) tends to infinity for absolute zero. In the neighborhood of $H_t$, the approximation is valid only when the temperature is sufficiently near the Néel temperature.

(ii) **High temperature case, 2**

The transition at $H_t$ between the screw state and the sine wave state found in the discussion above is continuous. It was pointed out in paper I, however, that such a transition is discontinuous at absolute zero. We may therefore expect that there would exist a critical temperature below which the continuous behavior of the transition breaks down. In order to find this critical temperature, we shall adopt a Fourier expansion method (together with the expansion (2·3)) which is based on the expectation that higher harmonics would appear with small amplitudes in the components of the moment vectors when the temperature is not far away from the Néel temperature. Namely, we put
and consider that the coefficients in these expansions progressively decrease.
Substituting (2.9) into (2.2), with the expansion (2.3) taken up to the second term, we have

\[ \mu \frac{\partial}{\partial \sigma^2} \left( \frac{H^{(m)}(\sigma, T)}{\sigma} \right) \cdot (\sigma_n^2 - \sigma^2) \frac{\sigma_{nn}}{\sigma} = \frac{\mu H}{\sigma} - \{J(q_0) - J(0)\} \xi_z - \{J(q_0) - J(2q_0)\} \xi_z \cos 2(nq_0 + \alpha) \]

\[ - \{J(q_0) - J(3q_0)\} \xi_z \cos 3(nq_0 + \alpha) - \cdots, \quad (2.10-1) \]

\[ \mu \frac{\partial}{\partial \sigma^2} \left( \frac{H^{(m)}(\sigma, T)}{\sigma} \right) \cdot (\sigma_n^2 - \sigma^2) \frac{\sigma_{nn}}{\sigma} = - \{J(q_0) - J(2q_0)\} \eta_3 \sin 2(nq_0 + \alpha) \]

\[ - \{J(q_0) - J(3q_0)\} \eta_3 \sin 3(nq_0 + \alpha) - \cdots, \quad (2.10-2) \]

where

\[ (\sigma_n^2 - \sigma^2) / \sigma^2 = a_0 + a_1 \cos (nq_0 + \alpha) + a_2 \cos 2(nq_0 + \alpha) + \cdots, \]

\[ a_0 = \xi_0^2 - 1 + \frac{1}{2} \xi_1^2 + \frac{1}{2} \xi_2^2 + \frac{1}{2} \xi_3^2 + \cdots + \frac{1}{2} \eta_1^2 + \frac{1}{2} \eta_2^2 + \frac{1}{2} \eta_3^2 + \cdots, \]

\[ a_1 = 2\xi_0 \xi_1 + \xi_1 \xi_2 + \xi_1 \xi_3 + \cdots + \eta_1 \eta_2 + \eta_1 \eta_3 + \cdots, \quad (2.11) \]

\[ a_2 = 2\xi_0 \xi_2 + \frac{1}{2} \xi_1^2 + \xi_1 \xi_3 + \cdots - \frac{1}{2} \eta_1^2 + \eta_1 \eta_2 + \cdots, \]

\[ a_3 = 2\xi_0 \xi_3 + \xi_1 \xi_2 + \cdots - \eta_1 \eta_2 + \cdots, \]

etc.

Comparing the coefficients of \(1, \cos (nq_0 + \alpha), \cos 2(nq_0 + \alpha), \cdots\) and \(\sin (nq_0 + \alpha), \sin 2(nq_0 + \alpha), \cdots\) on both sides of (2.10-1) and (2.10-2), we obtain equations to determine \(\xi_0, \xi_1, \xi_2, \cdots\) and \(\eta_1, \eta_2, \cdots\) as follows:

\[ - \frac{r}{2} \left( a_0 \xi_0 + \frac{1}{2} a_1 \xi_1 + \frac{1}{2} a_2 \xi_2 + \cdots \right) = \frac{H}{H_0} - \xi_0, \]

\[ - \frac{r}{2} \left( a_0 \xi_1 + a_1 \xi_0 + \frac{1}{2} a_1 \xi_1 + \frac{1}{2} a_2 \xi_2 + \cdots \right) = 0, \]

\[ - \frac{r}{2} \left( a_0 \xi_2 + a_1 \xi_0 + a_2 \xi_1 + \cdots \right) = - \frac{J(q_0) - J(2q_0)}{J(q_0) - J(0)} \cdot \xi_1, \]

\[ \cdots \cdots \cdots, \]

\[ - \frac{r}{2} \left( a_1 \eta_1 + \frac{1}{2} a_2 \eta_2 + \frac{1}{2} a_3 \eta_3 + \cdots \right) = 0, \quad (2.12) \]
where

\[ \tau = \frac{2 \mu \sigma^2}{J(q_0) - J(0)} \frac{\partial}{\partial \sigma^2} \left( \frac{H^{(m)}(\sigma, T)}{\sigma} \right) \quad (2.13) \]

In the approximation in which \( \xi \)'s and \( \eta \)'s other than \( \xi_0, \xi_1, \) and \( \eta_1 \) are neglected, we have from (2.12)

\[ \frac{\tau}{2} \left( \xi_0^2 - 1 + \frac{1}{2} \xi_1^2 + \frac{1}{2} \eta_1^2 \right) \xi_0 + \xi_0 \xi_1^2 = \frac{H}{H_0} - \xi_0, \]

\[ \frac{\tau}{2} \left( \xi_0^2 - 1 + \frac{3}{4} \xi_1^2 + \frac{1}{4} \eta_1^2 + 2 \xi_0^2 \right) \xi_1 = 0, \quad (2.12a) \]

\[ \frac{\tau}{2} \left( \xi_0^2 - 1 + \frac{1}{4} \xi_1^2 + \frac{3}{4} \eta_1^2 \right) \eta_1 = 0. \]

When \( \xi_0 \leq 1/2, \) we have from these equations

\[ \xi_1^2 = 1 - 4 \xi_0^2, \quad \eta_1 = 1, \quad (2.14) \]

\[ \left( 1 + \frac{\tau}{2} \right) \xi_0 - \frac{5}{2} \tau \xi_0^3 = \frac{H}{H_0}, \quad (2.15) \]

and when \( 1/2 \leq \xi_0 \leq 1, \) we have

\[ \xi_1 = 0, \quad \eta_1 = \frac{4}{3} (1 - \xi_0^2), \quad (2.16) \]

\[ \left( 1 - \frac{\tau}{6} \right) \xi_0 + \frac{\tau}{6} \xi_0^3 = \frac{H}{H_0}. \quad (2.17) \]

For \( \xi_0 > 1, \) both \( \xi_1, \) and \( \eta_1 \) vanish, and we have

\[ \left( 1 - \frac{\tau}{2} \right) \xi_0 + \frac{\tau}{2} \xi_0^3 = \frac{H}{H_0}. \quad (2.18) \]

If we neglect \( \tau, \) the results (2.14)–(2.18) reduce to those obtained in (i) (see (2.5), (2.6), and the constant term in (2.4), considering (2.8) as the definition of \( H_0). \) \( \tau \) defined by (2.13) is a decreasing function of temperature and it vanishes at the Néel point. If we put \( \mu H^{(m)}(\sigma, T) = kTB^{-1}(\sigma), \) we have near the Néel point

\[ \tau \approx \frac{2J(q_0)}{J(q_0) - J(0)} \frac{T_N - T}{T_N}. \quad (2.13a) \]

From (2.15), (2.17), and (2.18) we can calculate the magnetization (per atom), \( \mu \sigma \xi_0, \) as a function of \( H/H_0. \) In Fig. 1(a) is shown such a magnetization curve.
for small $\gamma$; it has a cusp at $\xi_0 = 1/2$, or at the field given by
\[ H_t = \left( \frac{1}{2} - \frac{\gamma}{16} \right) H_0. \] (2.19)

When the temperature is lowered and $\gamma$ becomes larger, the slope of the magnetization just below $H_t$ becomes steeper and finally it tends to infinity as can be seen from Eq. (2.15), at a temperature $T_0$ given by
\[ \gamma = \frac{8}{11}, \text{ or } \frac{T_N - T_s}{T_N} \approx 8 \frac{J(q_0) - J(0)}{2J(q_0)}. \] (2.20)

When the temperature is further lowered below this critical temperature, $T_s$, so that $\gamma$ becomes larger than $8/11$, the magnetization curve will appear as shown in Fig. 1(b). In this case, a discontinuous transition should occur at $H_t$ along the dotted vertical line separating equal areas on its both sides. The transition at $H_0$ is always of the second kind.

The approximation made above should be valid when the quantities
\[ \beta_k = \frac{J(q_0) - J(kq_0)}{J(q_0) - J(0)} \quad (k=2, 3, \ldots) \] (2.21)
are large compared with $\gamma$, as one sees from (2.12). For small $q_0$, $\beta_k$'s are large because $J(q_0)$ is the absolute maximum of $J(q)$ and $J(0)$ is a minimum whose value should not be much different from the value of $J(q_0)$. If we take into account those terms which are linear in $\gamma/\beta_k$ in solving Eq. (2.12), the harmonics higher than the third are to be neglected. The inclusion of the second and third harmonics shifts the transition field $H_t$ towards a lower field and the critical temperature $T_s$ towards a higher temperature. In the case where we have only $J_0, J_1,$ and $J_2$ as exchange constants, these shifts can be evaluated and
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It can be shown that the shift in $T_N - T_0$ and that in $H_i$ for $T > T_0$ amount to less than 10% as long as $q_0$ is less than 60°. However, when $q_0$ is near 90°, 120°, or 180°, the second and third harmonics can no longer be regarded as small, because the quantities $\gamma/\beta_k$ with $k = 2, 3$ are not small and eventually tend to infinity in the limit of these angles. For these cases, a special treatment such as that made in I will be required.

(iii) Arbitrary temperature case

When the temperature is further lowered and the parameters $\gamma/\beta_e$ calculated from (2·13) and (2·21) become comparable to or larger than unity, the calculation given in (ii) fails. However, if the applied field is either weak or close to the critical field $H_0$ given by (2·8), we can solve Eq. (2·2) with a sufficient accuracy throughout the whole range of temperature. In the expansion (2·3), the coefficients of $(\sigma^2 - \sigma^2)$, $(\sigma^4 - \sigma^4)$, ... increase indefinitely as temperature approaches to absolute zero, since the saturation moment $\sigma$ becomes independent of the magnitude of the molecular field, but at the same time $\sigma_n$ tends to $\sigma$ for the same reason, so that the series converges well at any temperature in the mentioned two cases. In fact, (2·3) is a power series of $H$ in the case of weak field and a power series of $H_0 - H$ in the case of $H$ close to $H_0$. On the other hand, the Fourier expansion (2·9) should generally be valid, and in the weak field case the coefficients $\xi_1$ and $\eta_1$ should approximately be equal to 1 and other coefficients should be small, while when $H$ is close to $H_0$ one expects $\xi_2$ to be approximately equal to 1 and other coefficients to be small. We shall take the expansion (2·3) up to the second term and calculate the effect of the field linear in $H$ in the weak field case and linear in $H_0 - H$ in the strong field case.

When $H$ is small, we put $\xi_1 = 1 + \xi'_1$, $\eta_1 = 1 + \eta'_1$ and consider the quantities $\xi_0$, $\xi_1$, $\eta_0$, $\eta_1$, $\eta_2$, $\eta_3$, ... to be proportional to $H$. Then linearizing Eq. (2·12) with respect to these small quantities, we obtain

$$\frac{T}{4} (2\xi_0 + \xi_1 + \eta_1) = \frac{H}{H_0} - \xi_0,$$

$$\frac{T}{4} (3\xi_1' + \eta_1' + \xi_2 + \eta_2) = 0,$$

$$\frac{T}{4} (2\xi_0 + \xi_3 + \eta_3) = -\beta_2 \xi_2,$$

$$\cdots \cdots \cdots$$

$$\frac{T}{4} (\xi_1' + 3\eta_1' - \xi_3 - \eta_3) = 0,$$

$$\frac{T}{4} (2\xi_0 + \xi_1 + \eta_1) = -\beta_1 \eta_1,$$

$$\cdots \cdots \cdots$$
The solution of these equations gives the Fourier coefficients of the $x$- and $y$-components of the magnetization vectors. In particular, one obtains $\xi_0$ from the first, third, and last equations and the corresponding initial susceptibility (per atom) as

\[
x_0 = \frac{\xi_0 \sigma \mu}{H} = \frac{2\beta_2 + \gamma}{2\beta_2 + (1 + \beta_2)\gamma} \cdot \frac{\mu^2}{J(q_0) - J(0)}. \tag{2.22}
\]

When $H$ is close to $H_0$, we put $\xi_0 = 1 - \xi_0'$ and consider $\xi_0$, $\xi_1$, $\xi_2$, $\xi_3$, ... to be of the order of $H_0 - H$, $(H_0 - H)^{1/2}$, $H_0 - H$, $(H_0 - H)^{3/2}$, ..., respectively, and $\eta_1$, $\eta_2$, $\eta_3$, ... of the order of $(H_0 - H)^{1/2}$, $(H_0 - H)^{3/2}$, ..., respectively. Then, taking terms of the lowest order in $(2 \cdot 12)$, we obtain the following four equations:

\[
\frac{\gamma}{4} (-4\xi_0' + 3\xi_1 + \eta_1^3) = \frac{H}{H_0} - 1 + \xi_0',
\]

\[
\frac{\gamma}{4} (2\xi_1) = 0,
\]

\[
\frac{\gamma}{4} (4\xi_1 + \xi_2^2 - \eta_1) = -\beta_1 \xi_2,
\]

\[
\frac{\gamma}{4} (2\xi_1 \eta_1) = -\beta_1 \eta_2.
\]

When $H_0 > H$, these equations give

\[
\xi_0' = \frac{3\beta_2 + 2\gamma}{3\beta_2 + (2 + \beta_2)\gamma} \cdot \frac{H_0 - H}{H_0}, \quad \xi_1 = \eta_2 = 0,
\]

\[
\xi_1 = \frac{2\gamma}{3\beta_2 + 2\gamma} \xi_0', \quad \eta_1' = \frac{8(\beta_2 + \gamma)}{3\beta_2 + 2\gamma} \xi_0',
\]

while when $H_0 < H$, they give

\[
-\xi_0' = \frac{1}{1 + \gamma} \cdot \frac{H - H_0}{H_0}, \quad \xi_1 = \eta_2 = \xi_3 = \eta_3 = 0.
\]

Correspondingly, the susceptibility in the vicinity of $H_0$ is obtained as

\[
x_1 = \frac{\xi_0' \sigma \mu}{H_0 - H} = \frac{3\beta_2 + 2\gamma}{3\beta_2 + (2 + \beta_2)\gamma} \cdot \frac{\mu^2}{J(q_0) - J(0)}, \quad (H < H_0) \tag{2.23}
\]

\[
x_2 = \frac{-\xi_0' \sigma \mu}{H - H_0} = \frac{1}{1 + \gamma} \cdot \frac{\mu^2}{J(q_0) - J(0)}, \quad (H > H_0) \tag{2.24}
\]

The susceptibility formula (2.22) is the same as that derived by Yoshi-mori\(^1\) for the case that exchange interactions between those layers which are separated by two or more layers can be neglected. As temperature goes down to zero, $\gamma$ becomes infinite, and (2.22), (2.23) and (2.24) tend to $x_0 = z_1/(1 + \beta_2)$, $x_1 = 2z_1/(2 + \beta_2)$,
and \( z_s = 0 \), respectively, in agreement with the results obtained in I, where

\[
\chi_l = \frac{\mu^2}{J(q_0) - J(0)} \tag{2.25}
\]

is the perpendicular susceptibility which corresponds to the applied field perpendicular to the screw plane in the absence of anisotropy energy. As temperature approaches to the Néel point, \( z_o, z_1, \) and \( z_2 \) tend to \( z_l \) since \( \gamma \) tends to zero. In Fig. 2 is shown the temperature dependence of the susceptibilities \( z_o, z_1, \) and \( z_2 \). They increase steeply near the Néel point when \( q_0 \) is small, since \( 2J(q_0) / (J(q_0) - J(0)) \) is large compared with unity.

We shall finally discuss the transition at low temperatures between the screw and oscillating spin structures. We shall assume that the expression for the free energy (per atom) at low fields,

\[
F(H) = F(0) - \frac{1}{2} z_o H^2, \tag{2.26}
\]

and that at high fields,

\[
F(H) = F(H_0) + \mu \sigma (H_0 - H) - \frac{1}{2} z_1 (H_0 - H)^2, \tag{2.27}
\]

are both valid near the transition field. Noting that \( \sigma_n = \sigma \) at \( H = 0 \) and at \( H = H_0 \) and that the entropy terms in \( F(0) \) and \( F(H_0) \) are therefore identical, we can put \( F(H_0) \) equal to \( F(0) - (1/2) \mu \sigma H_0 \). Then we obtain the transition field at a given temperature from the intersection of the two free energy curves given by (2.26) and (2.27). The result is as follows:

\[
H_t = \frac{1}{\beta_1 + \gamma} \left[ \frac{2\beta_2 + (1 + \beta_2) \gamma}{3\beta_2 + (2 + \beta_2) \gamma} \right] H_0, \tag{2.28}
\]

where we have written \( \mu \sigma / H_0 = z_1 \) (which, in the present case, is identical with (2.25)). Using (2.22), (2.23), and (2.24), we have

\[
H_t = \frac{1}{\beta_1 + \gamma} \left[ \{2\beta_2 + (1 + \beta_2) \gamma\}^{1/3} \{3\beta_2 + (2 + \beta_2) \gamma\}^{1/3} - \{2\beta_2 + (1 + \beta_2) \gamma\} \right] H_0. \tag{2.29}
\]

The temperature dependence of \( H_t \) is implicitly expressed through \( \gamma \) and \( H_0 \). The value of \( H_t / H_0 \) falls in the range between 0.5 and 0.414 for any values of...
§ 3. Effect of anisotropy energy within the plane

In this section, we shall consider the case that there is an anisotropy energy of twofold, fourfold or sixfold symmetry within the plane of the layers. We shall again assume that the magnetic moment vectors of the layers are confined within the plane of the layers.

With increasing temperature the anisotropy energies of fourfold and sixfold symmetry become negligibly small compared with the exchange energy difference \( \{J(q_0) - J(0)\} \sigma^2/2 \), but the anisotropy energy of twofold symmetry does not become negligible and it plays an important role near the Néel temperature in such a way that it makes the moment vectors oscillate sinusoidally in the easy direction.\(^{26}\) This is known in the high temperature magnetic structure of erbium and in the magnetic structure of thulium over the whole temperature range. It is, therefore, appropriate to consider separately the case of twofold anisotropy energy at high temperatures and the case of twofold and otherfold anisotropy energies at low temperatures. It is also convenient to discuss separately the case in which the field is applied parallel to one of the easy axes and the case in which it is applied parallel to one of the hard axes.

If we take the \( x \)-axis along one of the easy axes and the \( y \)-axis perpendicular to it in the plane of the layers, the anisotropy energy of \( p \)-fold symmetry may be written as

\[
W_a = -\frac{1}{2} V_p \sum_n \{ (\sigma_{nx} + i\sigma_{ny})^p + (\sigma_{nx} - i\sigma_{ny})^p \},
\]

where \( V_p \) may depend on temperature. The corresponding effective field on \( \sigma_n \) is \(-\partial W_a/\mu \partial \sigma_n\), so that Eq. (2·2) is now written as

\[
\mu H^{(m)}(\sigma_n, T) \frac{\sigma_n}{\sigma_n} = \mu H + \sum_m J_{|m-n|} \sigma_m - \frac{\partial}{\partial \sigma_n} W_a.
\]

At low temperatures the magnitude of the moment vectors is little affected by the applied field, and in such a case the anisotropy energy \( W_a \) can be considered to be a function of the directions alone of the moment vectors. Equation (3·2) is then useful only when the component perpendicular to \( \sigma_n \) is considered. Thus, multiplying both sides of (3·2) by \( \sigma_n \) vectorially, we have the following equation to determine the direction of \( \sigma_n \):

\[
0 = \mu H \times \sigma_n + \sum_m J_{|m-n|} \sigma_m \times \sigma_n - \frac{\partial}{\partial \sigma_n} W_a \times \sigma_n.
\]

If we write

\[
\sigma_{nx} = \sigma \cos \varphi_n, \quad \sigma_{ny} = \sigma \sin \varphi_n,
\]

We have

\[
0 = \mu H \times \sigma_n + \sum_m J_{|m-n|} \sigma_m \times \sigma_n - \frac{\partial}{\partial \sigma_n} W_a \times \sigma_n.
\]
the above equation can be written

$$0 = \mu H \sigma \sin \varphi_n - \sigma^2 \sum_{m} J_{m-n} \sin (\varphi_m - \varphi_n) + p V_p \sigma^p \sin p \varphi_n. \quad (3.4)$$

This is of course the equation of torque balance, and can be derived from the minimization of energy:

$$E = - \mu H \sigma \sum_{n} \cos \varphi_n - \frac{1}{2} \sigma^2 \sum_{m,n} J_{m-n} \cos (\varphi_m - \varphi_n) - V_p \sigma^p \sum_{n} \cos p \varphi_n$$

$$= \text{min.} \quad (3.5)$$

(i) **Effect of twofold anisotropy energy at high temperatures; field applied parallel to the easy axis**

We shall first study the spin arrangement at high enough temperatures, up to the Néel point, when there is an anisotropy energy of twofold symmetry within the plane, and we shall treat separately the cases of fields parallel to and perpendicular to the easy axis. Expansion (2.3), taken up to the second term, can be used when $\sigma^2$ and $\sigma^4$ are small at high temperatures (and also when $\sigma^2 - \sigma^4$ is small at low temperatures). However, it is not convenient in the present case to put $\mu H^{(m)}(\sigma, T)/\sigma$ equal to $J(q_0)$, as in § 2, since the Néel temperature is not determined by this relation in the limit of $\sigma = 0$. Namely, in the vicinity of the Néel temperature, where the second term of the expansion (2.3) can be neglected, we have from (3.2) for $H = 0$:

$$\frac{\mu H^{(m)}(\sigma, T)}{\sigma_n} = \frac{\sigma_{nx}}{\sigma_n} = \sum_{m} J_{m-n} \sigma_{mx} + 2V_2 \sigma_{ns},$$

$$\frac{\mu H^{(m)}(\sigma, T)}{\sigma_n} = \frac{\sigma_{ny}}{\sigma_n} = \sum_{m} J_{m-n} \sigma_{my} - 2V_2 \sigma_{ny}, \quad (3.6)$$

The solution of these equations is either $\sigma_{nx} \propto \cos (nq_0 + \alpha)$, $\sigma_{ny} = 0$, with

$$\lim_{\sigma \to 0} \frac{\mu H^{(m)}(\sigma, T)}{\sigma} = J(q_0) + 2V_2,$$

or $\sigma_{nx} = 0$, $\sigma_{ny} \propto \sin (nq_0 + \alpha)$, with

$$\lim_{\sigma \to 0} \frac{\mu H^{(m)}(\sigma, T)}{\sigma} = J(q_0) - 2V_2.$$

Evidently, the Néel point is determined by the former equation, since the latter equation will give a lower Néel point. Thus, we have a sinusoidal oscillation of the $x$-component in a certain range of temperature below the Néel point. It is now convenient to define $\sigma$ as a function of $T$ by

$$\mu H^{(m)}(\sigma, T)/\sigma = J(q_0) + 2V_2 \quad (3.7)$$

and make use of (2.3), taken up to the second term, and (3.2). However, in doing so, it is not meant that $\sigma_n$ reduces to $\sigma$ for $H = 0$ in the present case, except for $T \to T_N$, unlike in the previous case of no anisotropy energy within
the plane; \( \sigma \) represents merely the origin of the expansion (2.3).

Now we expand \( \sigma_{nx} \) and \( \sigma_{ny} \) in Fourier series as in (2.9). Then we have equations similar to (2.10-1) and (2.10-2). Equation (2.10-1) remains valid but (2.10-2) is so modified that there appears an additional term \(-4V_{d}\sigma_{ny}\) on the right-hand side. At high temperatures where \( \gamma/\beta \) is small compared with unity, we can follow the procedure developed in (ii) of the preceding section, and we have, in place of (2.12a), the following equations (only the third equation of (2.12a) being changed):

\[
\frac{\gamma}{2} \left( \xi_o^2 - 1 + \frac{3}{2} \xi_i^2 + \frac{1}{2} \eta_i^2 \right) \xi_i = -\frac{\mu H}{j(q_0) - j(0)} \xi_o,
\]

\[
\frac{\gamma}{2} \left( 3\xi_o^2 - 1 + \frac{3}{4} \xi_i^2 + \frac{1}{4} \eta_i^2 \right) \xi_i = 0,
\]

\[
\frac{\gamma}{2} \left( \xi_o^2 - 1 + \frac{1}{4} \xi_i^2 + \frac{3}{4} \eta_i^2 \right) \eta_i = -\frac{\gamma}{3} \eta_i,
\]

where

\[
\gamma_i = 12V_d/|j(q_0) - j(0)|.
\]

In the highest temperature range, where \( \gamma < \gamma_1 \) as will be seen soon later, we have \( \gamma_i = 0 \) at low fields and a solution of (3.8) as

\[
\left( 1 + \frac{\gamma}{2} \right) \xi_i = \frac{5}{2} \eta_i^2 = -\frac{\mu H}{j(q_0) - j(0)} \xi_o,
\]

\[
\xi_i^2 = \frac{4}{3} (1 - 3\xi_o^2).
\]

The spins oscillate sinusoidally along the easy axis (the field direction) with an amplitude \( \sigma \xi_i \) given by (3.11) and with a center of oscillation \( \sigma \xi_o \) calculated from (3.10), the latter increasing and the former decreasing with increasing field, until the field reaches the value \( H_{10} \) given by

\[
\mu H_{10} = \left( 1 - \frac{\gamma}{3} \right) \frac{1}{\sqrt{3}} |j(q_0) - j(0)| \sigma,
\]

where the oscillation ceases (\( \xi_o = 0 \)). Above this field, all the moment vectors align parallel to the field and vary according to

\[
\left( 1 - \frac{\gamma}{2} \right) \xi_o + \frac{\gamma}{2} \xi_i^2 = -\frac{\mu H}{j(q_0) - j(0)} \xi_0 \sigma
\]

(cf. (2.18)). It can be shown that here the solution with \( \gamma_i \neq 0 \) is unstable for any field strength.

As temperature is lowered and \( \gamma \) comes to exceed \( \gamma_1 \), the sinusoidal structure changes into an elliptic one at zero field. The solution of (3.8) in this temperature range at low fields is given by (3.10), together with
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\[ \xi_i^2 = 1 - 4\xi_\phi^2 + \frac{\gamma_2}{3\gamma}, \quad (3.14) \]

\[ \eta_i^2 = 1 - \frac{\gamma_2}{\gamma}, \quad (3.15) \]

The lengths of the two axes of the ellipse at zero field are in the ratio \( \xi : \eta = (1 + \gamma_2/3\gamma)^{1/2} : (1 - \gamma_2/\gamma)^{1/2} \), so that \( \gamma \) must be larger than \( \gamma_2 \). As the field is increased, the center of the ellipse shifts towards the field direction, \( \xi \) decreases, but \( \eta \) remains constant. At the field where \( \xi \) vanishes, namely at \( \mu H_{at} = \left(1 - \frac{5}{24} \gamma_2 - \frac{\gamma}{8}\right) \frac{1}{2} \sqrt{1 + \frac{\gamma_2}{3\gamma} (J(q_0) - J(0)) \sigma}, \quad (3.15) \)

the elliptic oscillation transforms into a transverse sinusoidal oscillation. Then we have the following solution of \( (3.8) : \)

\[ \left(1 - \frac{\gamma}{6} - \frac{2\gamma_2}{9}\right) \xi_\phi + \frac{\gamma}{6} \xi_\phi^3 = \frac{\mu H}{\{J(q_0) - J(0)\} \sigma}, \quad (3.16) \]

This transverse oscillation ceases at \( \mu H_{ts} = \left(1 - \frac{\gamma_2}{3}\right) \sqrt{1 - \frac{2}{3} \gamma_2 (J(q_0) - J(0)) \sigma} \quad (3.17) \)

Above this field the magnetization follows Eq. \( (3.13) \) (see Fig. 3(a)).

The transition at \( H_{at} \) is of the second kind (continuous) only when \( \gamma < 8/11 - (5/11) \gamma_2 \), and in order that this inequality be satisfied for \( \gamma > \gamma_2, \gamma_2 \) must be smaller than 1/2. For larger values of \( \gamma \) (namely, at lower temperatures), the transition is of the first kind (discontinuous) and should occur at a field, \( H_{at1} \), which is a little higher than \( H_{at} \) given by \( (3.15) \).

The arguments given above are valid for small \( \gamma_2 \). When \( \gamma_2 > 1/2 \), we have to be more careful. In this case, the transition at \( H_{at} \) is of the second kind only when \( 1/2 > \gamma \). For \( \gamma_2 > \gamma > 1/2 \), it is of the first kind and occurs at a field, \( H_{at1} \) (see below, Eq. \( (3.19) \)), which is a little higher than \( H_{at} \) given by \( (3.12) \). For \( \gamma > \gamma_2 \), the transition between elliptic oscillation and transverse sinusoidal oscillation is also of the first kind and occurs at a field \( H_{at1} \) (see below, Eq. \( (3.18) \)) which is a little higher than \( H_{at} \) given by \( (3.15) \). There is, however, a certain range of \( \gamma \) larger than \( \gamma_2 \) where elliptic oscillation transforms directly

* The suffix 1 is added to a transition field (such as \( H_{at} \) between the elliptic and transverse sinusoidal structures) to indicate that the transition is of the first kind; this convention was also used in § 2.
into parallel alignment at a field $H_{ao}$ without passing through a transverse sinusoidal oscillation (see Fig. 3(b) to (d)). It can be shown that this range of $\gamma$ extends to infinity when $2 \leq \gamma_1 \leq 6$, so that in this case we have no transverse sinusoidal oscillation as a stable state (Fig. 3(c)).

To obtain $H_{ao}$ analytically, we retain terms linear in $H$ in the free energy difference between the elliptic oscillation and parallel alignment. This can be calculated from the corresponding magnetization curves. Then we have

$$\mu H_{ao} = \frac{r_1^2 - 12 - 2r_1 y + 12}{24(\gamma - 2)} \{J(q_0) - J(0)\} \sigma.$$  \hfill (3.18)

This expression is useful only when $\mu H_{ao}$ is not large compared with $\{J(q_0) - J(0)\} \sigma$. $H_{ao}$ given by (3.18) decreases for increasing $\gamma$ and vanishes at $\gamma = (r_1^2 - 12)/(2r_1 - 12)$, provided this is positive, namely, $r_1 > 6$. When

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**Fig. 3. Structure changes in a screw system with twofold anisotropy energy within the easy plane at high temperatures; field applied parallel to the easy axis.**

- ↑ Ferromagnetisms
- ↓ longitudinal sinusoidal oscillation
- O elliptic oscillation
- Ψ transverse sinusoidal oscillation with a ferromagnetic component are all indicated by arrows and other signs; $r_2 = 12V \gamma \{J(q_0) - J(0)\}$. The zero of $\gamma$ corresponds to the Néel point and a larger value of $\gamma$ corresponds to a lower temperature. Transitions indicated by $H$ with a subscript 1 are of the first kind and other transitions are of the second kind.
6 + 2\sqrt{6} > \gamma_2 > 6 \) (Fig. 3 (d)), this critical value of \( \gamma \) is larger than \( \gamma_2 \), so that the elliptic structure transforms into parallel alignment (ferromagnetic arrangement)* as we pass through this value of \( \gamma \) (or the corresponding temperature). For \( \gamma_2 > 6 + 2\sqrt{6} \), we have only the transition between longitudinal sinusoidal oscillation and parallel alignment at \( H_{\text{tot}} \) for \( \gamma > 1/2 \), and this transition field can be calculated in a similar way as

\[
\mu H_{\text{tot}} = \frac{12\gamma - \gamma^2 - 12}{24(\gamma - 2)} \{ J(q_0) - J(0) \} \sigma .
\]

This field vanishes at \( \gamma = 6 + 2\sqrt{6} \), so that for \( \gamma > 6 + 2\sqrt{6} \) we have only parallel alignment (Fig. 3 (e)).

(ii) Effect of twofold anisotropy energy at high temperatures; field applied parallel to the hard axis

In the case where the external field is along the hard axis, the problem is a little more complicated. There can be such a case that the total magnetization is oblique to the applied field and varies with the field strength. First we shall assume that the moment vectors arrange themselves symmetrically with respect to the direction of the applied field. This case is quite simple to treat.

The field is now applied along the \( y \)-axis and \((3.7)\) will still be used. Putting

\[
\sigma_{y0}/\sigma = \xi_1 \cos(nq_0 + \alpha), \quad \sigma_{y0}/\sigma = \eta_0 + \eta_1 \sin(nq_0 + \alpha),
\]

we have, in place of \((3.8)\), the following equations:

\[
\frac{\gamma}{2} \left( \eta_0^2 - 1 + \frac{3}{4} \xi_1^2 + \frac{1}{4} \eta_1^2 \right) \xi_1 = 0,
\]

\[
\frac{\gamma}{2} \left( \eta_0^2 - 1 + \frac{1}{2} \xi_1^2 + \frac{3}{2} \eta_1^2 \right) \eta_0 = \frac{\mu H}{\{ J(q_0) - J(0) \} \sigma} - \frac{\gamma}{3} \eta_0 - \frac{\gamma_2}{3} \eta_0 ,
\]

\[
\frac{\gamma}{2} \left( 3\eta_0^2 - 1 + \frac{1}{4} \xi_1^2 + \frac{3}{4} \eta_1^2 \right) \eta_1 = - \frac{\gamma_2}{3} \eta_1 .
\]

These equations can be solved easily as before and we have the following results.

At high temperatures where \( \gamma < \gamma_1 \), the center of the sinusoidal oscillation in the direction of the easy axis is shifted by the field towards the field direction and its amplitude is decreased, in the manner expressed by

\[
\left( 1 + \frac{\gamma_2}{3} - \frac{\gamma}{6} \right) \eta_0 + \frac{\gamma}{6} \eta_0^3 = \frac{\mu H}{\{ J(q_0) - J(0) \} \sigma} ,
\]

\[
\xi_1^2 = \frac{4}{3} (1-\eta_0^2), \quad \eta_1 = 0.
\]

This oscillation ceases when \( \eta_0 \) reaches unity, namely, above \( H'_0 \) given by

\*) Ferromagnetism occurs only when \( q_0 \) is small. If \( q_0 \) is closer to \( \pi \) than to 0, we expect antiferromagnetism.
\[ \mu H_0' = \left( 1 + \frac{\bar{\gamma}_z}{3} \right) \{ J(q_0) - J(0) \} \sigma. \]  

(3.23)

At lower temperatures where \( \bar{\gamma} > \bar{\gamma}_z \), we have an elliptic oscillation at low fields, provided \( \bar{\gamma}_z \) is small enough, as given by

\[ \left( 1 - \frac{\bar{\gamma}_z}{3} + \frac{\bar{\gamma}}{2} \right) \gamma_0 - \frac{5}{2} \bar{\gamma} \gamma_0^2 = \frac{\mu H}{\{ J(q_0) - J(0) \} \sigma}, \]

(3.24)

\[ \xi^2 = 1 + \frac{\bar{\gamma}_z}{3\bar{\gamma}}, \quad \gamma_0^2 = 1 - 4\gamma_0^2 - \frac{\bar{\gamma}_z}{\bar{\gamma}}, \]

and this oscillation transforms above

\[ \mu H_0' = \left( 1 + \frac{7}{24} \frac{\bar{\gamma}_z - \bar{\gamma}}{8} \right) \frac{1}{2} \sqrt{1 - \frac{\bar{\gamma}_z}{\bar{\gamma}}} \{ J(q_0) - J(0) \} \sigma \]

(3.25)

into sinusoidal oscillation given by (3.22), and finally transforms into parallel alignment above \( H_0' \) given by (3.23). The transition at \( H_0' \) is continuous or discontinuous (at \( H_0' \), a little higher than \( H_0' \)) according as whether \( \bar{\gamma} \) is smaller or larger than \( 8/11 + (37/33)\gamma_1 \), while the transition \( H_0' \) is always continuous. These results are valid for \( \bar{\gamma}_z < 2 \) and are summarized in Fig. 4(a).

We have seen in (i) that, when \( \bar{\gamma}_z > 6 \), ferromagnetism appears in the range of temperature in which \( \bar{\gamma} > 6 + 2\sqrt{6} \). In this case, we have magnetic moment vectors aligned parallel to the easy axis in no external field. When there is a field along the hard axis in this case, the magnetization will be tipped towards this direction. A question then arises whether or not an oscillation of the moment vectors will set in when the field exceeds a certain critical value. In order to solve this question, one may put

\[ \frac{\sigma_{ax}}{\sigma} = \xi_0 + \xi_1 \cos(nq_0 + \alpha), \]

\[ \frac{\sigma_{ay}}{\sigma} = \gamma_0 + \gamma_1 \cos(nq_0 + \alpha), \]

and set up equations for \( \xi_0, \xi_1, \gamma_0, \gamma_1 \) similar to (3.21). Although it is not easy to solve these equations for the general case, the threshold field, \( H_0'' \), at which infinitesimal oscillation sets in can be found to be

\[ \mu H_0'' = \frac{1}{3} \{ \gamma_1 (\bar{\gamma}_z - 3) (1 - \frac{3}{\bar{\gamma}}) \}^{1/2} \{ J(q_0) - J(0) \} \sigma. \]

(3.26)

Below this field, \( \xi_0 \) and \( \gamma_0 \) are given by

\[ \xi^3 = 1 - \frac{2}{\bar{\gamma}} - \gamma_0^2, \quad \gamma_0 = \frac{3\mu H}{\gamma_1 J(q_0) - J(0) \sigma}. \]

(3.27)

Since \( H_0'' \) is lower than \( H_0' \) given by (3.23), one might expect that above \( H_0'' \) the amplitude of oscillation at an oblique position would increase with increasing field and then this oscillation would transform into sinusoidal oscillation, given by (3.22), which is symmetrical with respect to the field direction. However, the field,
below which the symmetrical oscillation begins to be asymmetrical, is calculated to be

\[ H_{t^*} = \frac{\gamma_2}{3} \left[ \frac{\gamma + 7\gamma_2 - 30 + 5\sqrt{\gamma + \gamma_2 - 6}}{3\gamma + 5\gamma_2 - 18 + 3\sqrt{\gamma + \gamma_2 - 6}} \right] \times \frac{\{J(q_0) - J(0)\} \sigma}{\mu} \]

which is lower than \( H_0^* \). Therefore, both \( H_0^* \) and \( H_{t^*} \) have no real meaning, i.e., the oblique parallel alignment must transform directly and discontinuously into sinusoidal oscillation before it starts oscillating (see Fig. 4 (d)). It can be shown that we arrive at the same conclusion with different phase constants \( \alpha \) and \( \beta \) for \( \sigma_{nz} \) and \( \sigma_{ny} \).

The transition field from oblique parallel alignment to sinusoidal oscillation can be calculated as before from the free energy difference between these two states. It is found to be

\[ \mu H_{\alpha^*} = \frac{1}{6} \sqrt{\frac{\gamma_2}{\gamma}} \left( \frac{\gamma^2 - 12\gamma + 12}{2\gamma_2 - 12} \right) \frac{\gamma_2}{\gamma - 6} \frac{\{J(q_0) - J(0)\} \sigma}{\mu} \]

Similarly, the transition field from oblique parallel alignment to elliptic oscillation...
tion is found to be

$$\mu H_n^{**} = \frac{1}{6} \sqrt{\frac{\gamma_2}{\gamma}} \bigg( \frac{2r_1^2 - 12\gamma - r_2^2 + 12}{(\gamma - 6)} \bigg) \{ J(q_o) - J(0) \} \sigma. \quad (3.30)$$

This transition takes place when $6 < \gamma_2 < 6 + 2\sqrt{6}$ (Fig. 4(c)) and $2 < \gamma_1 < 6$ (Fig. 4(b)).

(iii) Low temperature case; field applied parallel to one of the easy axes

We shall now study the magnetization process at low temperatures when there is an anisotropy energy of $p$-fold symmetry, particularly the case of $p = 6$. It will be assumed that the anisotropy energy is not so large as to modify drastically the spin arrangement which we would have for no anisotropy energy at zero external field; namely, we assume that we have a simple screw structure at zero field. It is also assumed that the turn angle of the screw, $q_0$, is not a simple integral multiple of $2\pi/p$, so that the moment vectors of the layers cover uniformly the circle of radius $\sigma$. (If $q_0$ is close to an integral multiple of $2\pi/p$, the anisotropy energy will make the moment vectors fit in its valleys.) The anisotropy energy would modify the uniform rotation of the moment vectors in such a way that their $x$-and $y$-components contain, beside the main Fourier component with wave number $q_0$, higher Fourier components with $(p - 1)q_0$, $(p + 1)q_0$, etc., but we shall disregard them. We shall also be interested mostly in the high field behavior of the system.

When the field is applied along one of the easy axes and is high enough, the moment vectors will align parallel to the field direction. However, below a certain critical field $H_0$, they will oscillate about the field direction, with increasing amplitude for decreasing field. Since the last term of (3.4), which comes from the anisotropy energy, is equivalent to increasing $\mu H\sigma$ by $pV_\sigma \sigma^{p-1}$ when the amplitude is small, it will immediately be seen that the critical field in the presence of the anisotropy energy is given by

$$\mu H_0 = \{ J(q_0) - J(0) \} \sigma - pV_\sigma \sigma^{p-1}, \quad (3.31)$$

instead of (2.8). The susceptibility just below $H_0$ can be calculated by putting

$$\varphi_n = \eta \sin(nq_0 + \alpha) \quad (3.32)$$

and using (3.4) or (3.5) to determine $\eta$. Then we have

$$\chi_1 = \frac{2\mu^2}{(2 + \beta_2) \{ J(q_0) - J(0) \} + (p^4 - p^4) V_\sigma \sigma^{p-1}}, \quad (3.33)$$

$$\eta = 4 \{ x_1 (H_0 - H) / \mu \sigma \}^{1/2}. \quad (3.34)$$

Now it is interesting to observe that we have a negative term $-p^4 V_\sigma \sigma^{p-2}$ in the denominator of (3.33), which has come from the second term of the power series expansion of $pV_\sigma \sigma^{p-1} \sin p\varphi_n$ in (3.4) and hence has minus sign. This term can be very large even if $V_\sigma \sigma^{p-1}$ is not large ($p^4 = 1296$ for $p = 6$). There-
fore, it is possible that $z_1$ is negative. In such a case, the magnetization curve in the neighborhood of $H_0$ would appear like the curve in Fig. 1(b) in the neighborhood of $H_t$. Evidently, in such a case the power series expansion of the anisotropy torque term is no longer permissible, except in the immediate neighborhood of $H_0$. Even in the case of positive $z_1$, the power series expansion of that term soon fails when the field decreases below $H_0$. We shall therefore treat the problem more exactly.

We shall start with the minimum principle (3.5), taking $\sigma=1$ for the sake of simplicity. As a trial function to solve this minimum problem, we shall take

$$\sin \frac{\varphi_n}{2} = \xi \sin (nq_0 + \alpha),$$

(3.35)

instead of (3.32), although these two are equivalent when $\xi$ is small. The preference of (3.35) to (3.32) is a matter of mathematical convenience. For a more refined treatment, one may include higher harmonics, and in fact an infinite Fourier series is able to express any moment vector arrangement within the range of $2\pi$, including a pure screw, provided the period is given. However, a single Fourier term already makes our calculation sufficiently complicated.

Substituting (3.35) in (3.5) and expanding the first two terms of the latter in powers of $\xi$, while exactly retaining the last anisotropy energy term, we have, up to $\xi^4$,

$$E_N = -\frac{1}{2} J(0) - \mu H - V_p$$

$$- \{J(q_0) - J(0) - \mu H - V_p f_p(\xi^2)\} \xi^4$$

$$+ \frac{1}{4} \{3J(q_0) - 2J(0) - J(2q_0)\} \xi^4,$$

(3.36)

where

$$1 - \xi^2 f_p(\xi^2) = \frac{1}{2\pi} \int_0^{2\pi} \cos [2p \sin^{-1}(\xi \sin \varphi)] d\varphi,$$

(3.37)

which is the average of $\cos p\varphi_n$ over a very large number, $N$, of layers, since $nq_0 + \alpha$ covers uniformly the angular range of $2\pi$ and thus can be replaced by a continuous variable $\varphi$. The function $f_p(x)$ is a polynomial of the $(p-1)$th degree,

$$f_p(x) = p^2 - \frac{p^2(p^2-1)}{(2!)^2} x + \frac{p^4(p^2-1)(p^2-2^2)}{(3!)^2} x^3 +$$

$$\ldots \ldots + (-1)^{p-1} \frac{p^2(p^2-1)\cdots(p^2-(p-1)^2)}{(p!)^2} x^{p-1}.$$  

(3.38)

In Fig. 5 we show $f_p(x)$ for $p=6$.

Minimizing (3.36) with respect to $\xi$, we have the condition,
Fig. 5. \( f_x(x) = 36 - 315x + 1120x^2 - 1890x^3 + 1512x^4 - 462x^5 \).

\[
\begin{align*}
- \{J(q_0) - J(0) - \mu H - V_p f_p(x)\} \\
+ \frac{1}{2} \{3J(q_0) - 2J(0) - J(2q_0) + 2V_p f_p'(x)\} x = 0,
\end{align*}
\]  

(3.39)

where \( x = \xi^2 \). This equation can be rewritten as

\[
\mu H = \{J(q_0) - J(0)\} - V_p f_p(x)
\]

\[ - \left[ \frac{1}{2} \left( 2 + \beta \right) \{J(q_0) - J(0)\} + V_p f_p'(x) \right] x, \]

(3.39a)

where \( \beta = \{J(q_0) - J(2q_0)\}/\{J(q_0) - J(0)\} \), as defined by (2.21).

Now, the transition field between the state of sinusoidal oscillation and the state of parallel alignment can be calculated by equating their energies. This transition field, \( H_0 \), is given by (3.31) when \( \xi \) is positive, in which case \( \xi \) vanishes at \( H_0 \), but it will be shifted to a higher field when \( \xi \) is negative, the transition then being between a state of a finite amplitude \( \xi \) and parallel alignment \((\xi = 0)\). Putting the energies of the two states equal to each other, we have from (3.36)

\[
- \{J(q_0) - J(0) - \mu H_0 - V_p f_p(x)\} + \frac{1}{4} \{3J(q_0) - 2J(0) - J(2q_0)\} x = 0.
\]  

(3.40)

This condition and Eq. (3.39) for \( H = H_0 \) give equations

\[
X = \frac{V_p}{J(q_0) - J(0)} = - \frac{2 + \beta}{4 f_p'(x)}
\]  

(3.41)

and

\[
Y = \frac{\mu H_0}{J(q_0) - J(0)} = \frac{\mu H_0}{V_p} \left[ \frac{x - f_p(x)}{f_p'(x)} \right]
\]  

(3.42)
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$x$ can be determined from (3·41),* and substituting it in (3·42), we can calculate $H_0$ as a function of $V_p$.

When the field is further decreased, the sinusoidal oscillation may transform into a screw. For the latter the energy expression (3·36) is no longer valid because (3·35) fails. The energy for the screw can, however, be approximated by $-1/2 \cdot J(q_0) - 1/2 \cdot \chi_0 H$, where $\chi_0$ is the initial susceptibility which is obtained from (2·22) by making $\gamma$ infinite:**

$$\chi_0 = \frac{\mu^2}{(1+\beta_2) \{J(q_0) - J(0)\}}.$$

Equating the energy for the screw to (3·36) and using (3·43), we have an equation which, together with (3·39), determines the transition field $H_t$. Writing $X = V_p / \{J(q_0) - J(0)\}$, $Y = \mu H_t / \{J(q_0) - J(0)\}$, as in (3·41) and (3·42), we have two equations:

$$\frac{1}{1+\beta_2} Y^2 - 2(1-x)Y - 2\{1-xf_p(x)\}X + \left\{1-2x + \frac{1}{2} (2+\beta_2) x^2 \right\} = 0,$$

$$Y + \{f_p(x) + xf_p'(x)\} X - \left\{1 - \frac{1}{2} (2+\beta_2) x \right\} = 0.$$

For the numerical calculations of $H_0$ and $H_t$, it is convenient to draw straight lines (3·39b) in the $XY$-plane by giving various values to $x$ and to look for their cross-points with vertical straight lines (3·41) for $H_0$ or with parabolas (3·44) for $H_t$. Figures 6(a) and 6(b) show examples of such a diagram for $p=6$, taking $\beta_1 = 8$ and $\beta_2 = 3$, respectively. The dashed line in the same figures corresponds to an approximate equation for $H_t$:

$$\frac{\mu H_t}{J(q_0) - J(0)} = \frac{1}{2} - \frac{V_3}{J(q_0) - J(0)}.$$

* It is possible that several different values of $x$ correspond to the same value of $X$. In such a case we have to take the largest $x$.

** This formula is not valid for the case of $p=2$, in which case the initial susceptibility depends on the direction of the applied field. When the field is applied along the easy axis or along the hard axis, we have respectively

$$\chi_0 = \frac{\mu^2}{(1+\beta_2) \{J(q_0) - J(0)\} + 4V_3}.$$

The susceptibility is larger when the field is along the easy axis, since those moment vectors which are perpendicular to the field direction and thus are pointing in the hard direction can turn easily towards the field direction. For a similar reason $\chi_0$ is smaller when the field is along the hard axis.
Fig. 6. Effect of anisotropy energy of sixfold symmetry within the easy plane and effect of external field on the spin structure; the field is applied along one of the easy axes. Transition at $H_0$ for $x < 1/252$ in Fig. (a) and for $X < 1/252$ in Fig. (b) is of the second kind; other transitions are of the first kind. Thin straight lines with indices 0.02, 0.04, etc., indicate the contours of constant amplitude of the sinusoidal oscillation and the numbers are values of $\xi^2$ defined by (3.35).

or $Y = 1/2 - X$. It can be seen that for small $V_x$ the screw structure at low fields transforms into a sinusoidally oscillating structure when one crosses over
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$H_t$ and then into parallel alignment above $H_0$, while for large $V_0$ a direct transition occurs from the screw to the parallel alignment at $H_t$. The value of $H_t$ in the latter case is obtained from (3.44) by putting $x=0$, namely, from

$$\frac{1}{1+\beta}, Y^2-2Y-2X+1=0,$$

$$Y = \mu H_t / \{J(q_0)-J(0)\}. \quad (3.44')$$

The transition at $H_0$ is of the second kind along the straight line marked with $x=0.00$ in Figs. 6(a) and 6(b) down to the point represented by a little black circle (at $\gamma=1/126$ or $\gamma=1/252$), where the line meets the curved lower branch for $H_0$. Along the latter the transition is of the first kind.

In the case of a twofold anisotropy energy, $H_t$ is higher than that given by (3.44) when the field is along the easy axis, because $\gamma_0$ is then larger. It can be shown in this case that the transition at $H_0$ is always of the second kind.

(iv) Low temperature case; field parallel to one of the hard axes

When the field is applied along one of the hard axes, we have the same formula (3.31) for $H_0$ (denoted by $H_0'$ in this case) and (3.33) for $\gamma_1$ (denoted by $\gamma_1'$) except that the sign of $V_0$ is reversed. Evidently we have no first kind transition at $H_0'$ in this case. To obtain $H_0'$, we have also only to reverse the sign of $X$ in Eqs. (3.44) and (3.39b). A complexity arises in this case, as in (ii), that an oscillation at an oblique position might come in as an intermediate stable configuration.

We now assume that the moment vectors oscillate sinusoidally about the intermediate direction between the field direction (hard axis) and one of the neighboring easy axes, and put

$$\sin \left( \frac{\varphi_0 - \varphi}{2} \right) = \xi \sin (n\varphi + \alpha), \quad (0 < \varphi < \pi/\beta) \quad (3.46)$$

where $\varphi$ denotes the angle between the direction of the magnetization and that of the external field. We substitute (3.46) in (3.5) and have

$$\frac{E}{N} = - \frac{1}{2} J(0) - \mu H \cos \varphi + V_0 \cos p\varphi$$

$$- \{J(q_0) - J(0) - \mu \cos P q_0 \cos p\varphi \cdot f_p'(x) \} \xi^2$$

$$+ \frac{1}{4} \{3J(q_0) - 2J(0) - J(2q_0) \} \xi^4. \quad (3.47)$$

Minimizing (3.47) with respect to $\xi$ and $\varphi$, we obtain the following conditions:

$$- \{1 - Y \cos \varphi + X \cos p\varphi \cdot f_p'(x) \} + \left\{ \frac{1}{2} (2+\beta_2) - X \cos p\varphi \cdot f_p'(x) \right\} x = 0,$$

$$(3.48)$$
\[ Y \sin \varphi (1 - x) - X P \sin \rho \varphi \{1 - xf_p(x)\} = 0, \quad (3.49) \]

where \( x = \frac{\xi^2}{x} \), \( X = V_p/\{J(q_0) - J(0)\} \), and \( Y = \mu H/\{J(q_0) - J(0)\} \), as before. Equations (3.48) and (3.49) are those to determine \( x \) and \( \varphi \).

We shall first look for the threshold field, \( H_{t''} \), at which the center of oscillation, or the direction of magnetization, begins to deviate from the direction of the external field towards the easy direction. Making \( c;o'\sim 0 \) in (3.48) and (3.49) we have

\[ Y = \frac{\mu H t'}{J(q_0) - J(0)} \left\{1 - (1/2)(2 + \beta_1) x\right\} - \frac{\rho^2 \{1 - xf_p(x)\} - (1 - x) \{f_p(x) + xf_p'(x)\}}{\rho^2 \{1 - xf_p(x)\} - (1 - x) \{f_p(x) + xf_p'(x)\}}, \quad (3.50) \]

If we eliminate \( x \) from these equations, we obtain \( H_{t''} \) as a function of \( V_p \).

Numerical calculations were made for \( \beta_1 = 8 \) and \( \beta_2 = 3 \) in the case of \( p = 6 \) and in the case of \( p = 2 \), and it was found that \( H_{t''} \) is lower than \( H_{t'} \), the latter being the transition field from screw to sinusoidal oscillation about the field direction. That \( H_{t''} < H_{t'} \) means that in these cases oblique sinusoidal oscillation cannot take place. This would possibly be true in the general case.

Although oscillation at an oblique position does not seem possible, the parallel alignment of the magnetic moment vectors oblique to the field direction should be stable at low fields if \( V_p \) is larger than \( 1/2 \cdot \{J(q_0) - J(0)\} \), namely, if the right-hand side of (3.45) is negative. For such an alignment the angle \( \varphi \) varies with field strength according to Eq. (3.49) with \( x = 0 \), namely,

\[ Y \sin \varphi - X P \sin \rho \varphi = 0. \quad (3.51) \]

The energy of this state is evidently

\[ \frac{E}{N} = -\frac{1}{2} J(0) - \mu H \cos \varphi + V_p \cos \rho \varphi. \quad (3.52) \]

Equating (3.52) to \( -1/2 \cdot J(q_0) - 1/2 \cdot \zeta_0 H \), where \( \zeta_0 \) is given by (3.43), we can determine the transition field, \( H_{t''} \), from screw to oblique ferromagnetic alignment. To simplify the calculation, we assume \( (\pi/\rho - \varphi) \) to be small and proportional to \( H \) and expand (3.51) and (3.52) in powers of \( (\pi/\rho - \varphi) \).

Then we have, up to terms quadratic in \( H \), an equation to determine \( H_{t''} \):

\[ \frac{1}{2} - \frac{Y}{\rho} \cos \frac{\pi}{\rho} + Y^2 \left\{\frac{1}{2(1 + \beta_1)} + \frac{3}{X} \sin^2 \frac{\pi}{\rho}\right\} = 0. \quad (H = H_{t''}) \quad (3.53) \]

Above \( H_{t''} \) we have oblique ferromagnetism, but the angle \( \varphi \) should decrease with increasing field and should vanish at \( H = \rho V_p/\mu \). However, since this field is lower than \( H_{t'} = (\{J(q_0) - J(0)\} \sigma/\mu + \rho V_p/\mu) \), there must be another transition from oblique ferromagnetism to sinusoidal oscillation about the field direction before \( H \) reaches \( \rho V_p/\mu \). Comparing energy expressions (3.52) and
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Fig. 7. Effect of anisotropy energy of sixfold symmetry within the easy plane and effect of external field on the spin structure; the field is applied along one of the hard axes.

Transition at $H_0'$ in both (a) and (b) is of the second kind and other transitions are all of the first kind. When the anisotropy energy is high, the spin vectors once aligned ferromagnetically in a direction oblique to the field at intermediate fields come into sinusoidal oscillation at higher fields and then align parallel to the field direction. $H_0''$ is the field at which the center of the sinusoidal oscillation begins to deviate from the field direction and $H_0'''$ is the field at which infinitesimal oscillation sets in for the oblique parallel alignment; they have no real meaning.
(3·36), with supplementary conditions (3·51) and (3·39), respectively (the sign of $V_p$ being reversed in (3·36) and (3·39)), we have the equation to determine this transition field $H_{t}^*$ as

$$\frac{3(p^2X-Y)^2}{2(p^2-p^3)X} - \frac{(1+p^2X-Y)^2}{(2+p^2)+(p^4-p^3)X} = 0. \quad (H=H_{t}^*) \quad (3·54)$$

Here we have assumed that $\varphi$ is small and its higher powers than the fourth can be neglected. When $X$ is large, the transition field is calculated from (3·54) to be approximately $\rho^2V_p-\left(2+\sqrt{6}\right)\{J(q_0)-J(0)\}$.

Figures 7(a) and 7(b) show diagrams for various spin arrangements. The curves for $H_{t}^{**}$ in these figures were actually calculated without making the approximation which lead to (3·53), so that they are a little more accurate. It may be remarked that these results are valid only when the anisotropy energy is expressed in the special form given by (3·1).

§ 4. Magnetization process in modified screw systems at low temperatures

(i) Introduction and general consideration

Various types of spin arrangement including linear sinusoidal arrangement, conical arrangement, and other modifications of the screw structure have been observed in heavier rare-earth metals. These have been considered to result from an interplay between the exchange energy and the crystalline field anisotropy energy which stabilizes the $c$-axis or a cone having the $c$-axis as the symmetry axis. In the present section we shall study the effect of external field on these modified screw structures.

If the $c$-axis is stabilized by the anisotropy energy, the magnetic moment vectors of the $c$-layers will tend to be confined in the $c$-direction. As a result, we have a longitudinal sine oscillation along the $c$-axis at sufficiently high temperatures, as we saw in § 3 (i) and (ii). This oscillation may change at low temperatures into an elliptic oscillation in a plane which contains the $c$-axis when the anisotropy energy is small, and the ellipticity, will become smaller as temperature is lowered. At the same time, a certain modulation in the rotation of the moment vectors will take place. The longitudinal sine oscillation may also persist down to absolute zero when the anisotropy energy is large, transforming into an antiphase domain structure as temperature is lowered. The magnetization process in such cases is expected to be somewhat similar to that studied in § 3 (i) and (ii), although in the present case there is no anisotropy energy confining the moment vectors in any particular plane which contains the $c$-axis, so far as the anisotropy energy is assumed to be axially symmetric, so that the moment vectors may for some field strengths deviate from such a plane, even if the field is applied along the $c$-axis. However, when there is a longitudinal oscillation along the $c$-axis, of more or less purely sinusoidal character, which is pos-
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Possible only at high temperatures, the magnetization process should completely be the same as that discussed in § 3 (i) and (ii); namely, when the field is applied either along the c-axis or perpendicularly to it, the center of oscillation shifts towards the field direction and above the field given by (3·12) and (3·23), respectively, the moment vectors point parallel to the field direction. When we have an elliptic or circular arrangement whose plane contains the c-axis, we may or may not have the same magnetization process as that discussed in § 3, depending on whether the anisotropy energy is large or not, but it is easy to imagine the magnetization process in this case to be as follows. When the field is applied along the c-axis, the initial structure of elliptic or circular oscillation will transform into a conical arrangement above a certain critical field, provided the anisotropy energy is axially symmetric, and then into parallel alignment above another critical field; when the field is applied perpendicularly to the c-axis, the plane of the moment vectors will first become perpendicular to the field direction and the moment vectors will be tipped towards the field direction, then for higher fields they will oscillate sinusoidally about the field direction in the plane which contains the c-axis and the field direction, either passing through a state of oblique alignment or without passing it, and finally become parallel to the field direction.

If a cone symmetrical about the c-axis is stabilized, we may have, depending on whether the exchange energy is large or small compared with the anisotropy energy, a screw arrangement with its rotation plane oblique to the c-axis or a conical arrangement, the cone being a little wider than the cone of the minimum anisotropy energy. Whether we have a conical arrangement or not depends also on the turn angle $\alpha_0$; for larger values of this angle we do not necessarily have a one-side conical arrangement, namely, the magnetization vectors of the layers may oscillate alternately on the up-cone and the down-cone.

It is not the purpose of the present section to study thoroughly all the conceivable complicated cases. We shall rather confine ourselves to a few typical cases of interest and study the main features of the magnetization process for them. We shall also confine ourselves to low temperatures since oblique screw arrangement and conical arrangement have been observed hitherto only at low temperatures. The magnetization of each layer will, therefore, be assumed to be completely saturated, while the anisotropy energy will be considered as being a function of temperature, or rather as a parameter whose different values correspond to different temperatures.

It will be assumed that the anisotropy energy is axially symmetric and is expressed as

$$\sum_n W_n (\cos^2 \theta_n),$$  \hspace{1cm} (4·1)

where $\theta_n$ is the angle between the magnetic moment vector of the n-th layer.
and the $c$-axis, or the $z$-axis as we shall call it.

(ii) **Screw arrangement with an inclined rotation plane**

If the anisotropy energy stabilizing a cone is small compared with the interlayer exchange energy, we have a screw arrangement in which the spins rotate uniformly in a plane which is in general oblique to the $z$-axis. The orientation of this plane is determined from the minimum of the anisotropy energy, namely,

$$\langle W_a (\sin^2 \theta \cdot \cos^2 (nq_0 + \alpha)) \rangle_n = \text{min},$$

where $\theta$ is the angle between the normal of the plane and the $z$-axis and $\langle \rangle_n$ means the average over $n$. This average can also be expressed as

$$\langle W_a (\sin^2 \theta \cdot \cos^2 (nq_0 + \alpha)) \rangle_n = \frac{1}{2\pi} \int_0^{2\pi} W_a (\sin^2 \theta \cdot \cos^2 \varphi) d\varphi.$$

When a field $H$ is applied along the $x$-axis, which is perpendicular to the $z$-axis, the normal to the rotation plane will come into the $xz$-plane. This is because the susceptibility perpendicular to the rotation plane, $\chi_\perp$, is greater than the susceptibility parallel to it, $\chi_0$. In the presence of a field, the angle $\theta$ will be determined from

$$-\frac{1}{2} H^2 (\chi_0 \cos^2 \theta + \chi_\perp \sin^2 \theta) + \langle W_a (\sin^2 \theta \cdot \cos^2 \varphi) \rangle_\varphi = \text{min}.$$  

as long as the change of the anisotropy energy due to the rearrangement of the moment vectors caused by the application of the field can be neglected. $\theta$ will increase with field strength and will tend to $\pi/2$. Although the variation of $\theta$ with field strength will depend on the functional form of $W_a$, and thus the critical field at which the rotation plane becomes perpendicular to the field direction cannot in general be calculated without the knowledge of $W_a$, we may consider the simplest case where $\theta$ varies continuously with field strength and tends to $\pi/2$. In this case the critical field $H_\theta$ is given by

$$H_\theta = \left[2 \langle W_a' (\cos^2 \varphi) \cos^2 \varphi \rangle_\varphi / (\chi_\perp - \chi_0) \right]^{1/2}.$$  

(One differentiates $(4 \cdot 3)$ with respect to $\sin^2 \theta$ and puts $\vartheta = \pi/2$.)

With a further increase in the field strength the moment vectors will begin to rotate on an elliptic cone whose axis is the field direction and whose longer principal axis is parallel to the $z$-axis, since the anisotropy energy is lower in the latter direction. Above another critical field, the $y$-components of the moment vectors will vanish and they will oscillate sinusoidally in the $xz$-plane. This critical field from cone to transverse sinusoidal oscillation, $H_\alpha$, can be calculated with the method developed in paper I, provided the amplitude of the sinusoidal oscillation at this field is so small that the power series expansion of the energy with respect to this amplitude can be made; in particular,
the amplitude must be smaller than the minimum angle between the conical surface of minimum anisotropy energy and the $x$-axis. The result is
\[
\mu H_{ct} = \frac{2 - \beta_2}{\beta_2} W_a'(0),
\]
where $\beta_2 = \frac{J(q_0) - J(2q_0)}{J(q_0) - J(0)}$ as before. With a further increase in the field strength, the amplitude of the oscillation in the $xz$-plane decreases and finally it vanishes at and above
\[
\mu H_0 = \frac{J(q_0) - J(0)}{2 W_a'(0)} - 2 W_a'(0).
\]
It is noted that $W_a'(0)$ is negative in the present case, so that $H_0 > H_{ct}$. Figure 8 shows schematically the structure changes mentioned here.

![Figure 8](image)

Fig. 8. Changes of spin structure caused by external field in the case where oblique screw is stabilized at $H=0$.

When the field is applied along the $z$-axis, $\theta$ will decrease and tend to zero. The critical field at which $\theta$ becomes zero is given by $[-W_a'(0)/(x_1 - x_0)]^{1/2}$. However, a discontinuous transition to $\theta=0$ at a lower field cannot be excluded, and it depends on the functional form of $W_a$. Since axial symmetry was assumed for the anisotropy energy, the moment vectors will become distributed on a cone for $\theta=0$ and they will finally become parallel to the $z$-axis above the field given by
\[
\mu H'_0 = \frac{J(q_0) - J(0)}{2 W_a'(1)} - 2 W_a'(0).
\]

(iii) Magnetization of a conical arrangement; the field is applied along the $x$-axis

If the anisotropy energy which stabilizes a cone is not small compared with the interlayer exchange energy and if $q_0$ is small, the moment vectors of the layers will arrange themselves on a cone. The angle $\theta_0$, which the moment vectors make with the $c$-axis, is determined from the minimum of the total energy:
\[
\frac{E}{N} = -\frac{1}{2} J(q_0) + \frac{1}{2} (J(q_0) - J(0)) \cos^2 \theta_0 + W_a (\cos^2 \theta_0) = \min,
\]
namely, from
\[
\frac{1}{2} (J(q_0) - J(0)) + W_a' (\cos^2 \theta_0) = 0.
\]
Whether we have a conical arrangement or a screw arrangement with inclined
rotation plane depends on the relative heights of the minimum value of (4·7) and the minimum value of (4·2) minus $J(q_0)/2$. In the present case, we assume that the former is lower.

When the field, applied along the $x$-axis, is small, the axis of the cone of the moment arrangement will be tilted towards the field direction. At the same time, each of the moment vectors will be tilted towards the field direction proportionally to their $y$-component. Therefore, we put

$$
\theta_n = \theta_0 + 2\xi \cos(nq_0 + \alpha),
$$

$$
\varphi_n = nq_0 + \alpha - 2\xi \sin(nq_0 + \alpha),
$$

(4·9)

$\theta_n$ and $\varphi_n$ being the polar and azimuthal angles of the moment vector of the $n$-th layer. We then expand the total energy in powers of $\xi$, $\zeta$, and $H$.

The total energy in the most general case can be written

$$
E = -\frac{1}{2} \sum_m \sum_n J_{m-n} \{ \cos \theta_m \cos \theta_n + \sin \theta_m \sin \theta_n \cos(\varphi_m - \varphi_n) \}
$$

$$
- \mu H \sum_n \sin \theta_n \cos \varphi_n + \sum_n W_a (\cos^4 \theta_n),
$$

(4·10)

so that by substituting (4·9) for $\theta_n$ and $\varphi_n$ and taking terms up to those quadratic in $\xi$, $\zeta$, and $H$ in the expansion of the above energy expression, we have for the energy per atom

$$
- \frac{1}{2} J(0) \cos^2 \theta_0 - \frac{1}{2} J(q_0) \sin^2 \theta_0 + W_a (\cos^4 \theta_0)
$$

$$
+ \frac{1}{2} \{ 2J(q_0) - J(0) - J(2q_0) \} \xi^2 \sin^2 \theta_0
$$

$$
- \{ J(0) - J(2q_0) \} \xi \zeta \sin \theta_0 \cos \theta_0
$$

$$
+ \frac{1}{2} \{ J(0) - J(2q_0) - 4W_a' (\cos^2 \theta_0) (1 - \tan^2 \theta_0) \}
$$

$$
+ 8W_a'' (\cos^2 \theta_0) \sin^2 \theta_0 \zeta^2 \cos^2 \theta_0
$$

$$
- \mu H (\xi \sin \theta_0 + \zeta \cos \theta_0).
$$

(4·11)

In order that the original conical arrangement be stable, the quadratic form with respect to $\xi$ and $\zeta$ in the above expression must be positive definite, namely, using (4·8) the following inequality must hold:

$$
2\beta_2 + (1 + \beta_1) \gamma_0 > 0,
$$

(4·12)

where

$$
\gamma_0 = \frac{4W_a'' (\cos^2 \theta_0) \sin^2 \theta_0}{J(q_0) - J(0) - \tan^2 \theta_0}
$$

(4·13)

and $\beta_1 = \{ J(q_0) - J(2q_0) \}/\{ J(q_0) - J(0) \}$ as before. Since $W_a'' (\cos^2 \theta_0)$ is posi-
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tive because (4·7) is minimum, \( \gamma_0 \) can be positive when \( J(q_0) - J(0) \) is small and \( \theta_0 \) is not too close to \( \pi/2 \), so that (4·12) will be satisfied. Minimizing (4·11) with respect to \( \xi \) and \( \zeta \), we have its minimum value as

\[
-\frac{1}{2} J(0) \cos^2 \theta_0 - \frac{1}{2} J(q_0) \sin^2 \theta_0 + W_a (\cos^2 \theta_0)
- \frac{1}{2} \frac{2 \beta_0 + \gamma_0}{2 \beta_0 + (1 + \beta_0) \gamma_0} \frac{\mu^2 H^2}{J(q_0) - J(0)}.
\]  
(4·14)

The coefficient of \(-H^2/2\) in (4·14) is the initial susceptibility of the system under consideration (compare it with (2.22)).

Now, at high fields, we may expect that the moment vectors oscillate in the vicinity of a certain polar angle \( \theta \) and zero azymuthal angle. We therefore put

\[
\theta_n = \theta + 2 \xi \cos (nq_0 + \alpha),
\]

\[
\phi_n = 2 \xi \sin (nq_0 + \alpha),
\]

and expand the energy in powers of \( \xi \) and \( \zeta \). We have no linear term, and up to quadratic terms we have

\[
\frac{E}{N} = -\frac{1}{2} J(0) - \mu H \sin \theta + W_a (\cos^2 \theta)
+ \left[ - (J(q_0) - J(0)) \sin \theta + \mu H \sin \theta \right] \xi^2
+ \left[ - (J(q_0) - J(0)) + \mu H \sin \theta - 2 W_a' (\cos^2 \theta) (\cos^2 \theta - \sin^2 \theta)
+ 4 W_a'' (\cos^2 \theta) \sin^2 \theta \cos^2 \theta \right] \zeta^2.
\]  
(4·16)

The value of \( \theta \) is to be determined from the minimum of the first line of (4·16), namely, from

\[
\mu H + 2 W_a' (\cos^2 \theta) \sin \theta = 0,
\]  
(4·17)

provided the amplitudes are small. Eliminating \( H \) from (4·16) by the use of (4·17), we have for the quadratic terms of (4·16) the following expression:

\[
\left[ - (J(q_0) - J(0)) - 2 W_a' (\cos^2 \theta) \right] \xi^2 \sin \theta
+ \left[ - (J(q_0) - J(0)) - 2 W_a' (\cos^2 \theta) \cos^2 \theta + 4 W_a'' (\cos^2 \theta) \sin^2 \theta \cos^2 \theta \right] \zeta^2.
\]  
(4·18)

The solution we are considering can be imagined by looking at (4·17) and (4·18). Equation (4·17), when multiplied by \( \cos \theta \), is the torque equation. Thus, at \( H=0 \), the \( \theta \) value has to be equal to the angle of the easy cone, or the angle of the minimum of the anisotropy energy, and thus has to be smaller than \( \theta_0 \). With increasing \( H \) the \( \theta \) value must increase. It will pass through \( \theta_0 \) and will tend to \( \pi/2 \), varying continuously or with jumps, depending on the functional form of the anisotropy energy. If the anisotropy energy has a single
minimum between $\theta=0$ and $\theta=\pi/2$ and varies smoothly without pronounced undulation, $\theta$ will increase continuously, and the coefficient of $\xi^2 \sin^2 \theta$ in (4·18) will behave so that it vanishes at $\theta=\theta_0$ in virtue of (4·8), is negative when $\theta_0<\theta$ and positive when $\theta>\theta_0$. More exactly speaking, this is so if $-W_\alpha'(\cos^2 \theta)$ is a monotonically increasing function of $\theta$ for $\theta>\theta_0$. Therefore, we can expect an oscillation in $\varphi$ when $\theta<\theta_0$ and no oscillation in $\varphi$ when $\theta>\theta_0$. The coefficient of $\xi^2$ behaves differently. Even in the case where the anisotropy energy varies in the simple way mentioned above, it attains a maximum in the neighborhood of the angle of the minimum of the anisotropy energy and then decreases with increasing $\theta$. At $\theta=\pi/2$ it has a negative value, namely, $-\{J(q_0)-J(0)\}$. Therefore, oscillation in $\theta$ must take place within a certain interval of $\theta$ ranging to $\pi/2$.

In this connection, two cases may be discriminated, namely, the case where $\gamma_\theta$ is positive and the case where it is negative. At $\theta=\theta_0$, the coefficient of $\xi^2$ in (4·18) can be written as $\gamma_\theta \{J(q_0)-J(0)\} \sin^2 \theta_0$, using (4·8) and (4·13), while the coefficient of $\xi^2 \sin^2 \theta_0$ vanishes. If, therefore, $\gamma_\theta$ is positive, no oscillation in the polar angle will take place in the neighborhood of $\theta_0$, while $\varphi$ will oscillate when $\theta$ is less than $\theta_0$. For this case of positive $\gamma_\theta$ we shall calculate the $\xi^2$-term which has to be added to the energy expression (4·16). It is convenient to put

$$\sin(\varphi_\alpha/2) = \xi \sin(nq_0 + \alpha), \quad (4·19)$$

in place of (4·15), since, then, the fourth order term will not contain $H$. A simple calculation gives the $\xi^2$-term as

$$\frac{1}{4} \{3J(q_0)-2J(0)-J(2q_0)\} \sin^3 \theta \cdot \xi^2. \quad (4·20)$$

By minimizing the sum of the $\xi^2$-term and (4·20) with respect to $\xi$ we have

$$\xi^2 = 2 \left[ \{J(q_0)-J(0)\} \sin^2 \theta - \mu H \sin \theta \right] \frac{3J(q_0)-2J(0)-J(2q_0)}{3J(q_0)-2J(0)-J(2q_0)} \sin^3 \theta, \quad (4·21)$$

provided the numerator of the right-hand side is positive, or $\theta<\theta_0$. With this result, the energy expression becomes

$$\frac{E}{N} = - \frac{1}{2} J(0) - \mu H \sin \theta + W_\alpha(\cos^2 \theta)$$

$$- \frac{\{J(q_0)-J(0)\} \sin \theta - \mu H}{3J(q_0)-2J(0)-J(2q_0)}.$$  \quad (4·22)

Because of the last term in this expression, the equation to determine $\theta$ as a function of $H$ is, to be more exact, no longer (4·17), but it is

$$\mu H + 2W_\alpha'(\cos^2 \theta) \sin \theta + \frac{2}{2+\beta_1} \left[ \{J(q_0)-J(0)\} \sin \theta - \mu H \right] = 0. \quad (4·23)$$
The angle $\theta$ will vary more slowly with $H$ than it would without the last term of this equation, but we shall not go further into this variation. Our main interest will be in asking whether or not there is a structure change between the initial state of magnetization (at low fields) and the oscillational state with an amplitude given by (4.21). The energy of the initial state given by (4.14) may be extrapolated to a field $H$ for which $\theta = \theta_0$, namely, to $H = (J(q_0) - J(0)) \sin \theta_0 / \mu$, and when this is done, it can be shown by a simple calculation that this energy value is higher than the energy value obtained from (4.22) by putting $\theta = \theta_0$. Therefore, we may conclude that there is a transition at a field lower than $(J(q_0) - J(0)) \sin \theta_0 / \mu$ between the initial state of conical arrangement (modified by the field) and the state in which $\varphi$ oscillates. This transition field may be calculated by comparing (4.14) and (4.22), $\theta$ in the latter being determined from (4.23). Although we have not carried out this calculation, the value of $\theta$ corresponding to this transition field should be smaller than $\theta_0$.

Above this transition field, the amplitude of oscillation in $\varphi$ will decrease with increasing field and the value of $\theta$ will increase up to $\theta_0$, where the oscillation will cease. The field for $\theta = \theta_0$ is given by

$$\mu H_e = (J(q_0) - J(0)) \sin \theta_0.$$  \hspace{1cm} (4.24)

With a further increase in $H$, the angle $\theta$ between the aligned moment vectors and the $c$-axis increase, in accordance with (4.17), until $H$ reaches the value for which the coefficient of $\xi^2$ in (4.16) vanishes. The oscillation in $\theta$ will begin, and this oscillation will probably further increase until $\theta$ reaches $\pi/2$; these changes are, however, not necessarily continuous, depending on the functional form of the anisotropy energy. After $\theta$ reaches $\pi/2$, the oscillation will diminish with increasing $H$ and it will finally vanish.

In the case of negative $r_0$, it can be shown that the energy (4.14) at $H = (J(q_0) - J(0)) \sin \theta_0 / \mu$ is lower than the value of the energy (4.22) at the same field, so that it is probable that the conical arrangement transforms directly into a state in which $\theta$ oscillates but $\varphi$ does not. However, the calculation in this case is complicated because the energy expression (4.16) is not sufficient for discussing this transition; we have to calculate energy terms up to the fourth order in $\xi$ and $\zeta$.

If $\theta$ tends continuously to $\pi/2$, the field for $\theta = \pi/2$ can be obtained from (4.17) as far as the amplitude of oscillation can be assumed to be small, namely, as*

$$\xi \mu H_{e/2} = -2W_\alpha'(0).$$  \hspace{1cm} (4.25)

Above this field the energy expression up to the second order in $\xi$ and $\zeta$ is given by

---

* $-2W_\alpha'(\cos^2 \theta) = (dW_\alpha/d\theta)/(\cos \theta \sin \theta)$, so that $W_\alpha'(0)$ may not be zero though $(dW_\alpha/d\theta)_{\pi/2}$ is zero.
\[
\frac{E}{N} = -\frac{1}{2} J(0) - \mu H + W_a(0) + \left[-\{J(q_0) - J(0)\} + \mu H\right] \xi^2 \\
+ \left[-\{J(q_0) - J(0)\} + \mu H + 2W'_a(0)\right] \zeta^2. 
\] (4.26)

The coefficient of \(\xi^2\) has been assumed to be positive for this range of field (since the absolute value of the second term in (4.17) was assumed to be an increasing function of \(\theta\), so that, referring to (4.8), \(\mu H\) must be larger than \(J(q_0) - J(0)\)), so that \(\xi\) must vanish. On the other hand, the coefficient of \(\zeta^2\) remains negative until the field reaches the value

\[
\mu H_b = \{J(q_0) - J(0)\} - 2W'_a(0). 
\] (4.27)

Above this field, all the moment vectors will align parallel to the field applied perpendicularly to the original cone axis. The assumption that \(2W'_a (\cos^2 \theta) \sin \theta\) is an increasing function of \(\theta\) for \(\theta > \theta_b\) is essential in the above argument. If otherwise, some or all of the intermediate state we have discussed may be skipped over.

Figure 9 represents schematically the structure changes which we have discussed in the present subsection.

---

**Fig. 9.** Changes of spin structure caused by external field in the case that conical structure is stabilized at \(H=0\). The anisotropy energy is assumed to have a single minimum between \(\theta = 0\) and \(\theta = \pi/2\) and to vary smoothly.

---

### § 5. Summary of §§ 2-4

We summarize here the assumptions, the mathematical methods, and the main results of §§ 2-4, as the descriptions in these sections are lengthy and the results obtained are various.

The system we have treated consists of equidistantly spaced ferromagnetic layers of spins exchange-coupled with coupling constants \(J_0\) (within the same
layer), $J_1$ (between neighboring layers), $J_2$ (between next-neighboring layers), etc. In the absence of external magnetic field, the magnetic moment vectors of the layers form a screw structure or a modified screw structure with a wave number $q_0$ at which the Fourier series

$$J(q) = \sum_{n} J_{1n} \exp(inq) = J_0 + 2J_1 \cos q + 2J_2 \cos 2q + \cdots$$

takes its absolute maximum value. The value of $q_0$ should not be equal to $\pi/2$ and $\pi/3$ (and of course 0 and $\pi$) for the reason mentioned in paper I, nor should it be close to $\pi/p$ when there is an anisotropy energy of $p$-fold symmetry; $q_0$ is often assumed to be small in order to make ferromagnetism appear more easily than antiferromagnetism. The exchange coupling constants were assumed to be unaffected by applying a field so that $q_0$ also is not affected. Under this assumption, the wave number of any periodic structure which appears with varying external field is very close to or exactly equal to $q_0$, so that it was assumed to be $q_0$.

Our calculations are based on the Weiss molecular field approximation. We start, therefore, with Eq. (2·2), which equates the molecular field that would give rise to a statistical average, $\mu \sigma_n$, of the atomic magnetic moment on the $n$-th layer at temperature $T$ to the external field plus exchange field. We make use of expansion (2·3) in powers of $\sigma_n^2 - \sigma^4$, where $\sigma$ corresponds to the value of $\sigma_n$ at zero external field, and take the first two terms for the actual calculations. When we take into consideration an anisotropy energy of the form (3·1) in § 3, Eq. (2·2) is replaced by Eq. (3·2), which includes the effective field due to this anisotropy energy, but we still use the expansion (2·3) for finite temperatures. For absolute zero, it was more convenient to start with the torque equation (3·4) or with the principle of minimum energy expressed by (3·5), both of which being derivable from (3·2). The principle of minimum energy was used also in § 4.

In §§ 2 and 3 it was assumed that the magnetic moment vectors of the layers were confined within the easy plane of magnetization, the field being applied in the same plane. In § 2 we dealt with the case of no anisotropy within the easy plane and in § 3 the case in which there is anisotropy. In § 2 (i) the second term of the power series expansion (2·3) was considered as a perturbing term, while in § 2 (ii) a more rigorous treatment was made by expanding the components of the magnetic moment vectors in Fourier series and solving a system of non-linear equations for the Fourier coefficients. The results for the magnetization curve are shown in Fig. 1 (a) and (b), the former being valid between the Néel temperature, $T_N$, and a temperature which corresponds to $\gamma = 8/11$ and the latter at lower temperatures, where $\gamma$ is defined by (2·13) and is such a measure of temperature that it vanishes at $T_N$ (see (2·13a)) and tends to infinity as $T$ approaches to zero. There are two kinks in the curve of Fig. 1(a); between zero field and $H_t$ the structure is essentially a screw, the magnetization in-
duced by the field being added to each layer magnetization; between $H_t$ and $H_0$ there is a sinusoidal oscillation transverse to the field direction whose amplitude diminishes with increasing field and finally vanishes at $H_0$; above $H_0$ all the moments of the layers align parallel to the field direction. $H_0$ and $H_t$ are given respectively by (2·8) and (2·19) and are functions of temperature. The slope of the lower branch of the magnetization curve at $H_t$ tends to infinity for $\gamma=8/11$ and becomes negative for lower temperatures, so that a discontinuous transition must occur at a field $H_0$ for such lower temperatures, as seen in Fig. 1(b). These results are valid at high enough temperatures for which $\gamma$ is small compared with $\beta_s$ defined by (2·21).

In § 2 (iii) was studied the case of arbitrary temperature. Exact treatment was, however, possible only for small field and in the vicinity of $H_0$. The susceptibilities for small fields and for fields less than and greater than $H_0$ are given by (2·22), (2·23), and (2·24), respectively, and their temperature dependence is shown in Fig. 2. Using the initial susceptibility and the susceptibility below $H_0$ and extrapolating the corresponding free energy expressions into the range of medium field strength, and comparing them, the transition field $H_{tt}$ was determined to be (2·29); this field is a little less than $H_0/2$.

In § 3 two cases were distinguished: the field is parallel to one of the easy axes and the field is parallel to one of the hard axes. In § 3 (i) and (ii) we treated a system with twofold anisotropy within the easy plane at high temperatures, with a method similar to that we used in § 2 (ii). In this case, the spin structure is sinusoidal between $T_N$ and the temperature corresponding to $\gamma=\gamma_1$, where $\gamma_1$ is defined by (3·9) and is proportional to the anisotropy constant. When the field is applied along one of the easy axes, we have the results shown in Fig. 3; various cases arise, depending on the value of $\gamma_1$. For example, when $\gamma_1$ is less than 1/2, we have Fig. 3(a); in the highest temperature range, there is a sinusoidal oscillation at low field, which transforms into parallel alignment at $H_0$ (from longitudinal sinusoidal oscillation to no oscillation); in the intermediate temperature range, we have elliptic oscillation at low field, which transforms into transverse sinusoidal oscillation at $H_{tt}$ (from elliptic to transverse) and then into parallel alignment at $H_0$; in the lowest temperature range, the same occurs as in the intermediate temperature range, but the first transition is discontinuous, of or of the first kind, so that the corresponding field is written as $H_{tt}$, other transitions being of the second kind in this case. When the field is along one of the hard axes, the structure changes are as shown in Fig. 4; also there are various cases. A peculiarity in this case is that for high anisotropy energy a parallel alignment oblique to the field direction (near the neighboring easy direction) appears as seen in Figs. 4 (b)-(d).

In § 3 (iii) and (iv) the behavior at low temperatures of a system with an anisotropy energy of $p$-fold symmetry within the easy plane was studied, particularly for $p=6$. The magnetization of each layer was assumed as saturat-
ed \((\sigma = 1)\) in most part of these subsections, but equations and formulas obtained can easily be converted into those for \(\sigma \neq 1\) if one can assume that the field can scarcely affect the magnitude of the magnetization. The anisotropy constant was, however, taken as a parameter which could vary with temperature. The mathematics is a little complicated in that when there is a sinusoidal oscillation, the anisotropy term cannot be expanded in powers of the amplitude of this oscillation and it has to be taken into account exactly. The results for \(p = 6\) are as shown in Figs. 6 and 7. In Fig. 6 (field parallel to an easy axis) we see the following features: for small anisotropy constant there is a discontinuous transition at \(H_t\) from screw to sinusoidal oscillation and then a continuous transition at \(H_0\) into parallel alignment; for intermediate anisotropy constant the second transition is also discontinuous; \(H_0\) decreases rapidly in these two ranges; for high anisotropy constant, there is a direct transition from screw to parallel alignment and this transition field \(H_t\) is almost independent of the value of the anisotropy constant. In Fig. 7 (field parallel to a hard direction) we see that there are always three structures and, moreover, an oblique parallel alignment appears when the anisotropy constant is large; \(H_0\) increases rapidly with increasing anisotropy constant in this case, while \(H_t\) remains nearly constant (a prime was attached to the transition fields in order to distinguish them from those in Fig. 6).

Finally, in § 4, we studied the magnetization process in a system having an anisotropy energy of axial symmetry which gives rise to: (1) longitudinal sinusoidal oscillation along the axis, (2) elliptic oscillation in an arbitrary plane which contains the axis, (3) oblique screw structure in which the magnetic moment vectors of the layers rotate in a plane which is oblique to the axis, and (4) conical arrangement of the magnetic moment vectors with the axis of the cone coinciding with the axis of anisotropy. The first two cases were discussed only briefly and qualitatively, because the magnetization processes in these cases are similar to those discussed in § 3 (i) and (ii), except that for elliptically oscillating structure at zero field a conical structure should appear for intermediate field applied along the axis of anisotropy and that the plane ellipse should be bent into an elliptic \(\pi < \alpha < \pi\) for low field applied perpendicularly to the axis of anisotropy. The third and fourth cases were treated more fully in § 4 (i) and (ii), respectively, particularly when the field is perpendicular to the anisotropy axis. However, a full discussion was not attempted, because structure changes should be sensitive to the functional form of the anisotropy energy.

The results obtained in § 4 (i) are illustrated in Fig. 8; there are, generally speaking, three transition fields separating four structures, namely, oblique screw, cone, sinusoidal oscillation, and parallel alignment. The first transition field, \(H_a\), was calculated assuming a transition of the second kind, although a transition of the first kind is not impossible; the second transition field, \(H_t\), was calculated assuming a small amplitude of transverse component of oscillation.
at and above this field; the last transition field, $H_5$, was also assumed to arise from a transition of the second kind. The case of field parallel to the anisotropy axis was briefly discussed at the end of this subsection.

In § 4 (ii) the magnetization process for a conical structure was discussed in some detail for field perpendicular to the anisotropy axis. (The parallel case was discussed by Cooper, Elliott, Nettel, and Suhl.15) Typical changes of structure in this case are illustrated in Fig. 9. When $q_0$ is not large and the cone is not very flat, namely, when $\gamma_0 > 0$, $\gamma_0$ being defined by (4.13), six different structures appear consecutively as the field is increased, the first transition being of the first kind and the others being of the second kind. Just below the second transition at $H_6$ the magnetic moment vectors oscillate on a small part of the original cone, the part which is nearest to the field direction. Above $H_6$ they align in the meridian plane, then they begin to oscillate in this plane, the center of oscillation shifting towards the field direction and finally coinciding with it, and they still oscillate with further increase in field strength until this reaches $H_7$, above which they align parallel. When $\gamma_0 < 0$, the second and third structures are skipped over. These results are, however, valid only when the anisotropy energy varies with $\theta$ monotonically and smoothly between its minimum at $\theta_0$ and its maximum at $\pi/2$. Otherwise, some or all of the intermediate structures can be skipped over and, in some cases, there may be more jumps.

§ 6. Comparison with experimental results

Qualitative comparison between the theoretical results predicted in the present paper and experiments so far obtained will be made in the present section.

Magnetization measurements with single crystals have been made by Legvold, Spedding et al. on Dy,16 Er,17 and Ho,18 and a neutron diffraction study of Ho single crystal has been made by Koehler.19 Also a neutron diffraction study of polycrystalline MnAu, was made by Herpin and Mériel.10

In the case of MnAu, a simple helical structure with a propagation vector parallel to the crystalline tetragonal axis was found to appear below $365^\circ$K (the Néel temperature) and the turn angle was found to decrease from 51° at 300°K to 46° at 125°K and then to increase to 47° at 87°K. On application of a field in the tetragonal basal plane—since for the investigated diffraction lines, 101± and 002±, the magnetic field was applied perpendicular to the plane containing the incident and diffracted beams so that it was in the basal plane of the reflecting crystallites—the transition helical-sinusoidal-ferromagnetic was observed, the first transition being at 9.6 koe and the second transition in the range of 14 to 16 koe. If we assume no anisotropy within the basal plane and identify these transition fields with $H_n$ and $H_5$ of § 2, then it appears that the ratio $H_n/H_5$ observed is too large, since theory predicts for it a value slightly less than 1/2. If there is an anisotropy in the basal plane, this ratio should great-
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Magnetically be affected by changing the direction of the field. A study with a single crystal is, therefore, desirable.

Dy also shows a simple helical arrangement between 179°K (the Néel temperature) and 87°K (below which ferromagnetism appears). Magnetization measurements made from 160°K to 90°K with a field applied in the hexagonal basal plane show an apparently discontinuous increase of magnetization at a field which varies with temperature, followed by a gradual increase going to saturation. If we identify this field with $H_u$ of § 2, the theory predicts that it should be nearly proportional to the magnetization of the individual layers for zero field as a function of temperature, because $H_u$ is proportional to the latter and $H_u/H_o$ is nearly equal to 1/2. Thus, $H_u$ should increase with decreasing temperature. The experimental fact is just the contrary: the transition field decreases linearly with decreasing temperature, from 10.5 koe at 160°K to nearly zero at 90°K. There is no appreciable anisotropy in the basal plane down to 100°K. It is noted, however, that observation showed that the turn angle decreased linearly from 43.2° at 179°K to 26.5° at 87°K, which means that the other factor, $J(q_0) - J(0)$, in Eq. (8) decreases, possibly over-compensating the effect of the increase in magnetization. In this connection, it is suggested that it may be interesting to measure closely the magnetization curve in the range between 160°K and 179°K, where the magnetization of the individual layers varies rapidly with temperature; namely, we may have a sinusoidal oscillation about the field direction in the immediate range above the transition field.

In Er, in the temperature range of 80°K to 52°K, where we have a longitudinal sinusoidal oscillation along the c-axis, and also in the range of 52°K to 20°K, where there is an additional transverse oscillation, experiment seems to show a single transition from oscillation to ferromagnetism. For the former temperature range, our prediction supports this, as one may see from Figs. 3 and 4.

Holmium was investigated in detail by Koehler by neutron diffraction. The Néel temperature is 133°K; between this and 35°K the structure is helical with $q_0$ varying linearly from 50° to 36°, between 35°K and 19°K it is still helical but $q_0$ remains constant, and below 19°K the layer magnetization vectors rotate on a flat cone with a smaller value of $q_0$ which corresponds to a turn angle of 30°. Above 80°K there is no appreciable anisotropy within the basal plane as far as neutron diffraction observation could confirm, although magnetization measurements by Legvold, Spedding et al. show some anisotropy. In this temperature range, up to 100°K, a transition to a sinusoidally oscillating structure was observed by applying a field in the basal plane, which was then followed by another transition to ferromagnetism. In the range below, down to 40°K, where there is anisotropy, Koehler observed two oscillating structures which appeared one after another and showed high intensity of harmonics. The appearance of two oscillating structures is not understandable by the present theory. When the field was applied below this temperature along the b-axis (the easy
direction of magnetization in the basal plane, transitions helix to oscillation, then to ferromagnetism were observed down to 35°K. Then, down to 20°K, a direct transition from helix to ferromagnetism was observed. This can be understood by referring to Fig. 6(a) or (b). When the field was applied along the a-axis (the hard direction), between 40°K and 32°K, still two phases of oscillation were observed between helix and ferromagnetism, of which the second phase had a small ferromagnetic component perpendicular to the a-axis; this again cannot be understood by our theory—we looked for the possibility of a structure in which the moment vectors oscillate about a direction which is oblique to the hard axis, but we found no stable structure of this kind (see § 4). Further below, between 32°K and 25°K, with a field applied along the a-axis, transitions helix to oscillation, then to ferromagnetic alignment along the b-axis were observed and between 25°K and 20°K a direct transition from helix to ferromagnetic alignment along the b-axis was observed; the latter can be understood by referring to Fig. 7(a) or (b), but our theory does not predict an intermediate phase of oscillation. In all the above, the transition fields decreased with decreasing temperature, as it was the case in Dy, and the wave numbers of the various phases decreased as they appeared one after another, the latter fact being outside our simplified theory which assumed a constant wave number. Below 20°K there is a conical structure at zero field. In this temperature range, Koehler observed a transition from cone to ferromagnetism oblique to the applied field and then transition to ferromagnetism parallel to the b-axis. Referring to Fig. 9 and to what was mentioned at the end of § 5, these transitions can be understood qualitatively, although our theory did not assume any azymuthal anisotropy and, therefore, has to be so modified as to make the ferromagnetism appear along the easy axis, even when the field is along the hard axis.

The comparison between theory and experiment described above leaves a number of unsatisfactory points on the side of the theory, while there is also a point which has to be improved on the side of the experiment, namely, the point that the single crystals used do not seem satisfactory. This is indicated by the fact the transitions observed were not always sharp and two phases often coexisted over a certain range of field.

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