

K. YOSIDA: One different point between Dr. Kaplan's treatment and ours is in the functional form of the anisotropy energy. Dr. Kaplan used the dipole-dipole type of anisotropy but we used the one-ion anisotropy which seems to be more realistic for rare-earth metals. If one uses the one-ion anisotropy, the temperature change in the period of the spin modulation becomes small compared with experimental results. On the other hand, according to Dr. Kaplan, if one uses the pseudodipolar anisotropy, the temperature dependence of the period can be explained for erbium because the ratio of the amplitude of the spin modulation along the c axis to that perpendicular to the c axis changes with temperature. However, this type of anisotropy can not explain the change of the pitch with temperature for dysprosium and holmium.

H. SUHL: May I ask the authors of the preceding three papers if they have considered the nature of elementary excitations above the state with the spins all aligned along the c -axis but varying sinusoidally in magnitude?

R. J. ELLIOTT: In the paper mentioned at the end of my preprint it is shown that when the sine wave is incommensurate with the lattice the elementary excitations cannot be conveniently described as waves. For example the ferromagnetic resonance will be very broad.

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Magnetization Process of a Spin Screw System

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1. Introduction

After the discovery of screw spin structure by Yoshimori¹⁾ (first reported by Nagamiya²⁾ at the International Conference on Magnetism at Grenoble, 1958) and later by Villain³⁾ and Kaplan⁴⁾, a number of substances with this structure have been found by neutron diffraction experiment. Of particular interest are modifications of screw structure found in rare earth metals by Oak Ridge people⁵⁾ and theoretically interpreted by Elliott⁶⁾, Kaplan⁷⁾, Miwa and Yosida⁸⁾, and also by Kitano⁹⁾. These modifications come about due to the effect of the anisotropy energy inherent in these metals. Another interesting phenomenon is the change of screw structure due to the action of an applied magnetic field, first observed by neutron diffraction experiment by Herpin and Mériel

in MnAu_2 ¹⁰⁾. They and also Enz¹¹⁾ developed a theory for this phenomenon, but the present paper deals with it more generally and with more mathematical rigour*).

* Herpin and Mériel assume J_1 and J_2 only and further assume that the anisotropy energy is sufficiently large to confine the spin vectors in the plane of the layers. Enz treats the limiting case of $q_0 \rightarrow 0$. These restrictions are removed in the present paper. Furthermore, the particular cases of $q_0 = 180^\circ, 90^\circ, 120^\circ$, which were not discussed by them, are studied in detail in the present paper. The merit of the present study lies in the calculation of the fourth order energy terms of Fourier amplitudes. For low fields, the fourth order term becomes infinite for $q_0 = 180^\circ$ and 90° and vanishes for $q_0 = 120^\circ$, which show that these cases have to be dealt with carefully. For high fields, the fourth order terms are essentially important when the second order terms become negative, namely when

Yoshimori already discussed the effect of applied field and that of anisotropy energy, but his arguments were confined to the case of small applied field and small anisotropy energy. We shall remove these restrictions. The system to be considered is such that the spins form ferromagnetic layers and the anisotropy energy stabilizes the spin vectors to be in the plane of these layers, so that they form a "proper screw" structure with screw axis perpendicular to this plane in the absence of external field. The field is supposed to be applied within this plane, and we confine ourselves to the absolute zero of temperature. The exchange coupling constant between adjacent layers (say, per pair of atoms) will be denoted by J_1 , that between next-nearest-neighbouring layers by J_2 , and so on. We define $J(q)$ by

$$J(q) = 2(J_1 \cos q + J_2 \cos 2q + \dots).$$

2. Weak Field Case

The spin vectors be confined in the plane of the layers and we denote by θ_n the angle between the magnetization vector of the n -th layer and the applied field. μ be the magnetic moment of each layer (say, per atom). Then the energy of the system can be expressed as

$$\begin{aligned} E = & -J_1 \sum_n \cos(\theta_{n+1} - \theta_n) \\ & -J_2 \sum_n \cos(\theta_{n+2} - \theta_n) - \dots \\ & -\mu H \sum_n \cos \theta_n. \end{aligned}$$

Minimization of this energy is satisfied by $\theta_{n+1} - \theta_n = q_0$, where q_0 is determined from $J(q_0) = \max.$, since the energy per layer is then expressed as $E/N = -(1/2)J(q_0)$. In the special case of $J(q) = 2J_1 \cos q + 2J_2 \cos 2q$, we have $\cos q_0 = -J_1/4J_2$ ($J_2 < 0$).

For small H , we put $\theta_n = nq + \alpha + \varepsilon_n$ and assume ε_n to be small. The energy E is expanded in powers of ε_n . Further, ε_n is expanded in Fourier series:

$$\varepsilon_n = \sum_{q'} \xi_{q'} e^{inq'} \quad (\xi_{-q'} = \xi_{q'}^*).$$

$H < H_0$, H_0 being defined in the text. The calculation of the fourth order terms for H slightly smaller than H_0 and a consequent mathematical proof that there is only one non-vanishing Fourier amplitude ξ_{q_0} in such a case constitute an advance over Herpin-Mériel and Enz's paper. However, these details are not given in the present abbreviated note. The full paper will appear in Progr. Theor. Phys., Kyoto.

The first order energy term per layer consists only of the imaginary part of $-\mu H \xi_{q_0} e^{-i\alpha}$ and the second order term is calculated to be

$$\sum_{q' > 0} [J(q) - \frac{1}{2}J(q+q') - \frac{1}{2}J(q-q')] |\xi_{q'}|^2.$$

The coefficient of $|\xi_{q'}|^2$ in this expression is positive as far as q is close to q_0 , so that the minimization of energy makes all $\xi_{q'}$ with $q' \neq q_0$ vanish. Also the real part of $\xi_{q_0} e^{-i\alpha}$ should vanish, and we have

$$|\xi_{q_0}| = -i \xi_{q_0} e^{-i\alpha} = \mu H / [2J(q_0) - J(2q_0) - J(0)].$$

The corresponding energy value is given by

$$E/N = -\frac{1}{2}J(q_0) - \frac{1}{2}\mu^2 H^2 / [2J(q_0) - J(2q_0) - J(0)].$$

The value of q can be calculated by further minimizing this energy; q is equal to q_0 for $H=0$ but it varies with H^2 as

$$q - q_0 = \frac{-2J'(2q_0)\mu^2 H^2}{J''(q_0)[2J(q_0) - J(2q_0) - J(0)]^2}.$$

The magnetization is obtained by differentiating $-E/N$ with respect to H , and for $H \rightarrow 0$ the susceptibility is given by

$$\chi_0 = \mu^2 / [2J(q_0) - J(2q_0) - J(0)],$$

which, in the special case of $J(q) = J_1 \cos q + J_2 \cos 2q$, becomes

$$\chi_0 = \mu^2 / [-8J_2(1 - \cos q_0)^2(1 + 2\cos q_0 + 2\cos^2 q_0)].$$

The angle θ_n varies as

$$\theta_n = nq + \alpha - 2|\xi_{q_0}| \sin(nq + \alpha).$$

Going over to the next approximation, we obtain ξ_{2q_0} as the next non-vanishing amplitude; the energy in this case was calculated to order H^4 .

3. High Field Case

At high fields, all the spin vectors will be either completely or nearly parallel to the field direction. Assuming that the spin vectors are confined in the plane of the layers, we put $\sin(\theta_n/2) = x_n$ and expand the energy in powers of x_n . Further we put

$$x_n = \sum_q \xi_q e^{inq} \quad (\xi_{-q} = \xi_q^*).$$

Then we have, up to the fourth power,

$$\begin{aligned} E/N = & -\frac{1}{2}J(0) - \mu H + 2 \sum_q [J(0) - J(q) + \mu H] |\xi_q|^2 \\ & + \sum_q \sum_{q'} \sum_{q''} [J(q) - J(q+q') - J(q+q'')] \\ & + J(q+q'+q'')] \xi_q \xi_{q'} \xi_{q''} \xi_{-q-q'-q''}. \end{aligned}$$

The coefficient of $|\xi_{q_0}|^2$ in this expression is minimum for $q=q_0$, so that for $H > H_0$ defined by $H_0 = [J(q_0) - J(0)]/\mu$ it is positive for all q . The minimum of energy is then realized with all $\xi_{q \neq 0} = 0$; the spins are parallel to the

applied field. For $H < H_0$, the coefficient is negative in the neighbourhood of q_0 . It can be proved with the use of the fourth order terms that all ξ_q with $q \neq q_0$ vanish. The non-vanishing amplitude ξ_{q_0} is then calculated to be

$$|\xi_{q_0}| = [\mu(H_0 - H)/(6J(q_0) - 2J(2q_0) - 4J(0))]^{\frac{1}{2}}.$$

The corresponding energy is expressed as

$$\frac{E}{N} = -\frac{1}{2}J(0) - \mu H - \frac{\mu^2(H_0 - H)^2}{3J(q_0) - J(2q_0) - 2J(0)}.$$

The differential susceptibility is constant, being given by the coefficient of $-(H_0 - H)^2$ multiplied by 2. In the case of $J(q) = J_1 \cos q + J_2 \cos 2q$, we have

$$\mu H_0 = -4J_2(1 - \cos q_0)^2$$

and

$$\begin{aligned} &3J(q_0) - J(2q_0) - 2J(0) \\ &= -4J_2(1 - \cos q_0)^2(3 + 4 \cos q_0 + 4 \cos^2 q_0). \end{aligned}$$

4. Transitions at Intermediate Fields

By comparing the energy expressions ob-

tained for low and high fields, the field H_t is determined where a discontinuous transition occurs between the screwing and sinusoidally waving structures. We have

$$H_t/H_0 = [(1 + \beta)(2 + \beta)]^{\frac{1}{2}} - (1 + \beta),$$

where

$$\beta = [J(q_0) - J(2q_0)]/\mu H_0 = (1 + 2 \cos q_0)^2.$$

H_t/H_0 is close to 1/2 and at minimum 0.414. If there is an anisotropy energy in the plane and if the field is applied along one of the easy directions, the high field energy will be lowered, so that H_t and H_0 will decrease.

If q_0 is near π , $\pi/2$, or $2\pi/3$, the special configuration of antiparallel or triangular spins will be favoured in certain ranges of field, and transition will occur twice, as shown schematically in Fig. 1. These ranges shown in this figure have been determined with J_1 and J_2 only.

So far we have assumed that the spin vectors are confined within the plane because of a strong out-of-plane anisotropy energy. If the latter is small, the field applied in the plane will convert the proper screw into a

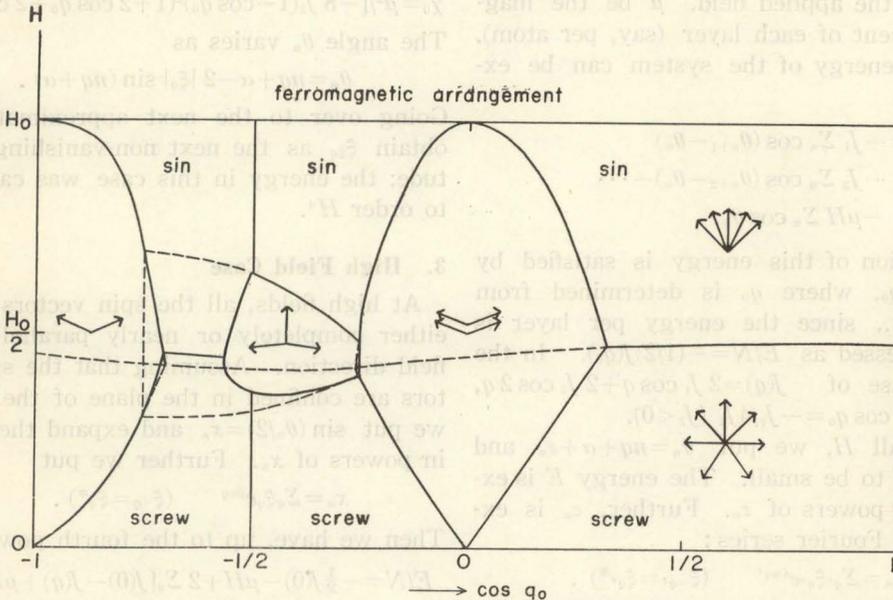


Fig. 1. Change of spin structure in the case of large anisotropy energy as a function of applied magnetic field H for various values of $\cos q_0$, calculated for $J(q) = 2J_1 \cos q + 2J_2 \cos 2q$. The domain for triangular spins in the neighbourhood of $\cos q_0 = -1/2$ and $H = H_0/2$, enclosed with solid curves, was calculated with the energy expression for low field which includes terms of H^4 ; the domain enclosed with broken curves was calculated with that which is valid only to the order of H^2 . Exact shape of this domain may therefore be different further. This domain and the domain for the pair of antiferromagnetic spins will have different shape depending on the functional form of $J(q)$.

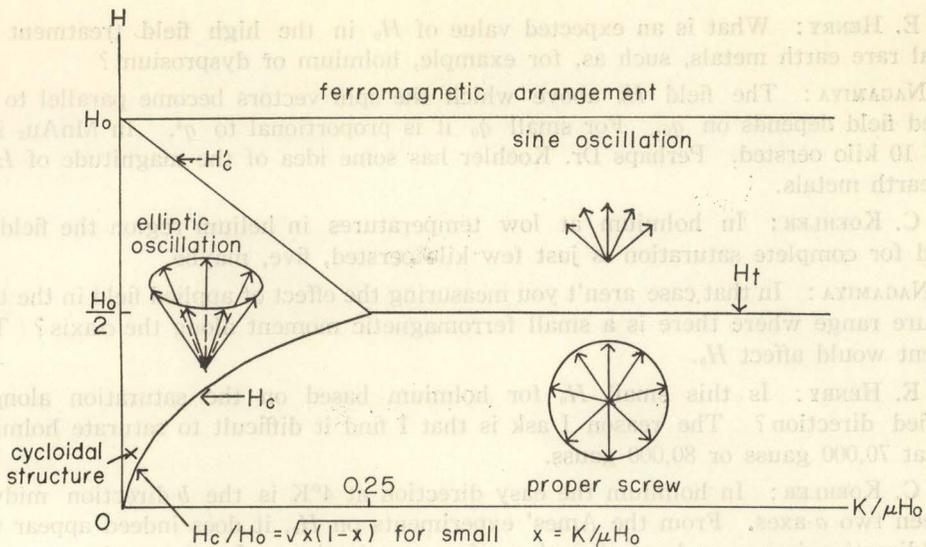


Fig. 2. Change of spin structure as a function of applied magnetic field and anisotropy constant K ; the anisotropy energy is assumed to be of the form of $K \cos^2$ (spin vector, z) with positive K . $\beta = [J(q_0) - J(2q_0)] / [J(q_0) - J(0)]$ is assumed to be large, i.e., q_0 to be small.

cycloid (the spin vectors rotating in the plane perpendicular to the field) at a field given by $H_c = [K/(\chi_{\perp} - \chi_0)]^{1/2}$, where K is the coefficient of the anisotropy energy of the \cos^2 -form and χ_{\perp} the susceptibility of the cycloid:

$$\chi_{\perp} = \mu^2 / [J(q_0) - J(0) - K].$$

At still higher fields the spin vectors rotate on an ellipsoidal cone, which finally collapses and coincides with the original plane at $H_c' = H_0 - 2K/\mu$. Such occurs for $K < \mu H_0/4$. Fig. 2 shows these transitions for the case of $\beta \rightarrow \infty$.

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DISCUSSION

K. TOMITA: Have you ever tried a quantum mechanical calculation of the similar type?

T. NAGAMIYA: No, but I do not expect any difference between the classical and quantum-mechanical treatments of the present problem.

I might add that Dr. Kitano is investigating the same problem at finite temperatures with the Weiss approximation. His preliminary results show that very similar phenomena persist to appear up to the Néel point.

K. TOMITA: We have looked at the same problem from the point of view of excitation using the method of Green's function, and found that at a certain value of the magnetic field the excitation frequency becomes negative or imaginary, i.e., the supposed structure becomes unstable. What do you think of the relation between the excitation criterion and the free energy criterion?

T. NAGAMIYA: I expect they are essentially the same, but I have to study your theory before answering definitely.

W. E. HENRY: What is an expected value of H_0 in the high field treatment for typical rare earth metals, such as, for example, holmium or dysprosium?

T. NAGAMIYA: The field H_0 above which the spin vectors become parallel to the applied field depends on q_0 . For small q_0 it is proportional to q^4 . In MnAu_2 it is about 10 kilo oersted. Perhaps Dr. Koehler has some idea of the magnitude of H_0 in rare earth metals.

W. C. KOEHLER: In holmium at low temperatures in helium region the field required for complete saturation is just few kilo oersted, five, maybe.

T. NAGAMIYA: In that case aren't you measuring the effect of applied field in the temperature range where there is a small ferromagnetic moment along the c -axis? This moment would affect H_0 .

W. E. HENRY: Is this small H_0 for holmium based on the saturation along a specified direction? The reason I ask is that I find it difficult to saturate holmium even at 70,000 gauss or 80,000 gauss.

W. C. KOEHLER: In holmium the easy direction at 4°K is the b -direction midway between two a -axes. From the Ames' experiments on H_0 , it does indeed appear that the a -direction is a very hard direction of magnetizations. In the early stages of magnetization of holmium at 64°K, there is apparently a different frequency for components of the moments parallel and perpendicular to the applied field. Is such effect predicted by your theory?

T. NAGAMIYA: No such effect is predicted by my theory, but I wish to study your experimental results.

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