

is also small numerically, and since it enters the expression of Eq. (28) along with the small weighting factor $R(K_s)$, the net effect on $dE_{\text{ex}}/dk|_{k=k_0}$ of the terms in (28) for $K_s \neq 0$ is negligible.

We have also calculated the total exchange energy, by numerical integration. The result for free electrons as given by Wigner and Seitz is -72.0 kg cal/mole. The effect of the periodic lattice has also been calculated by

Wigner and Seitz, and they find that to three decimal places, there is no difference between the free electron and periodic value. Our result is essentially the same, although we get a slightly different numerical value for the difference between the free electron and periodic case, viz., about 0.005 kg cal/mole. The smallness of this correction is of course a consequence of the rapid decrease of $g(x,y)$ and $R(K_s)$ with K_s .

Spin-Wave Theory of Ferromagnetic Thin Films

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The spin-wave theory in the Holstein-Primakoff formulation is applied to thin ferromagnetic films. A magnetic anisotropy term is included in the Hamiltonian. A discussion is given of the dependence of the magnetic properties on the temperature and on the number n of monatomic layers of the thin film. The influence of possible parallel and perpendicular anisotropy is also discussed.

I. INTRODUCTION

THE first approach in the theory of ferromagnetic thin films is due to Kittel.¹ He showed that when the thickness of the thin film is smaller than approximately 10^{-5} cm, the film becomes a single domain, magnetized in a direction parallel to its surface.

By applying Bloch's theory of spin waves, Klein *et al.*^{2,3} have studied such a single domain for various lattices. It should be mentioned that they have not taken into account the effect of magnetic anisotropy. This fact gives rise to a divergence in the final results for the magnetization if the state of the zero spin-wave vector is included in the sums which appear. With a view to avoiding this difficulty, Klein *et al.* have omitted this state in the corresponding sums, but they have not given a complete justification of this procedure.

Recently, Döring⁴ tried to give a complete discussion of the questions which arise in Klein's calculation. He justified the omission of the zero spin-wave vector state, but showed that even in this case the correct calculations lead to a strange dependence of the magnetization on the dimensions of the surface of the thin film. Döring showed also that by introducing a magnetic anisotropy term in the Hamiltonian, this situation will no longer appear, i.e., the magnetization will no longer depend on the dimensions of the surface

of the thin film. However, as is known,⁵ when the anisotropy term is taken into account, it is no longer necessary to omit the zero spin-wave vector state and the divergences in the calculation of the magnetization will no longer appear. Döring has also discussed the cyclic condition for the perpendicular axis of the film, and has recalculated the magnetization, going further than Klein *et al.* to higher order terms.⁶ It should be mentioned that Döring has not considered the case in which the magnetic anisotropy is perpendicular to the surface of the thin film.

In this paper, the spin-wave theory in the Holstein-Primakoff (H-P) formulation⁷ will be developed for thin ferromagnetic films. We shall not take into account the spin-wave interactions, which, as Dyson⁸ and Oguchi⁹ have shown, do not influence the first approximation of the H-P method. Oguchi has concluded in his discussion,¹⁰ which applies entirely to our case, that the first approximation in the H-P method gives the essential features of the problem, and that all the correction terms are quite small at low temperatures. In this manner, we restrict ourselves to the first approximation of the H-P method in this paper.

⁵ C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951). The authors showed that the magnetic anisotropy eliminates the divergences in the magnetization of a monatomic layer.

⁶ In order to calculate these terms, Döring performed some approximations, some of which were not entirely justified.

⁷ T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

⁸ F. J. Dyson, Phys. Rev. **102**, 1217, 1230 (1956).

⁹ T. Oguchi, Phys. Rev. **117**, 117 (1960). See also, F. Keffer and T. Oguchi, *ibid.* **117**, 718 (1960).

¹⁰ See reference 9, especially p. 122.

¹ C. Kittel, Phys. Rev. **70**, 965 (1946).

² M. J. Klein and R. S. Smith, Phys. Rev. **81**, 378 (1951).

³ S. J. Glass and M. J. Klein, Phys. Rev. **109**, 288 (1958).

⁴ W. Döring, Z. Naturforsch. **16a**, 1008, 1146 (1961).

We shall calculate the dependence of the spontaneous magnetization on the temperature and the number n of monatomic layers of the thin film. Also, the influence of magnetic anisotropy on the magnetic properties of thin films is considered. To this aim, we will introduce in the Hamiltonian a magnetic anisotropy term, which takes into account the possibility of a parallel and a perpendicular magnetic anisotropy. We note that the influence of magnetic anisotropy on the magnetic properties of ferromagnetic thin films has also been studied, by using the trace theory of Heisenberg, in some previous papers of Corciovei,^{11,12} who developed Valenta's method.¹³ Recently, Corciovei and Ghika¹⁴ have passed to the third-order approximation in the trace theory of Heisenberg.

In the present paper, the influence of a small perpendicular anisotropy¹⁵ is found to be unimportant. On the contrary, the parallel magnetic anisotropy has a very interesting influence on the family of curves which give the dependence of spontaneous magnetization on T for various n .

II. THE FIRST-ORDER APPROXIMATION IN THE H-P METHOD

In our study of the ferromagnetism of a thin film, we shall proceed with the H-P method for the spin-wave theory, in contrast to Klein *et al.*^{2,3} and Döring,⁴ who have applied the results of spin-wave theory in Bloch's formulation.

We shall consider that the thin film is a cubic simple lattice, whose unit vectors are \mathbf{i}_x , \mathbf{i}_y , and \mathbf{i}_z , so that \mathbf{i}_z is perpendicular to the surface of the thin film.¹⁶ The thin film can be divided into n monatomic layers parallel to the surface of the thin film, which shall be numbered by ν ($\nu=1, 2, \dots, n$). In this manner, the position of an atom is given by the number of the monatomic layer to which it belongs and by the vector \mathbf{j} in the plane of the layer. Evidently,

$$\mathbf{j} = a(j_y \mathbf{i}_y + j_z \mathbf{i}_z), \quad (1)$$

where a is the lattice constant, j_y and j_z are integers ($j_y, j_z=1, 2, \dots, N$), and N^2 is the number of atoms in a layer which is supposed to be a square. Thus, any atom can be denoted by $\nu \mathbf{j}$.

The Hamiltonian \mathcal{H} will contain an exchange term \mathcal{H}_{ex} , an anisotropy term $\mathcal{H}_{\text{anis}}$, and the Zeeman term \mathcal{H}_Z . We shall denote the spin operator of the $\nu \mathbf{j}$ atom by $\mathbf{S}_{\nu \mathbf{j}}$ and its components in the three directions \mathbf{i}_x , \mathbf{i}_y , and \mathbf{i}_z by $S_{\nu \mathbf{j}x}$, $S_{\nu \mathbf{j}y}$, and $S_{\nu \mathbf{j}z}$, respectively. In the

exchange term, we shall consider only the interactions between nearest neighbor atoms. In this manner,

$$\mathcal{H}_{\text{ex}} = -J \sum_{\langle \nu \mathbf{j}, \nu' \mathbf{j}' \rangle} \mathbf{S}_{\nu \mathbf{j}} \mathbf{S}_{\nu' \mathbf{j}'}, \quad (2)$$

where J is twice the exchange integral corresponding to two nearest neighbors and by $\langle \rangle$ we indicate summation only on the pairs of nearest-neighbor atoms.

We shall assume that the magnetization axis, which is situated in the plane of the film, is the \mathbf{i}_z axis which does not represent a restriction of the problem. We can, thus, suppose that there is a small magnetic anisotropy in the same direction, which we shall call parallel anisotropy. As is known,¹ there exists also a perpendicular magnetic anisotropy corresponding to the \mathbf{i}_x axis.¹⁷ To the parallel and perpendicular magnetic anisotropy will correspond two internal anisotropy magnetic fields, H_{\parallel} and H_{\perp} which are not equal. In a manner similar to that of Döring,⁴ we can now write the anisotropy term of the Hamiltonian in the form

$$\mathcal{H}_{\text{anis}} = -gm_B (H_{\perp} \sum_{\nu \mathbf{j}} S_{\nu \mathbf{j}x} + H_{\parallel} \sum_{\nu \mathbf{j}} S_{\nu \mathbf{j}z}), \quad (3)$$

where g is the gyromagnetic factor, and m_B is the Bohr magneton. As for the Zeeman term, it can be written in the form

$$\mathcal{H}_Z = -gm_B \sum_{\nu \mathbf{j}} (H_x S_{\nu \mathbf{j}x} + H_y S_{\nu \mathbf{j}y} + H_z S_{\nu \mathbf{j}z}), \quad (4)$$

where H_x , H_y , and H_z are the components of the external magnetic field \mathbf{H} in the three directions.

The total Hamiltonian \mathcal{H} will be

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{anis}} + \mathcal{H}_Z. \quad (5)$$

In the H-P theory, the creation and annihilation operators $a_{\nu \mathbf{j}}^*$ and $a_{\nu \mathbf{j}}$, for each atomic spin, which satisfy the following commutations rules are introduced:

$$\begin{aligned} a_{\nu \mathbf{j}} a_{\nu' \mathbf{j}'}^* - a_{\nu' \mathbf{j}'}^* a_{\nu \mathbf{j}} &= \delta_{\nu \nu'} \delta_{\mathbf{j} \mathbf{j}'}, \\ a_{\nu \mathbf{j}} a_{\nu' \mathbf{j}'} - a_{\nu' \mathbf{j}'} a_{\nu \mathbf{j}} &= a_{\nu \mathbf{j}}^* a_{\nu' \mathbf{j}'}^* - a_{\nu' \mathbf{j}'}^* a_{\nu \mathbf{j}} = 0, \end{aligned}$$

and the spin operators are related to these operators by the relations

$$\begin{aligned} S_{\nu \mathbf{j}x} + i S_{\nu \mathbf{j}y} &= (2S)^{1/2} f_{\nu \mathbf{j}}(S) a_{\nu \mathbf{j}}, \\ S_{\nu \mathbf{j}x} - i S_{\nu \mathbf{j}y} &= (2S)^{1/2} a_{\nu \mathbf{j}}^* f_{\nu \mathbf{j}}(S), \\ S_{\nu \mathbf{j}z} &= S - a_{\nu \mathbf{j}}^* a_{\nu \mathbf{j}}, \end{aligned} \quad (7)$$

where

$$f_{\nu \mathbf{j}}(S) = (1 - a_{\nu \mathbf{j}}^* a_{\nu \mathbf{j}} / 2S)^{1/2}. \quad (7')$$

As is known, we can write the series expansion of $f_{\nu \mathbf{j}}(S)$ in powers of $a_{\nu \mathbf{j}}^* a_{\nu \mathbf{j}}$, but we shall not enter into the complexity of this problem.⁹ As mentioned in the introduction,¹⁰ we shall restrict ourselves, in order to obtain the essential features of the problem at low temperatures, to the first approximation of the H-P method. In this approximation, $f_{\nu \mathbf{j}}(S)$ may be replaced

¹⁷ For justification see, also, footnote 16.

¹¹ A. Corciovei, Czech. J. Phys. **10**, 568, 917 (1960).

¹² A. Corciovei, J. Phys. Chem. Solids **20**, 162 (1961).

¹³ L. Valenta, Czech. J. Phys. **7**, 127, 136 (1957).

¹⁴ A. Corciovei and G. Ghika, Czech. J. Phys. **22**, 278 (1962).

¹⁵ We note that in references 12 and 14 the effect of hypothetical very large internal perpendicular magnetic anisotropy fields is studied. Such effects will not be studied in the present paper.

¹⁶ It should be stated that we assume that the cubic lattice is not perfect, but is compressed or dilated along the \mathbf{i}_x direction.

by 1. Performing the calculations, the three terms of the Hamiltonian become

$$\begin{aligned}
 \mathcal{H}_{\text{ex}} = & -JS \sum_{\nu} \sum_{\langle jj' \rangle} (a_{\nu j} a_{\nu j'}^* + a_{\nu j}^* a_{\nu j'}) - JS \sum_{\langle \nu \nu' \rangle} \sum_{\mathbf{j}} (a_{\nu j} a_{\nu' j}^* + a_{\nu j}^* a_{\nu' j}) + JS \sum_{\nu} \sum_{\langle jj' \rangle} (a_{\nu j}^* a_{\nu j} + a_{\nu j'}^* a_{\nu j'}) \\
 & + JS \sum_{\langle \nu \nu' \rangle} \sum_{\mathbf{j}} (a_{\nu j}^* a_{\nu j} + a_{\nu' j}^* a_{\nu' j}) - JS^2 n N^2 z_0 / 2 - JS^2 (n-1) N^2 z_1, \\
 \mathcal{H}_{\text{anis}} = & -g m_B H_1 (S/2)^{1/2} \sum_{\nu} \sum_{\mathbf{j}} (a_{\nu j} + a_{\nu j}^*) - g m_B H_{11} \sum_{\nu} \sum_{\mathbf{j}} (S - a_{\nu j}^* a_{\nu j}), \\
 \mathcal{H}_Z = & -g m_B H_x (S/2)^{1/2} \sum_{\nu} \sum_{\mathbf{j}} (a_{\nu j} + a_{\nu j}^*) + i g m_B H_y (S/2)^{1/2} \sum_{\nu} \sum_{\mathbf{j}} (a_{\nu j} - a_{\nu j}^*) - g m_B H_z \sum_{\nu} \sum_{\mathbf{j}} (S - a_{\nu j}^* a_{\nu j}),
 \end{aligned} \tag{8}$$

where z_0 is the number of nearest neighbors of any atom in the same layer, and z_1 is the number of nearest neighbors of any atom in a given nearest layer of the layer of the atom under consideration. Evidently, in our case $z_0 = 4$ and $z_1 = 1$.

III. CANONICAL TRANSFORMATIONS

We shall now perform some canonical transformations, in order to transform the Hamiltonian into a sum of independent oscillators. Firstly, it is possible to perform a Fourier transformation and to introduce new creation and annihilation operators $a_{\nu \mathbf{h}}^*$ and $a_{\nu \mathbf{h}}$, where \mathbf{h} is the propagation vector within the layer, namely,

$$\mathbf{h} = (2\pi/N a) (m_y \mathbf{i}_y + m_z \mathbf{i}_z) = h_y \mathbf{i}_y + h_z \mathbf{i}_z, \tag{9}$$

where m_y and m_z are integer numbers such that

$$-N/2 < m_y, \quad m_z < N/2.$$

The Fourier transformation is

$$\begin{aligned}
 a_{\nu \mathbf{j}} &= (1/N) \sum_{\mathbf{h}} \exp(-i\mathbf{h}\mathbf{j}) a_{\nu \mathbf{h}}, \\
 a_{\nu \mathbf{j}}^* &= (1/N) \sum_{\mathbf{h}} \exp(i\mathbf{h}\mathbf{j}) a_{\nu \mathbf{h}}^*,
 \end{aligned} \tag{10}$$

and the new operators satisfy the following commutation relations:

$$\begin{aligned}
 a_{\nu \mathbf{h}} a_{\nu' \mathbf{h}'}^* - a_{\nu' \mathbf{h}'}^* a_{\nu \mathbf{h}} &= \delta_{\nu \nu'} \delta_{\mathbf{h} \mathbf{h}'}, \\
 a_{\nu \mathbf{h}} a_{\nu' \mathbf{h}'} - a_{\nu' \mathbf{h}'} a_{\nu \mathbf{h}} &= a_{\nu \mathbf{h}}^* a_{\nu' \mathbf{h}'}^* - a_{\nu' \mathbf{h}'}^* a_{\nu \mathbf{h}} = 0.
 \end{aligned} \tag{11}$$

It is now evident that (10) is also a canonical transformation.

We introduce the notation

$$\gamma_{\mathbf{h}} = (1/z_0) \sum_{\substack{\langle jj' \rangle \\ \mathbf{j} \text{ given}}} \exp[i\mathbf{h}(\mathbf{j} - \mathbf{j}')], \tag{12}$$

where in the sum only the pairs with a fixed \mathbf{j} are considered. It is evident that for cubic simple lattices we have

$$\gamma_{\mathbf{h}} = \frac{1}{2} (\cosh h_y a + \cosh h_z a). \tag{12'}$$

Taking into account the fact that

$$\sum_{\mathbf{j}} \exp(-i\mathbf{h}\mathbf{j}) = N \delta_{0\mathbf{j}}, \tag{13}$$

it is easy to show that

$$\begin{aligned}
 \mathcal{H}_{\text{ex}} = & - (JS z_0 / 2) \sum_{\nu} \sum_{\mathbf{h}} \gamma_{\mathbf{h}} (a_{\nu \mathbf{h}} a_{\nu \mathbf{h}}^* + a_{\nu \mathbf{h}}^* a_{\nu \mathbf{h}}) - JS \sum_{\langle \nu \nu' \rangle} \sum_{\mathbf{h}} (a_{\nu \mathbf{h}} a_{\nu' \mathbf{h}}^* + a_{\nu \mathbf{h}}^* a_{\nu' \mathbf{h}}) + JS \sum_{\nu} (z_0 + z_{1\nu}) \sum_{\mathbf{h}} a_{\nu \mathbf{h}}^* a_{\nu \mathbf{h}} \\
 & - JS^2 n N^2 z_0 / 2 - JS^2 (n-1) N^2 z_1, \\
 \mathcal{H}_{\text{anis}} = & -g m_B H_1 (N^2 S / 2)^{1/2} \sum_{\nu} (a_{\nu 0} + a_{\nu 0}^*) + g m_B H_{11} \sum_{\nu} \sum_{\mathbf{h}} a_{\nu \mathbf{h}}^* a_{\nu \mathbf{h}} - g m_B H_{11} n N^2 S, \\
 \mathcal{H}_Z = & -g m_B H_x (N^2 S / 2)^{1/2} \sum_{\nu} (a_{\nu 0} + a_{\nu 0}^*) - g m_B H_y (N^2 S / 2)^{1/2} \sum_{\nu} (a_{\nu 0} - a_{\nu 0}^*) + g m_B H_z \sum_{\nu} \sum_{\mathbf{h}} a_{\nu \mathbf{h}}^* a_{\nu \mathbf{h}} - g m_B H_z n N^2 S,
 \end{aligned} \tag{14}$$

where

$$\begin{aligned}
 z_{1\nu} &= 2 \quad \text{for } \nu = 2, 3, \dots, n-1, \\
 &= 1 \quad \text{for } \nu = 1, n.
 \end{aligned} \tag{15}$$

In this manner, the sums $\langle \rangle$ have been eliminated. In addition, we introduce the canonical conjugated operators $Q_{\nu \mathbf{h}}$, $P_{\nu \mathbf{h}}$ by the relations

$$a_{\nu \mathbf{h}} = (1/2)^{1/2} (Q_{\nu \mathbf{h}} + iP_{\nu \mathbf{h}}), \quad a_{\nu \mathbf{h}}^* = (1/2)^{1/2} (Q_{\nu \mathbf{h}} - iP_{\nu \mathbf{h}}). \tag{16}$$

We obtain

$$Q_{\nu \mathbf{h}} P_{\nu' \mathbf{h}'} - P_{\nu' \mathbf{h}'} Q_{\nu \mathbf{h}} = i \delta_{\nu \nu'} \delta_{\mathbf{h} \mathbf{h}'}, \tag{17}$$

the other commutators are zero. The three terms of the Hamiltonian can be written

$$\begin{aligned} \mathcal{H}_{\text{ox}} &= -(JSz_0/2) \sum_{\nu} \sum_{\mathbf{h}} \gamma_{\mathbf{h}} (Q_{\nu\mathbf{h}}^2 + P_{\nu\mathbf{h}}^2) - JS \sum_{\langle\nu\nu'\rangle} \sum_{\mathbf{h}} (Q_{\nu\mathbf{h}}Q_{\nu'\mathbf{h}} + P_{\nu\mathbf{h}}P_{\nu'\mathbf{h}}) + (JS/2) \sum_{\nu} (z_0 + z_{1\nu}) \sum_{\mathbf{h}} (Q_{\nu\mathbf{h}}^2 + P_{\nu\mathbf{h}}^2) \\ &\quad - JS(S+1)nN^2(z_0/2) - JS(S+1)(n-1)N^2z_1, \\ \mathcal{H}_{\text{anis}} &= -gm_B H_1 (N^2S)^{1/2} \sum_{\nu} Q_{\nu 0} + (gm_B H_{11}/2) \sum_{\nu} \sum_{\mathbf{h}} (Q_{\nu\mathbf{h}}^2 + P_{\nu\mathbf{h}}^2) - gm_B n N^2 H_{11} (S + \frac{1}{2}), \\ \mathcal{H}_Z &= -gm_B H_x (N^2S)^{1/2} \sum_{\nu} Q_{\nu 0} - gm_B H_y (N^2S)^{1/2} \sum_{\nu} P_{\nu 0} + (gm_B H_z/2) \sum_{\nu} \sum_{\mathbf{h}} (Q_{\nu\mathbf{h}}^2 + P_{\nu\mathbf{h}}^2) - gm_B n N^2 H_z (S + \frac{1}{2}). \end{aligned} \tag{18}$$

We introduce the notations

$$w_0 = -JS(S+1)nN^2(z_0/2) - JS(S+1)(n-1)N^2z_1, \quad \alpha = \frac{1}{2}JS, \quad \alpha_{\mathbf{h}} = -\frac{1}{2}JSz_0\gamma_{\mathbf{h}} + \frac{1}{2}JS(z_0 + 2). \tag{19}$$

In this manner, the total Hamiltonian \mathcal{H} becomes

$$\begin{aligned} \mathcal{H} &= w_0 - gm_B n N^2 (H_x + H_{11}) (S + \frac{1}{2}) + \sum_{\mathbf{h}} \{ (\alpha_{\mathbf{h}} + gm_B H_x/2 + gm_B H_{11}/2) \sum_{\nu} (Q_{\nu\mathbf{h}}^2 + P_{\nu\mathbf{h}}^2) \\ &\quad - \alpha (Q_{1\mathbf{h}}^2 + P_{1\mathbf{h}}^2 + Q_{n\mathbf{h}}^2 + P_{n\mathbf{h}}^2) - 2\alpha \sum_{\langle\nu\nu'\rangle} (Q_{\nu\mathbf{h}}Q_{\nu'\mathbf{h}} + P_{\nu\mathbf{h}}P_{\nu'\mathbf{h}}) \} - gm_B (H_x + H_{11}) (N^2S)^{1/2} \sum_{\nu} Q_{\nu 0} \\ &\quad - gm_B H_y (N^2S)^{1/2} \sum_{\nu} P_{\nu 0}. \end{aligned} \tag{20}$$

In this expression, for every \mathbf{h} the oscillations for various ν are still coupled.

IV. THE COMPLETE DIAGONALIZATION OF THE HAMILTONIAN

With a view to a complete separation of the Hamiltonian in a sum of terms of independent oscillators, we shall perform a new transformation:

$$Q_{\nu\mathbf{h}} = \sum_{\tau=1}^n T^{(\mathbf{h})}_{\nu\tau} \bar{q}_{\tau\mathbf{h}}, \quad P_{\nu\mathbf{h}} = \sum_{\tau=1}^n T^{(\mathbf{h})}_{\nu\tau} \bar{p}_{\tau\mathbf{h}}, \tag{21}$$

for which we impose the conditions

$$\begin{aligned} \sum_{\nu} Q_{\nu\mathbf{h}}^2 &= \sum_{\tau} \bar{q}_{\tau\mathbf{h}}^2, \quad \sum_{\nu} P_{\nu\mathbf{h}}^2 = \sum_{\tau} \bar{p}_{\tau\mathbf{h}}^2, \\ Q_{1\mathbf{h}}^2 + Q_{n\mathbf{h}}^2 + 2 \sum_{\langle\nu\nu'\rangle} Q_{\nu\mathbf{h}}Q_{\nu'\mathbf{h}} &= \sum_{\tau} x^{(\mathbf{h})}_{\tau} \bar{q}_{\tau\mathbf{h}}^2, \\ P_{1\mathbf{h}}^2 + P_{n\mathbf{h}}^2 + 2 \sum_{\langle\nu\nu'\rangle} P_{\nu\mathbf{h}}P_{\nu'\mathbf{h}} &= \sum_{\tau} x^{(\mathbf{h})}_{\tau} \bar{p}_{\tau\mathbf{h}}^2. \end{aligned} \tag{22}$$

These conditions can be transformed into

$$\sum_{\nu} T^{(\mathbf{h})}_{\nu\tau} T^{(\mathbf{h})}_{\nu\tau'} = \delta_{\tau\tau'}, \tag{23}$$

and

$$\begin{aligned} (-x^{(\mathbf{h})}_{\tau} + 1)T^{(\mathbf{h})}_{1\tau} + T^{(\mathbf{h})}_{2\tau} &= 0, \\ -x^{(\mathbf{h})}_{\tau} T^{(\mathbf{h})}_{\nu\tau} + (T^{(\mathbf{h})}_{\nu+1,\tau} + T^{(\mathbf{h})}_{\nu-1,\tau}) &= 0, \\ &\text{for } \nu = 2, 3, \dots, n-1, \\ (-x^{(\mathbf{h})}_{\tau} + 1)T^{(\mathbf{h})}_{n\tau} + T^{(\mathbf{h})}_{n-1,\tau} &= 0. \end{aligned} \tag{24}$$

Using (23) it is easy to verify that $\bar{q}_{\tau\mathbf{h}}, \bar{p}_{\tau'\mathbf{h}}$ satisfy the rule

$$\bar{q}_{\tau\mathbf{h}}\bar{p}_{\tau'\mathbf{h}} - \bar{p}_{\tau'\mathbf{h}}\bar{q}_{\tau\mathbf{h}} = i\delta_{\tau\tau'}\delta_{\mathbf{h}\mathbf{h}'}. \tag{25}$$

One sees that the solutions (24) are independent of \mathbf{h} . We can, thus, omit the superscript (\mathbf{h}) in $T^{(\mathbf{h})}_{\nu\tau}$ and $x^{(\mathbf{h})}_{\tau}$ and write simply $T_{\nu\tau}$ and x_{τ} .

The solution $T_{\nu\tau}$ of (24) must not vanish; it is, thus, necessary to impose the condition

$$\begin{vmatrix} 1-x & 1 & 0 & \dots & 0 \\ 1 & -x & 1 & \dots & 0 \\ 0 & 1 & -x & \dots & 0 \\ & & \dots & \dots & \\ 0 & \dots & -x & 1 & 0 \\ 0 & \dots & 1 & -x & 1 \\ 0 & \dots & 0 & 1 & 1-x \end{vmatrix} = 0. \tag{25}$$

It is evident that the problem discussed above occurs for $n \geq 2$. In the case $n=1$, the canonical transformation is no longer necessary, and it can be seen formally that $x=2$.

The solution of (25) is simple when n is small. For example, one obtains for $n=2: x_1=2, x_2=0$; for $n=3: x_1=2, x_2=1, x_3=-1$; and for $n=4: x_1=2, x_2=\sqrt{2}, x_3=0, x_4=-\sqrt{2}$. It is possible to show¹⁸ that for a given n the solutions x_{τ} can be written

$$x_{\tau} = 2 \cos[(\pi/n)(\tau-1)], \quad \tau = 1, 2, \dots, n, \tag{26}$$

because the secular problem is very similar to that which appears in the vibration of a linear chain of atoms. It is to be noted that condition (26) corresponds to the introduction of a cyclic condition in the \mathbf{i}_x direction of a thin film with $2n$ layers instead of n .

¹⁸ C. Hemmer, thesis, Nordita Publication 27, 1959 (unpublished). See also reference 4, p. 1148.

Performing the transformation (21), the Hamiltonian corresponding to n monatomic layers takes the form

$$\mathcal{H} = w_0 - gm_B n N^2 (H_x + H_{11}) (S + \frac{1}{2}) + \sum_{\mathbf{h}} \sum_{\tau} \{ (\alpha_{\mathbf{h}} + gm_B H_x / 2 + gm_B H_{11} / 2) (\bar{q}_{\tau \mathbf{h}}^2 + \bar{p}_{\tau \mathbf{h}}^2) \}$$

$$- \alpha \sum_{\mathbf{h}} \sum_{\tau} x_{\tau} (\bar{q}_{\tau \mathbf{h}}^2 + \bar{p}_{\tau \mathbf{h}}^2) - gm_B (H_x + H_{11}) (N^2 S)^{1/2} \sum_{\tau} (\sum_{\nu} T_{\nu \tau}) \bar{q}_{\tau 0} - gm_B H_y (N^2 S)^{1/2} \sum_{\tau} (\sum_{\nu} T_{\nu \tau}) \bar{p}_{\tau 0}. \quad (27)$$

Finally, we perform the transformation

$$\bar{q}_{\tau 0} = q_{\tau 0} + [gm_B (H_x + H_{11}) (N^2 S)^{1/2} \sum_{\nu} T_{\nu \tau}] / [2\alpha_0 + gm_B (H_x + H_{11}) - 2\alpha x_{\tau}],$$

$$\bar{p}_{\tau 0} = p_{\tau 0} + [gm_B H_y (N^2 S)^{1/2} \sum_{\nu} T_{\nu \tau}] / [2\alpha_0 + gm_B (H_x + H_{11}) - 2\alpha x_{\tau}], \quad (28)$$

$$\bar{q}_{\tau \mathbf{h}} = q_{\tau \mathbf{h}}, \quad \bar{p}_{\tau \mathbf{h}} = p_{\tau \mathbf{h}}, \quad \text{for } \mathbf{h} \neq 0,$$

where α_0 is $\alpha_{\mathbf{h}=0}$. The transformation is evidently canonical and the new operators satisfy the commutation rules

$$q_{\tau \mathbf{h}} p_{\tau' \mathbf{h}'} - p_{\tau' \mathbf{h}'} q_{\tau \mathbf{h}} = i \delta_{\tau \tau'} \delta_{\mathbf{h} \mathbf{h}'}$$

With the transformation (28), the Hamiltonian becomes a sum of oscillators:

$$\mathcal{H} = w_0 + \sum_{\mathbf{h}} \sum_{\tau} [(\alpha_{\mathbf{h}} + gm_B H_x / 2 + gm_B H_{11} / 2 - \alpha x_{\tau})$$

$$\times (q_{\tau \mathbf{h}}^2 + p_{\tau \mathbf{h}}^2)] + w_x (H_x + H_{11})^2 + w_y H_y^2$$

$$+ w_z (H_x + H_{11}), \quad (29)$$

where we have introduced the notations

$$w_x = - \sum_{\tau} \frac{g^2 m_B^2 N^2 S (\sum_{\nu} T_{\nu \tau})^2}{4(\alpha_0 + gm_B H_x / 2 + gm_B H_{11} / 2 - \alpha x_{\tau})},$$

$$w_y = - \sum_{\tau} \frac{g^2 m_B^2 N^2 S (\sum_{\nu} T_{\nu \tau})^2}{4(\alpha_0 + gm_B H_x / 2 + gm_B H_{11} / 2 - \alpha x_{\tau})}, \quad (30)$$

$$w_z = - gm_B n N^2 (S + \frac{1}{2}).$$

On the other hand, the quantization can be performed according to the rule

$$Aq^2 + Bp^2 \rightarrow (2n+1)(AB)^{1/2},$$

where n is an integer quantum number.

In this manner, the Hamiltonian will have the following eigenvalues:

$$E_{\mathbf{n}} = w_0 + w_x (H_x + H_{11})^2 + w_y H_y^2 + w_z (H_x + H_{11})$$

$$+ \sum_{\tau} \sum_{\mathbf{h}} w_{\tau \mathbf{h}} (2n_{\tau \mathbf{h}} + 1), \quad (31)$$

where

$$w_{\tau \mathbf{h}} = \alpha_{\mathbf{h}} + gm_B (H_x + H_{11}) / 2 - \alpha x_{\tau}, \quad (32)$$

and $n_{\tau \mathbf{h}}$ are the (positive or zero) corresponding quantum numbers. By \mathbf{n} we have designated the set of all $n_{\tau \mathbf{h}}$.

Now we can calculate the partition function Z . We have

$$Z = \sum_{\mathbf{n}} \exp[-E_{\mathbf{n}} / (kT)],$$

where k is Boltzmann's constant and T is the temperature. It is easy to show that we obtain

$$Z = \exp\left(-\frac{w_0 + w_x (H_x + H_{11})^2 + w_y H_y^2 + w_z (H_x + H_{11})}{kT}\right)$$

$$\times \prod_{\tau} \prod_{\mathbf{h}} \frac{e^{-w_{\tau \mathbf{h}} / (kT)}}{1 - e^{-2w_{\tau \mathbf{h}} / (kT)}}. \quad (33)$$

With the aid of Z , we can calculate the free energy $F = -kT \ln Z$ in the form

$$F = w_0 + w_x (H_x + H_{11})^2 + w_y H_y^2 + w_z (H_x + H_{11})$$

$$+ \sum_{\tau} \sum_{\mathbf{h}} w_{\tau \mathbf{h}} + kT \sum_{\tau} \sum_{\mathbf{h}} \ln(1 - e^{-2w_{\tau \mathbf{h}} / (kT)}). \quad (34)$$

V. THE MAGNETIZATION OF THIN FILM

The components of magnetization on various axes are given by

$$M_x = -\partial F / \partial H_x, \quad M_y = -\partial F / \partial H_y,$$

$$M_z = -\partial F / \partial H_z. \quad (35)$$

One obtains

$$M_x = -2w_x (H_x + H_{11}), \quad M_y = -2w_y H_y, \quad (36)$$

and

$$M_z = -w_z - \sum_{\tau} \sum_{\mathbf{h}} \left[\left(1 + \frac{2e^{-2w_{\tau \mathbf{h}} / (kT)}}{1 - e^{-2w_{\tau \mathbf{h}} / (kT)}} \right) \frac{\partial w_{\tau \mathbf{h}}}{\partial H_z} \right]. \quad (37)$$

When \mathbf{H} vanishes we obtain the spontaneous magnetization. It is seen that the spontaneous magnetization on the Oy axis vanishes. Also, since the order of magnitude of H_1 is 10 Oe to 100 Oe, we have $w_x H_1 \ll w_z$ and, thus, the spontaneous magnetization perpendicular to the thin film can be neglected as compared to the spontaneous magnetization on the Oz axis. Further, we shall consider only the spontaneous magnetization in the plane of the thin film on the Oz axis.

In formula (37) we can go from the sum to an integral over $dh_y dh_z$ and, for the spontaneous magneti-

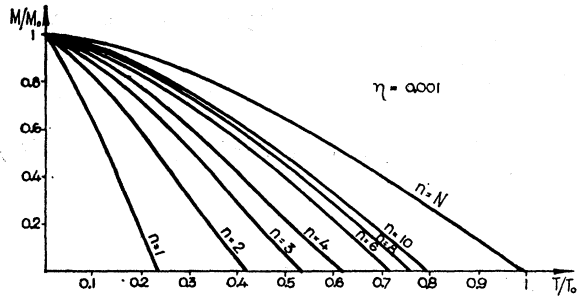


FIG. 1. Magnetization vs temperature for $\eta=0.001$ and for different values of the thickness.

zation, we obtain

$$M_z = gm_B n N^2 (S + \frac{1}{2}) - \sum_{\tau} \frac{N^2 a^2}{(2\pi)^2} \left[\int_{-\pi/a}^{\pi/a} \int_{-\pi/a}^{\pi/a} \left(1 + \frac{2e^{-2w_{\tau h}} h'(kT)}{1 - e^{-2w_{\tau h}} h'(kT)} \right) \times \frac{\partial w_{\tau h}}{\partial H_z} dh_y dh_z \right]_{H_z=0}, \quad (38)$$

where we have also introduced the normalization factor $(Na/2\pi)^2$. The main contribution to (38) arises near the point $h_y = h_z = 0$, as can be seen from (32) and (19). We can, thus, approximate γ_h which appears in α_h by

$$\gamma_h = 1 - \frac{1}{4} [(h_y a)^2 + (h_z a)^2], \quad (36)$$

and the integration can be performed by using polar coordinates

$$h_y = (\sigma/a) \cos\theta, \quad h_z = (\sigma/a) \sin\theta \quad (40)$$

and replacing the square domain of integration with a circle of equal surface. In this manner, we obtain

$$M_z = gm_B n N^2 (S + \frac{1}{2}) - \left(\frac{N}{2\pi} \right)^2 \sum_{\tau} \left[\int_0^{2\sqrt{\pi}} \int_0^{2\pi} \left(1 + \frac{2e^{-2w_{\tau}(\sigma)} h'(kT)}{1 - e^{-2w_{\tau}(\sigma)} h'(kT)} \right) \times \frac{\partial w_{\tau}(\sigma)}{\partial H_z} \sigma d\sigma d\theta \right]_{H_z=0}, \quad (41)$$

where $w_{\tau}(\sigma)$ is obtained from (32) and (19) and has the expression

$$w_{\tau}(\sigma) = JS\sigma^2/2 - JSx_{\tau}/2 + JS + gm_B H_z/2 + gm_B H_{11}/2. \quad (42)$$

By performing the integration, it is easy to show that

$$M_z = M_0 \left\{ 1 - \frac{\varphi}{4\pi n S} \times \sum_{\tau=1}^n \ln \frac{1 - \exp[-(2/\varphi)(2\pi + 1 - x_{\tau}/2 + \eta)]}{1 - \exp[-(2/\varphi)(1 - x_{\tau}/2 + \eta)]} \right\}, \quad (43)$$

where we have introduced the notations

$$M_0 = gm_B S n N^2, \quad \varphi = kT/(JS), \quad (44)$$

$$\eta = gm_B H_{11}/(2JS).$$

We note that η is the ratio between the "parallel magnetic anisotropy constant" $gm_B H_{11}$ and the "exchange constant" $2SJ$. In general, we have $\eta \ll 1$. Expression (43) gives us the general dependence of the magnetization on the temperature, the thickness, the parallel magnetic anisotropy, the exchange integral, and the spin.

For $\tau=1$, we have $1 - x_{\tau}/2 = 0$. In this manner, it is evident that the quantity η eliminates in (43) the divergency at $\tau=1$. The unnatural procedure of Döring⁴ of eliminating the divergency without introducing the magnetic anisotropy leads to some difficulties which cannot be overcome in a consistent manner. In our opinion, only by taking into account the parallel magnetic anisotropy, can we study the problem in a consistent manner. As we have seen, the perpendicular magnetic anisotropy introduces a component of the magnetization on the Ox axis which, of course, for the usual small values of H_{11} , can be neglected as compared with M_z . In our approximation, the perpendicular anisotropy does not influence the magnetization on the Oz axis.

Formula (43) shows that for small values of n , M_z decreases linearly with T . When n becomes very large, we must obtain the well-known $T^{3/2}$ decrease of the magnetization which corresponds to the bulk material. Let us demonstrate this fact. For simplicity we shall put $\eta=0$ and we shall neglect the quantities

$$\exp[-(2/\varphi)(2\pi + 1 - x_{\tau}/2 + \eta)],$$

which are practically vanishing in (43) at low temperatures. Passing from sum to integral and putting $\tau-1=y$, we obtain

$$M_z = M_0 \left[1 - \frac{\varphi}{4\pi n S} \int_0^n \ln \frac{1}{1 - e^{-\pi^2 y^2 / (n^2 \varphi)}} dy \right],$$

where we have also performed a series development in the vicinity of $y=0$, where the main contribution

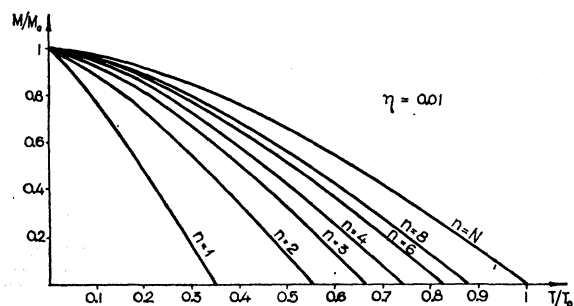


FIG. 2. Magnetization vs temperature for $\eta=0.01$ and for different values of the thickness.

of the integral appears. Further, we introduce $u = \pi y / (n\sqrt{\varphi})$ and obtain

$$M_z = M_0 \left[1 - \frac{\varphi^{3/2}}{4\pi^2 S} \int_0^\infty \ln \frac{1}{1 - e^{-u^2}} du \right],$$

where we have approximated the upper limit of the integral with infinity at low temperatures.

We observe that

$$\int_0^\infty \ln[1/(1 - e^{-u^2})] du = (\pi^{1/2}/2)\zeta(3/2),$$

where ζ is Riemann's function. As is known, $\zeta(3/2) = 2.612$. In this way, we obtain finally

$$\begin{aligned} M_z &= M_0 \left[1 - 2.612\pi^{1/2} \varphi^{3/2} / (8\pi^2 S) \right] \\ &= M_0 \left[1 - \frac{0.117}{2S} \varphi^{3/2} \right], \quad (45) \end{aligned}$$

which is exactly the result known for a ferromagnetic cubic solid body.¹⁹ We can introduce the quantity T_0 defined from the condition

$$0.117 \times (kT_0)^{3/2} / (2J^{3/2} S^{5/2}) = 1.$$

In this manner, (45) becomes

$$M_z = M_0 \left[1 - (T/T_0)^{3/2} \right]. \quad (46)$$

For $T = T_0$, M_z vanishes and, thus, in this theory, T_0 can be considered the Curie temperature of the bulk material.

The transition from the linear decrease with T of the magnetization for small n to the $T^{3/2}$ decrease for large values of n can be seen in Figs. 1 and 2. Figure 1 shows the dependence of the magnetization on T for small values of n and for $S = \frac{1}{2}$ and $\eta = 0.001$. Figure 2 shows the same family of curves, but for $\eta = 0.01$. This value of η is larger than the experimental values of η for the solid body. In Figs. 1 and 2 we have also shown [from Eq. (46)] the dependence of the magnetization on T for the bulk material (n of the order of magnitude of N) and for $\eta = 0$. We observe that the temperature

¹⁹ See, for example, R. E. Peierls, *Quantum Theory of Solids* (Clarendon Press, Oxford, 1955), p. 172, formula (42). For comparison with Peierls we observe that in (45), J is the exchange energy and $2S$ is the number of $\frac{1}{2}$ spins of any atom. We note that the correct numerical constant has been obtained.

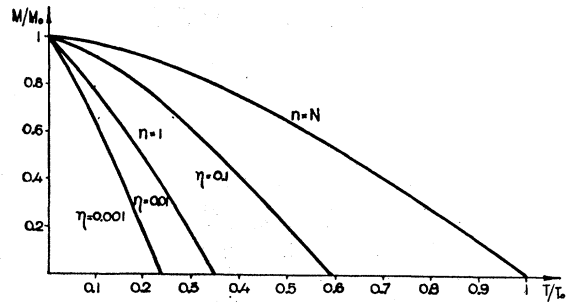


FIG. 3. Magnetization vs temperature for $n=1$, and for different values of the coefficient η .

where the magnetization vanishes is the Curie temperature, which increases with increasing n and η . However, as is known, in the H-P method, the results are only qualitative in this range of temperatures. In Fig. 3, the dependence of the magnetization on T for $n=1$ and $\eta=0.1, 0.01$, and 0.001 , respectively, is given.

It is very interesting to note the dependence on η of the family of curves given in Figs. 1-3. For a given temperature and a given n , the magnetization increases with increasing η . It is very possible that η depends on n , i.e., that it increases when n decreases.²⁰ On the other hand, it is well known that for n smaller than 10, the experimental results are very unsatisfactory, different authors giving different results for the same material.²¹ In particular, Neugebauer²² obtains larger values for the magnetization than other authors. In this manner, the results of Neugebauer seem to indicate a greater η for very thin films than for the solid body. For example, it can be seen in Fig. 2 that for $\eta=0.01$ for n greater than 10, the dependence and the values of the magnetization become practically the same as for the solid body.

It is, of course, evident that accurate experimental results will permit the calculation of the corresponding η and, thus, the magnetic anisotropy for various n , and will also clarify other problems which arise in the study of very thin ferromagnetic film.

²⁰ It is not the purpose of this paper to explain such a dependence.

²¹ See, for example, C. P. Bean, in *Proceedings of an International Conference on Structures and Properties on Thin Films, Bolton Landing, New York, 1959* (John Wiley & Sons, Inc., New York, 1959), p. 331.

²² C. A. Neugebauer, *Phys. Rev.* **116**, 1441 (1959).