angular distributions of neutrons with energies greater than say 90 percent of the incident proton energy should be due principally to the target nucleon momentum distribution, and that consequently such measurements would yield considerable information concerning momentum distributions in nuclei.

The authors wish to express their appreciation to Professor Edwin M. McMillan for his interest and advice, to Dr. Geoffrey Chew and Mr. Lester Baumhoff for valuable discussions on the theoretical aspects of the experiment, and to Mr. James Vale and the cyclotron operating crew for their cooperation and assistance.

PHYSICAL REVIEW VOLUME 81, NUMBER 3 FEBRUARY 1, 1951

Thin Ferromagnetic Films*

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(Received June 15, 1950)

The magnetic properties of thin films of ferromagnetic materials have been studied by means of the Bloch spin-wave theory. The spontaneous magnetization depends upon the number of atomic layers, G_z, in the film. For thicknesses below a "critical" thickness, which depends on the temperature and film dimensions, the spontaneous magnetization decreases rapidly as the number of atomic layers is decreased. The temperature dependence of the spontaneous magnetization varies from a $T^{\frac{3}{2}}$ law for very thick films to a linear function of T for a monolayer film. The spontaneous magnetization, at fixed temperature and film thickness, decreases as the film dimension increases.

I. INTRODUCTION

IT has been shown by Bloch[1] that one- and two-dimensional lattices, in contrast to three-dimensional lattices, should not exhibit spontaneous magnetization, even when the exchange integral is positive. This conclusion was reached by the use of the spin-wave method, which was introduced by Bloch as an approximate way of treating the Heisenberg[2] model of a ferromagnet. The spin-wave method provides a reasonable approximation to the lowest energy states of the system, as shown by Bethe,[3] but only for these; and conclusions reached by this method are therefore expected to be valid only at low temperatures when the magnetization is near its saturation value. It should be mentioned that Bloch's result on the absence of spontaneous magnetization in two-dimensional lattices has been substantiated by the calculations of Weiss[4] using the Bethe-Peierls method;[5] on the other hand, recent work by Ekstein[6] contradicts this result, leaving this point in doubt.

If one accepts Bloch's result, it follows that the magnetic properties of a slab of ferromagnetic material should vary with the thickness of the slab and should show a transition from ferromagnetic behavior for thick slabs to paramagnetic behavior for sufficiently thin films. We have studied the dependence of the spontaneous magnetization on the thickness of the sample, using the method of spin-waves, in order to determine the nature of the transitional behavior to be expected. An experimental investigation of the magnetic properties of thin films would clearly be desirable, since it could serve as a check on the correctness of conclusions drawn from the spin-wave theory.

II. CALCULATIONS AND RESULTS

On the basis of the usual approximations of the spin-wave method one obtains, in the case of a simple cubic lattice of atoms having spin $\frac{1}{2}$, the spontaneous magnetization:[7]

$$M=\beta N\left[1-\frac{2}{N}\sum_{\lambda_x,\lambda_y,\lambda_z}\left\{\exp\left[\frac{2J}{kT}\sum_{i=x,y,z}\times\left(1-\cos\frac{2\pi\lambda_i}{G_i}\right)\right]-1\right\}^{-1}\right]. \quad (1)$$

Here $2\pi\lambda_x/G_x$ is the x component of the spin-wave vector: G_x is the x dimension of the crystal in units of the lattice parameter and λ_x is an integer ($\lambda_x=0, 1, 2, \cdots G_x-1$). β is the Bohr magneton, J is the exchange integral between nearest neighbors, and N is the total number of atoms in the crystal ($N=G_xG_yG_z$).

If G_x, G_y, G_z are large numbers, then the variables $K_\lambda^{(x)}=2\pi\lambda_x/G_x$, etc., change by small steps and one can approximate the sums by integrals. Thus, for a three-

* This work was supported in part by the ONR.
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dimensional crystal the standard treatment[7] gives

$$M^{(3\text{ dim})}=\beta N\left[1-\frac{2(4\pi)}{(2\pi)^3}\int_0^\infty \frac{K^2dK}{\exp(JK^2/kT)-1}\right]$$
$$=\beta N[1-0.13(kT/J)^{\frac{3}{2}}]. \quad (2)$$

(The cosines have been expanded to first order.) This is the Bloch $T^{\frac{3}{2}}$ formula for the spontaneous magnetization. Similarly, for a two-dimensional lattice:

$$M^{(2\text{ dim})}=\beta N\left[1-\frac{2(2\pi)}{(2\pi)^2}\int_0^\infty \frac{KdK}{\exp(JK^2/kT)-1}\right]. \quad (3)$$

This integral diverges at the lower limit, a result which is interpreted to mean that the plane lattice has no spontaneous magnetization. For this case, the basic assumption of the Bloch theory, that the magnetization is close to the saturation value, is invalid.

In the case of a thin film (thin in the z direction) G_x and G_y are large numbers, taken equal to G for convenience; and, therefore, one can integrate over λ_x and λ_y but not over λ_z. One obtains

$$M=\beta N\left\{1-\frac{2G^2(2\pi)}{N(2\pi)^2}\sum_{\lambda_z=0}^{G_z-1}\int_{2\pi/G}^\infty \times\frac{KdK}{\exp(J/kT)[K^2+2-2\cos(2\pi\lambda_z/G_z)]-1}\right\}. \quad (4)$$

The lower limit on the integral is not zero, because in the original sum of Eq. (1) the states with $\lambda_x=\lambda_y=\lambda_z=0$ have been excluded. The reasons for making this omission will be discussed in the Appendix. The upper limit is set equal to infinity without appreciable error because the important contributions to the integral come from the region of small K when $J/kT\gg 1$, (a condition necessary for the applicability of the Bloch method).

This integral can now be evaluated with the result that

$$M=\beta N\left[1-\frac{kT}{J}\frac{1}{2\pi G_z}\sum_{\lambda_z=0}^{G_z-1}(-)\times\ln\left\{1-\left(1-\frac{J}{kT}\frac{4\pi^2}{G^2}\right)\exp[-f(\lambda_z)]\right\}\right], \quad (5)$$

where

$$f(\lambda_z)=(2J/kT)[1-\cos(2\pi\lambda_z/G_z)]. \quad (6)$$

Only the first-order term in $\exp[(J/kT)\cdot(4\pi^2/G^2)]$ has been retained.

We have carried out the summation in Eq. (5) by numerical methods for films varying in thickness from 1 to 128 layers of atoms. Representative results are plotted in Fig. 1, which shows the relative magnetization M/M_0 as a function of the reduced temperature T/T_B for different values of G_z, the thickness expressed in atomic layers. Here, $M_0=\beta N$, the magnetic moment for complete saturation; and T_B is the characteristic temperature determined from the Bloch $T^{\frac{3}{2}}$ law Eq. (2); i.e.,

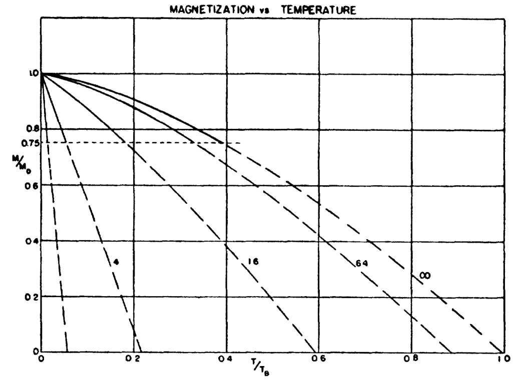

FIG. 1. The relative magnetization M/M_0 as function of the reduced temperature T/T_B for films of different thickness. The integers on the curves represent the number of layers of atoms. The film is square, 2×10^7 atoms on a side. The dashed line at $M/M_0=0.75$ indicates that the predictions of the theory are not expected to be valid below this limit. The value 0.75 is arbitrary.

$$T_B^{\frac{3}{2}}=(1/0.13)(J/k)^{\frac{3}{2}};\quad T_B=3.9J/k. \quad (7)$$

The curve for $G_z=\infty$ is a plot of Eq. (2). G, the linear dimension of the film in units of the lattice parameter has been taken as 2×10^7 in Fig. 1.

It will be noted that the curves have been drawn dashed for magnetizations less than $0.75M_0$. This is to stress the point that valid conclusions cannot be drawn from the spin-wave theory unless the magnetization[8] is near its saturation value M_0.

Perhaps the most important conclusion to be drawn from Fig. 1 is that, for the assumed values of the parameters, significant deviations from the three-dimensional spontaneous magnetization occur for films less than about 60 atomic layers in thickness. At any given temperature the spontaneous magnetization decreases rapidly with decreasing film thickness below this "critical" number of atomic layers. Furthermore, for these sufficiently thin films the spontaneous magnetization falls off more sharply with increasing temperature than $T^{\frac{3}{2}}$, approaching a linear function of temperature.[9]

It should be noted that we include a curve for $G_z=1$, i.e., the two-dimensional lattice. This curve has been calculated from Eq. (4) rather than from Eq. (3). The

[8] For example, the curve for $G_z=\infty$, the $T^{\frac{3}{2}}$ law does not correspond to experimental curves except at low temperatures, below about $\frac{1}{3}$ the Curie temperature. See the curves in R. H. Fowler, *Statistical Mechanics* (The Macmillan Company, New York, 1936), pp. 500–501.

[9] The curve drawn for a monolayer film is strictly of the form $M/M_0=1-aT$. This is incompatible with Nernst's theorem which requires that $(\partial M/\partial T)_{T=0}=0$. The reason for the discrepancy is that the integration over λ_x and λ_y is not permitted at very low temperatures ($kT\leqq 2\pi J/G$) and the magnetization should then be calculated directly from Eq. (1). The authors would like to thank Dr. Charles Kittel for raising this question.

reason for the lack of divergence in Eq. (4) is, of course, that the lower limit on the integral is not zero, because states with $\lambda_x=\lambda_y=\lambda_z=0$ have been omitted. (See Appendix.) The curve implies that at sufficiently low temperatures ($T<0.2J/k$), even a monolayer film will be ferromagnetic. (A result analogous to this has been found by Osborne[10] in a study of the two-dimensional ideal Bose-Einstein gas, a closely related problem.)

In addition to its dependence on T and G_z shown in Fig. 1 and discussed above, the spontaneous magnetization depends on G, the linear dimension of the film expressed in units of the lattice parameter. When conditions are such that all terms beyond the first term of the sum in Eq. (5) can be neglected, this dependence is very simple, and we find:

$$\partial(M/M_0)/\partial \ln G = -(kT/J)(1/\pi G_z). \qquad (8)$$

The condition for the validity of Eq. (8) is that the excitation energy of a state with nonzero z-wave number $2\pi\lambda_z/G_z$ be large compared to kT; i.e., $(J/kT)4\pi^2/G_z^2)\gg 1$. When this condition is satisfied, M/M_0 is a linear function of $\ln G$: M/M_0 decreases as $\ln G$ increases, T and G_z (film thickness) held constant.

When Eq. (8) is not valid, one can still obtain some information on the dependence of M/M_0 on G; namely, by considering the full sum in Eq. (5), it can be shown that for $G>G_z^2$, $\partial(M/M_0)/\partial \ln G<0$, implying that for sufficiently large G, M/M_0 goes to zero. (The bound $G>G_z^2$ is merely a convenient one in estimating the partial derivative since the functional dependence is quite complicated.)

III. DISCUSSION

Let us now give a qualitative physical discussion which may help to clarify the results which have been derived above. Consider the energy levels of the system as given by the spin-wave theory. They have the form:[7]

$$E(n_{\lambda})=C+J\sum_{\lambda_x,\lambda_y,\lambda_z}\left[\sum_{i=x,y,z}\left(1-\cos\frac{2\pi\lambda_i}{G_i}\right)\right]n_{\lambda}, \qquad (9)$$

where C is a constant and n_{λ} is an integer which gives the number of spin-wave quanta in the state characterized by λ_x, λ_y, λ_z.

When $G_z\ll G(=G_x=G_y)$, these energy levels correspond to a spectrum in which the spacing of states differing in their λ_x- or λ_y-values but having the same value of λ_z is very small compared to the spacing of states which differ only in their λ_z-values. In particular there are G^2 states with $\lambda_z=0$ (which may be called two-dimensional states); and, since for $\lambda_z\neq 0$ the lowest energy level is ($\lambda_x=\lambda_y=0$, $\lambda_z=1$), for which the excitation energy is $\cong J(2\pi^2/G_z^2)$, it is easy to show that there are $\cong\pi(G/G_z)^2$ two-dimensional states having energies lower than the first state with $\lambda_z\neq 0$. (Similarly, between the states ($\lambda_x=\lambda_y=0$, $\lambda_z=1$) and ($\lambda_x=\lambda_y=0$, $\lambda_z=2$) there are approximately $3\pi(G/G_z)^2$ states, etc.)

These remarks indicate, therefore, that when G_z is sufficiently small compared to G, i.e., when the film thickness is sufficiently small compared to its linear dimension, only the states determined by the first few values of λ_z make any important contribution in the relevant low temperature region. In the calculations given above this means that only the first few terms of the sum in Eq. (5) are appreciable. As a consequence, when $G_z/G\ll 1$, the magnetic properties are essentially determined by the two-dimensional states, giving rise to the results discussed quantitatively in Sec. II above.

The authors would like to thank the referee for several valuable suggestions.

APPENDIX

We shall now discuss the omission of the states with $\lambda_x=\lambda_y=\lambda_z=0$. These states of zero spin-wave vector have a simple classical interpretation: they correspond to all spins being parallel with the spin system aligned in an arbitrary direction. In the absence of an external magnetic field, these states have no excitation energy, as may be verified from Eq. (9). The contribution of these states to the magnetization is infinite as can be seen from Eq. (1).

The origin of this infinite term in the magnetization can be traced to the following circumstance. The sum of the n_{λ} (the occupation numbers of the spin-wave states) is a measure of the number of reversed spins in the lattice. Consequently, this sum has a maximum value. In carrying out the partition sum (from which Eq. (1) is derived) one should therefore impose a restriction on the possible values of the n_{λ}; namely, their sum should be less than a fixed number, proportional to N. Actually the partition sum is evaluated without imposing this restriction: the n_{λ} are allowed to take on all integer values from zero to infinity. This (incorrect) method of evaluating the partition sum leads directly to the infinity in question.

We have not been able to perform the calculation of the partition sum rigorously, but we have made another type of approximate calculation. We have restricted the occupation numbers n_{λ} by setting their sum equal to one fixed value determined to fit experimental data for the spontaneous magnetization. While this method is only an *a posteriori* one, the result is of interest: the troublesome states, with $\lambda_x=\lambda_y=\lambda_z=0$, make a completely negligible contribution to the magnetization, rather then the infinite one obtained by the usual method.

The result of this approximate calculation and the physical unreality of the infinite term in question have been our justification for omitting the zero wave number states from our treatment. A rigorous calculation would, of course, settle the subject conclusively.

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