SOVIET PHYSICS JETP VOLUME 12, NUMBER 5 MAY, 1961

CONTRIBUTION TO THE THEORY OF FERRO- AND ANTIFERROMAGNETISM

V. L. GINZBURG and V. M. FAĬN

Radiophysics Institute, Gor'kiĭ State University

Submitted to JETP editor May 26, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) **39**, 1323-1338 (November, 1960).

A simple approximate method is developed for the determination of the magnetization of the lattice or sublattice and of other quantities in ferro- and antiferromagnetic materials. The method is applicable over the entire range of temperatures, and takes into account the dependence on the dimensions and form of the magnetic system. The question is discussed of the antiferromagnetism and ferromagnetism of small particles and polymer chains, an aggregate of which may be considered, under given conditions, to form a "paramagnetic fluid."

ONE of the highly interesting current problems is the study of ferromagnetism, and especially antiferromagnetism, of finely-disperse substances, polymers, and possibly, individual macromolecules. (We are concerned here also with the paramagnetic region, i.e., with temperatures above the transition temperature.) It is clear even from general considerations that magnetic methods can be highly effective in the study of the cited objects. Hence we shall limit ourselves here only to referring to the experimental data that point to the anomalous magnetic properties of certain nucleic acids and artificial polymers.[1] The nature of these effects has not yet been elucidated, but even if it is not related to antiferromagnetism, as we assume,[2] an analysis of the properties of ferro- and antiferromagnetic materials of the "polymeric type" will nevertheless be necessary. For this purpose, we must have a simple method for the approximate calculation of the magnetization of the lattice or sublattice and of other quantities, as functions of the dimensions and form of the specimens (small particles, films, polymer chains, etc.) over as wide a temperature range as possible.

Such a method is developed below, based on a certain self-consistent generalization of the theory of spin waves. The results obtained may also be of interest in the case of bulk ferro- and antiferromagnetic materials, since the expressions derived are approximately correct for all temperatures. Here we have based our treatment on the ordinary model of localized spins with exchange interaction (see, e. g., reference 3), which in itself deviates greatly from reality. However, the results thus obtained have, to a considerable degree, a general character. That is, they are independent of the model within broad limits, and may be derived in a semi-phenomenological manner.[4-6] It would be somewhat more consistent to develop the theory directly by the latter method, but both approaches are permissible in the final analysis. However, we found it more convenient to proceed from the model representation. (Besides, while in the semi-phenomenological approach one commonly limits the treatment to the terms which are quadratic in the components of the wave vector, we have used more general expressions.)

1. FERROMAGNETISM

We shall write the Hamiltonian of a system of spins $\hbar \mathbf{S}_j$ in the usual form:[3,7]

$$\mathcal{H} = -J\sum_{jl}\mathbf{S}_j\mathbf{S}_{j+l} - \mu H\sum_j S_j^z - \mu H_A\sum_j S_j^z,$$

$$\mu = ge\hbar/2mc, \quad J > 0, \tag{1}$$

$$\{S_j^x S_k^y\} = S_j^x S_k^y - S_k^y S_j^x$$

$$= i\delta_{jk}S_j^z, \ldots, (S_j^x)^2 + (S_j^y)^2 + (S_j^z)^2 = S(S+1). \tag{2}$$

In Eq. (1), the summation over l is carried out over nearest neighbors, each pair of spins being represented twice. (Thus, the quantity commonly designated as the exchange integral is exactly equal to J, while the interaction energy of a pair of spins is equal to $2J\mathbf{S}_j\mathbf{S}_k$.) H is the magnetic field intensity, and H_A is the effective field of the magnetic anisotropy, both lying in the direction of the z axis. In Eq. (1), we can also easily take into account anisotropic terms of a more general form, but shall not do this here.

We transform to the variables

$$S_\lambda = N^{-1/2}\sum_j \exp(i\lambda j) S_j, \qquad S_j = N^{-1/2}\sum_\lambda S_\lambda \exp(-i\lambda j)$$

$$\sum_j \exp i(\mu - j\lambda) = N\delta_{\lambda\mu}, \tag{3}$$

where N is the total number of particles (spins), j is the radius vector of the j-th lattice point, and the wave vector λ also takes on N different values. If the spins are located at the points of a simple rectangular (orthorhombic) lattice with lattice parameters a_1, a_2, and a_3 along the axes $x \equiv 1$, $y \equiv 2$, and $z \equiv 3$, while the specimen has dimensions n_1a_1, n_2a_2, and n_3a_3 along these axes,

$$N = n_1n_2n_3, \qquad \lambda_i = 2\pi m_i / n_i a_i, \quad -n_i/2 \leqslant m_i \leqslant n_i/2,$$

$$m_i = 0, \pm 1, \pm 2 \ldots .$$

Here we assume that $n_i \gg 1$; in the contrary case, boundary conditions are essential, and then, for example, $m_i \leq n_i/2 - 1$. The limitation to the case in which $n_i \gg 1$ is not obligatory, and does not prevent an approximate study of very small objects, for example, with $n_i \gtrsim 10$.

In terms of the new variables,

$$\mathcal{H} = -JZ\sum_\lambda \gamma_\lambda S_\lambda S_{-\lambda} - \mu(H + H_A)N^{1/2}S_0^z,$$

$$\{S_\lambda^x S_\rho^y\} = iN^{-1/2}S_{\lambda+\rho}^z \text{ etc.}, \qquad Z\gamma_\lambda = \sum_l \exp(i\lambda l), \tag{4}$$

where Z is the number of nearest neighbors. Hence,

$$\dot{S}_\rho^x = \frac{i}{\hbar}\{\mathcal{H}S_\rho^x\} = (\omega_H + \omega_A)S_\rho^y + \frac{\omega_0}{2}N^{-1/2}\sum_\lambda(\gamma_\lambda - \gamma_{\lambda-\rho})(S_\lambda^z S_{\rho-\lambda}^y - S_\lambda^y S_{\rho-\lambda}^z);$$

$$\dot{S}_\rho^y = -(\omega_H + \omega_A)S_\rho^x + \frac{\omega_0}{2}N^{1/2}\sum_\lambda(\gamma_\lambda - \gamma_{\lambda-\rho})(S_\lambda^x S_{\rho-\lambda}^z - S_\lambda^z S_{\rho-\lambda}^x),$$

$$\omega_H = \mu H/\hbar = geH/2mc, \quad \omega_A = \mu H_A/\hbar, \qquad \omega_0 = 2JZ/\hbar. \tag{5}$$

We shall now consider that the system has been magnetized along the z axis, so that the mean value of the z component of the total spin $S_{\text{total}}^z = \sum S_j^z = N^{1/2}S_0^z$ is large in comparison with the root-mean-square values of the quantities $S_{\text{total}}^{x,y} = \sum S_j^{x,y} = N^{1/2}S_0^{x,y}$. The quantity S_0^z will be considered to be a classical quantity, and will be conveniently designated as $N^{1/2}\eta S$, where ηS is the mean value of the z component of the total spin per particle. This condition has the following form, where the brackets $< >$ denote the statistical mean value:

$$N\eta S = \langle S_{\text{total}}^z\rangle \gg \langle (S_{\text{total}}^{x,y})^2\rangle^{1/2}. \tag{6}$$

As we shall see, this condition is not at all sufficient, but when it is obeyed, the linearization of Eq. (5) may lead to reasonable results. Proceeding thus, i.e., replacing S_0^z by $\langle S_0^z\rangle = N^{1/2}\eta S$, we have

$$\dot{S}_\lambda^x = \omega_\lambda S_\lambda^y, \qquad \dot{S}_\lambda^y = -\omega_\lambda S_\lambda^x,$$

$$\omega_\lambda = \omega_H + \omega_{A0}\eta + \omega_0\eta S[1 - (\gamma_\lambda + \gamma_{-\lambda})/2], \tag{7}$$

Here we assume that $\omega_A = \omega_{A_0}\eta$, since the field H_A itself vanishes for $\eta = 0$ in the self-consistent approximation, and in the simplest case, H_A is proportional to η (i.e., $H_A = H_{A_0}\eta$, where $H_{A_0} = \text{const.}$). Equation (7) is derived from the Hamiltonian*

$$\mathcal{H}_0 = \sum_\lambda{}' \frac{\hbar\omega_\lambda}{\eta S}(S_\lambda^x S_{-\lambda}^x + S_\lambda^y S_{-\lambda}^y)$$

$$= \sum_\lambda{}' \frac{\omega_\lambda}{2}(P_{\lambda 1}^2 + Q_{\lambda 1}^2 + P_{\lambda 2}^2 + Q_{\lambda 2}^2)$$

$$= \sum_\lambda{}' \hbar\omega_\lambda(n_{\lambda 1} + n_{\lambda 2} + 1) = \sum_\lambda \hbar\omega_\lambda\left(n_\lambda + \frac{1}{2}\right),$$

$$\{S_\lambda^x S_{-\lambda}^y\} = iN^{-1/2}S_0^z = i\eta S, \qquad S_{\pm\lambda}^x = \sqrt{\eta S/2\hbar}\,(Q_{\lambda 1} \pm iQ_{\lambda 2}),$$

$$S_{\pm\lambda}^y = \sqrt{\eta S/2\hbar}\,(P_{\lambda 1} \pm iP_{\lambda 2}), \quad \{P_{\lambda l}Q_{\mu m}\} = -i\hbar\delta_{lm}\delta_{\lambda\mu}. \tag{8}$$

Thus, the excitation energy is equal to a sum of oscillator energies.

If we assume that $\eta = 1$, the expression obtained obviously corresponds exactly with the ordinary spin-wave approximation. At low temperatures, under the assumption that saturation ($\eta = 1$) occurs at T = 0, the spin-wave approximation for ferromagnetic materials is very good.[3,7] On the contrary, at increased temperatures, and especially at $T \sim \Theta$ (Θ is the Curie point), the ordinary spin-wave approximation is completely unsuitable. This latter situation is quite understandable, since with the decrease in the mean spin ηS occurring as the temperature is raised, the frequency of the spin waves must decrease [see Eq. (7)], a fact not taken into account. In this regard, we may hope that, without fixing the parameter η, but rather, by determining it by the self-consistent method, we may construct a theory having a region of applicability considerably greater than that in which $\eta = \text{const.}$

To determine η, we shall use the exact operator equation

$$(S_j^x)^2 + (S_j^y)^2 + (S_j^z)^2 = S(S+1),$$

averaged over the states of the system. If we also take into account [see Eq. (8)] the fact that

---

*The prime denotes summation over positive values only of $\boldsymbol{\lambda} = (\lambda_1, \lambda_2, \lambda_3)$. In order to avoid introduction of the quantum numbers n_{λ_1} and n_{λ_2} into Eq. (8), we have assumed that $n_\lambda = n_{\lambda_1}$ ($\lambda \geq 0$) and $n_\lambda = n_{\lambda_2}$ ($\lambda < 0$).

$$\langle S_j^x S_j^x + S_j^y S_j^y\rangle = \frac{1}{N}\Big\langle\sum_\lambda (S_\lambda^x S_{-\lambda}^x + S_\lambda^y S_{-\lambda}^y)\Big\rangle$$
$$= 2\eta\frac{S}{N}\sum\Big(n_\lambda + \frac{1}{2}\Big),$$

we have

$$\frac{2\eta S}{N}\sum n_\lambda + \eta S + \langle (S_j^z)^2\rangle = S(S+1). \qquad (9)$$

Obviously, for spin ½, $(S_j^z)^2 = 1/4$. Then, in a state of thermal equilibrium,* in which $\bar{n}_\lambda = [\exp(h\omega_\lambda/kT) - 1]^{-1}$, we obtain the following equation for η:

$$\eta = 1 - \frac{2\eta}{N}\sum_\lambda\Big[\exp\frac{\hbar\omega_\lambda}{kT} - 1\Big]^{-1},$$
$$\omega_\lambda = \omega_H + \omega_{A0}\eta + \frac{\omega_0}{2}\Big(1 - \frac{\gamma_\lambda + \gamma_{-\lambda}}{2}\Big)\eta. \qquad (10)$$

At low temperatures, at which $1 - \eta \ll 1$, Eq. (10) coincides with the result of the ordinary spin-wave theory:[3]

$$SN(1-\eta) = \sum_\lambda\Big[\exp\frac{\hbar\omega_\lambda}{kT} - 1\Big]^{-1},$$
$$\omega_\lambda = \omega_H + \omega_{A0} + \omega_0 S\Big(1 - \frac{\gamma_\lambda + \gamma_{-\lambda}}{2}\Big). \qquad (11)$$

If in Eq. (9) we assume that $\langle (S_j^z)^2\rangle = \eta^2 S^2$ for spin S > ½, then on going to low temperatures we obtain Eq. (11), with the substitution of S + ½ for S. In order to establish complete correspondence, we may assume that

$$\langle (S_j^z)^2\rangle = \eta^2 S^2 - S(\eta^2 - 1)/2.$$

Then the equation for η is:

$$(\eta^2 - 1)\Big(S - \frac{1}{2}\Big) + \eta = 1 - \frac{2\eta}{N}\sum\Big(\exp\frac{\hbar\omega_\lambda}{kT} - 1\Big)^{-1},$$
$$\omega_\lambda = \omega_H + \omega_{A0}\eta + \omega_0 S\Big(1 - \frac{\gamma_\lambda + \gamma_{-\lambda}}{2}\Big)\eta. \qquad (12)$$

For S = ½, this equation goes over into Eq. (10); at T → 0, it goes over into Eq. (11); while for T → Θ it gives a reasonable result (see below). In other respects, however, it is not as firmly based as Eq. (10).

In the high-temperature region (near the Curie point Θ), the parameter $\eta \ll 1$, and $\eta(\Theta) = 0$. Hence, by using the expansion

$$(e^x - 1)^{-1} = 1/x - 1/2 + x/12 - x^3/720 + \ldots$$

in the first-order approximation, for H = 0, we obtain

$$\eta^2 = A(1 - T/\Theta), \qquad \Theta = (S + 1/2)SJ/kF,$$
$$F = \frac{1}{N}\sum\Big(\delta + Z - \sum_{\mathbf{l}}\cos\boldsymbol{\lambda}\mathbf{l}\Big)^{-1}, \ A = \frac{S + 1/2}{S - 1/2 + (1+\delta)JSZ/3k\Theta},$$
$$\frac{1}{N}\sum_\lambda\Big(Z - \sum_{\mathbf{l}}\cos\boldsymbol{\lambda}\mathbf{l}\Big) = Z, \qquad \delta = \frac{\mu H_{A0}}{2JS}. \qquad (13)$$

Such a law for the magnetization

$$M = \mu SN\eta/V = \text{const}\sqrt{1 - T/\Theta}$$

(here V is the volume of the specimen) must be obtained when fluctuations are neglected.[8]

For a simple cubic lattice (with the number Z of nearest neighbors = 6, and with lattice parameter a), upon replacing summation by integration, for $\delta = 0$, we obtain

$$F = \frac{a^3}{(2\pi)^3}\int\frac{d\boldsymbol{\lambda}}{\Big(6 - \sum_{\mathbf{l}}\cos\boldsymbol{\lambda}\mathbf{l}\Big)}$$
$$= \frac{1}{2\pi^3}\iiint_0^\pi \frac{du\,dv\,dw}{3 - \cos u - \cos v - \cos w} = 0.2526. \qquad (14)$$

Thus, for spin S = ½, we have kΘ/J = 1.98 and $\eta^2 = 1.98(1 - T/\Theta)$. In addition, calculations carried out in the paramgnetic region for the model being studied under the same conditions give values kΘ/J = 1.93, 1.92, and 1.83.[9-11] The excellent agreement obtained is better than might be expected. This may apparently be explained by the smallness of the role of large values of **λ**. For the relatively long wavelengths in Eq. (14), the spin-wave approximation, starting with a state with magnetic moment $M(T) = \mu\eta(T)SN$, must be good.

We note that attempts to widen the field of applicability of the spin-wave theory by means of some self-consistent treatment have been made previously.[12-15] However, the method of calculation used by Heber[12] and Schafroth[13] seems to us to be more complex, while the results either do not apply at all[12] to the region T ~ Θ, or are poorly applicable. In the papers of Bogolyubov and Tyablikov,[14,15] double-time retarded and advanced Green's functions were used, and then Eq. (10) was derived approximately for spin ½, with $\omega_{A_0} = 0$. The method given above for deriving this equation seems to us to be not only simpler, but also physically clearer. In addition to the comparison of values of kΘ/J given above, the results obtained by Tyablikov[15] and the comparison of the latter with other solutions give evidence that Eq. (10) is correct at low temperatures within an accuracy of terms of the order of $T^{-5/2}$, inclusive. For H ≠ 0 and T > Θ, it is correct within an curacy of terms of the order of 1/T.

Thus, there is every ground to consider that the

---

*In the case of a spectrum of quasi-particles having energy levels depending on the energy, as is known, the ordinary formula for $\bar{n}$ may be derived from the expression for the entropy.

very simple self-consistent treatment applied here gives good results. The character of the approximation is not associated here with concrete expressions for the frequencies ω_λ. In other words, by using Eqs. (10) and (12) with frequencies $\omega_\lambda = \omega_H + \omega_A + f(\lambda)\eta$, where f is some reasonably selected function, we arrive at a phenomenological self-consistent theory of spin waves. The essential limitation here is actually the fact that the original Hamiltonian in Eq. (1) commutes with $S^z_{\text{total}} = \sum S^z_j$. We shall not carry out here the possible generalization of the self-consistent theory to the case in which there is an anisotropic interaction of a more general type. Rather, we shall consider further only ferromagnetic materials of small dimensions.

Within the framework of theory being discussed, we retain Eqs. (10) — (13) in this case, except that we cannot go from summation to integration in all directions.* Thus, for thin films, we must retain the summation in the direction perpendicular to the plane of the film. For long "fibers" we must apply summation in two directions, and for parallelepipeds, in all three directions. In the case of a simple cubic lattice with dimensions n_1a, n_2a, and n_3a: $N = n_1n_2n_3$, and

$$F = \frac{4}{n_1n_2n_3}\sum_{l_1l_2l_3}\left(\frac{\delta}{2} + 3 - \cos\frac{2\pi m_1}{n_1} - \cos\frac{2\pi m_2}{n_2} - \cos\frac{2\pi m_3}{n_3}\right)^{-1},$$

$$\frac{\delta}{2} = \frac{\mu H_{A0}}{4JS}, \qquad m_i = 0, 1, \ldots, n_i/2. \tag{15}$$

In the three-dimensional case in sufficiently large systems, the factor taking into account anisotropy plays in certain respects simply the role of a convergence factor, eliminating the infinite term in F for the case $m_1 = m_2 = m_3 = 0$ [see Eq. (15)].* However, in the one- and two-dimensional problems, the results already depend fundamentally on δ. Thus, for an infinite chain of spins we have:

$$F = \frac{1}{2\pi}\int_0^\pi \frac{du}{\delta/2 + 1 - \cos u} \approx \frac{1}{2\sqrt{\delta}}, \qquad \delta \ll 1. \tag{16}$$

In view of Eqs. (13) and (14), we have for such a chain,

$$\Theta = \left[\sqrt{2S}\left(S + \tfrac{1}{2}\right)/k\right]\sqrt{\mu H_{A0} J}.$$

At the same time, it follows from general statistical considerations[18] that a phase transition in an infinite one-dimensional system is impossible, i.e., $\Theta = 0$. The same is naturally true in the Ising model,[19] which corresponds to the limit of strong anisotropy.

With regard to the discussion above, we note firstly that the introduction of the field $H_A = H_{A_0}\eta$ is in itself an approximate method, and introduces an additional element of self-consistency. Actually, the true interaction in one separate direction has the form $\sum S^z_j S^z_{j+1}$, but this expression has been replaced by one of the type $\langle S^z\rangle \sum S^z_j = \eta S \sum S^z_j$. As a result, the effect of the anisotropy field is equivalent to the effect of a certain external magnetic field bringing about the appearance of a magnetic moment in the chain for $T \neq 0$. Secondly, even independently of the problem of the nature of the anisotropy field, the original introduction of the mean quantity $\langle S^z_j\rangle = \eta S$ is permissible in the statistical calculations, in general, only under the condition that the fluctuations are small. In other words, in addition to condition (6), the following condition must be fulfilled:

$$\langle(\Delta\eta)^2\rangle \ll \langle\eta\rangle^2 \equiv \eta^2, \qquad \Delta\eta = \eta - \langle\eta\rangle \tag{17}$$

(to avoid complicating the notation, we shall write

---

*The presence of spontaneous magnetization in ferromagnetic materials and the magnetic effects associated therewith may be taken into account to a certain degree by the introducduction of the field H_A. However, this field depends on the form and dimensions of the specimen, just as does the magnetization itself. We shall not consider this important magnetic moment in this article, just as we shall not in general analyze the question of the nature and magnitude of the anisotropy field H_A. For the antiferromagnetic materials, in which we are chiefly interested, the entire problem of the relation of H_A to the dimensions and form is, in a certain respect, considerably less critical, owing to the lack of spontaneous magnetization. However, in the case of the ferromagnetic materials, the corresponding relation may be taken into account to a certain degree as the result of the solution of magnetostatic problems. Besides, in both the ferromagnetic and the antiferromagnetic materials, the form and dimensions of the specimens exhibit, of course, an influence on sufficiently long waves. Hence, in the region of sufficiently low temperatures, the problem of the dependence of various quantities on the dimensions and form requires special study (for the latter remark, the authors wish to thank M. I. Kaganov).

*In this regard, one of the ways of calculating sums of the type of (15) is simply the omission of terms with $m_1 = m_2 = m_3 = 0$, anisotropy being simultaneously neglected (see, e.g., reference 16). The results of such a calculation are given below for illustration (for the most recent experimental data on films, see the paper of Neugebauer[17]):

$(n_1n_2n_3)=$	$(\infty\infty\infty)$	(64,64,64)	(32,32,32)	(16,16,16)	(4,4,4)	(4,4,16)	(4,4,64)
F=	0.2526	0.29	0.32	0.37	0.66	0.75	1.7
$(n_1n_2n_3)=$	(8,8,64)	(4,16,16)	(4,64,64)	(8,16,16)	(8,32,32)	(8,64,64)	(8,128,128)
F=	0.67	0.55	0.61	0.32	0.41	0.42	0.44

It is clear from Eqs. (13) and (15) that it is possible to neglect the terms with $m_1 = m_2 = m_3 = 0$ if $N\delta \gg 1$. When $N\delta \ll 1$, obviously $F \approx (N\delta)^{-1}$. However, in this case the conditions for validity of the approximation are not fulfilled (see criterion (21) below). Hence, and also in view of the requirement $n_i \gg 1$, by no means all given values of F can be used.

η for $<\eta>$ whenever this does not lead to confusion).

For a concrete application of condition (17), as well as for a more complete test of the linear approximation used here, we would, of course, have to determine all of the physical quantities from the next order of approximation. However, we shall not go through this involved procedure here, but rather, shall estimate the fluctuation of η within the framework of the semi-thermodynamic theory. Near the Curie point, neglecting fluctuations,[8] the thermodynamic potential per unit volume can be expressed in the form

$$\Phi = \Phi_0 + \alpha\eta^2 + \tfrac{1}{2}\beta\eta^4 + \rho(\mathrm{grad}\,\eta)^2 - \eta H\mu SNV^{-1}, \tag{18}$$

where $\eta(\mathrm{r})$ is the characteristic parameter,[18,8] for which we may also select the quantity $<\eta>$ used above, as applied to a state of incomplete thermodynamic equilibrium. Assuming that

$$\Delta\eta = \sum \eta_\lambda e^{i\lambda r},$$

as usual, we then determine the thermodynamic fluctuations at $H = 0$ (for more detail, see reference 8 and the literature cited there):

$$\langle|\eta_\lambda|^2\rangle = kT/(\Phi_0'' + 2\rho\lambda^2)V,$$

$$\langle(\Delta\eta)^2\rangle = \left\langle\int(\Delta\eta(\mathrm{r}))^2\,dV\right\rangle = \sum_\lambda \langle|\eta_\lambda|^2\rangle,$$

$$\Phi_0'' = (\partial^2\Phi/\partial\eta^2)_{\eta=\eta_0}, \qquad \Phi_0'' = -4\alpha \quad (\text{at } T < \Theta),$$

$$\eta_0^2 \equiv \langle\eta\rangle^2_{\mathbf{equil}} = -\alpha/\beta \approx \alpha'_\Theta(\Theta - T)/\beta_\Theta \quad (\text{at } T < \Theta),$$

$$\eta_0^2 = 0 \quad (\text{at } T > \Theta). \tag{19}$$

Equation (18) may be used under condition (17) at temperatures not too far from the Curie point (for $\eta \ll 1$), and for wave vectors $\lambda \ll \lambda_{\mathrm{max}} = \pi/a$. However, in order to make the estimates in Eq. (19), we must carry out the summation or integration up to values $\lambda \sim \lambda_{\mathrm{max}}$. As a result of such an estimate and the use of condition (17) for the three-dimensional case, criteria may be obtained, which have previously been discussed in reference 8. In real ferromagnetic materials, the situation is rather unclear, but apparently condition (17) is well obeyed when $\Delta T \gg 10^{-2}\Theta$ (in the case of metals this condition may be even weaker).

It is obvious from Eq. (19)that the fluctuations are greater in the two-dimensional, and especially in the one-dimensional case, than in the three-dimensional case. Thus, for an infinite chain

$$\langle(\Delta\eta)^2\rangle \sim \frac{kT}{2\pi}\int_0^{\lambda_{max}} \frac{d\lambda}{\Phi_0'' + 2\rho\lambda^2}$$

$$= \frac{kT}{2\pi\sqrt{2\rho\Phi_0''}}\tan^{-1}\left(\sqrt{\frac{2\rho}{\Phi_0''}}\,\lambda_{max}\right) \approx \frac{kT}{4\sqrt{2\rho\Phi_0''}} \tag{20}$$

Further, from Eqs. (13) and (16) we obtain

$$\eta^2 \sim (k\Theta/J)(1 - T/\Theta), \qquad k\Theta \sim S^{3/2}\sqrt{\mu H_{A0}J};$$

In view of (18) (or more exactly, from the condition that $\partial\Phi/\partial\eta = 0$), in the region $T > \Theta$ in the presence of a magnetic field

$$\eta = \mu SNH/2V\alpha = \mu SNH/2V\alpha'_\Theta(T - \Theta).$$

At the same time, we may derive from Eq. (12), for $H \neq 0$ and $T \gg \Theta$, the equation $\eta = (S + 1/2) \times \mu H/2kT$. Hence $\alpha'_\Theta \sim kN/V = k/a$ (in the one-dimensional case, $N/V = N/L = 1/a$, where L is the length of the chain and a is the distance between neighboring spins). Finally, the coefficient $\rho \sim Ja$, since for the shortest waves ($\lambda \sim \lambda_{\mathrm{max}} = \pi/a$), the energy $\rho(d\eta/dz)^2 \sim \rho\lambda^2_{\mathrm{max}} \sim \rho/a^2$ must be of the order of the exchange energy J/a per unit length of chain.

In view of all that has been said, condition (17) acquires the form

$$\eta^2 \sim \frac{k}{J}(\Theta - T) \gg \langle(\Delta\eta)^2\rangle \sim kT/10\sqrt{k(\Theta - T)J}$$

or

$$(\Theta - T)/\Theta \gg (J/k\Theta)^{1/3} \gtrsim 1$$

(We assume that $T \sim \Theta$, since all of the estimates refer to the region near the Curie point, and have been made with a corresponding degree of precision). The condition which we have derived obviously cannot be fulfilled. Thus, for an infinite chain the approximation is indeed inapplicable, in agreement with the general results. From this example, the procedure is clear by which we may test the region of applicability of our approximation.

We note that a finite chain (two- and three-dimensional systems not yet being discussed) can be ferromagnetic (possess a magnetic moment) even for $T \neq 0$ (for an infinite chain, long-range order essentially vanishes, while any finite set of spins may possess a magnetic moment differing from zero as the result of short-range order). In the case of finite systems, condition (6) is also essential. According to Eqs. (3) and (8),

$$S^x_{\mathbf{total}} = \sum_j S^x_j = N^{1/2}S^x_0,$$

$$\langle(S^x_{\mathbf{total}})^2\rangle = N\langle(S^x_0)^2\rangle$$

$$= \eta SN\left(\bar{n}_0 + \tfrac{1}{2}\right) = \eta SN\left\{\left(\exp\frac{\hbar\omega_{\lambda=0}}{kT} - 1\right)^{-1} + \tfrac{1}{2}\right\},$$

$$\omega_{\lambda=0} = \omega_H + \omega_{A0}\eta.$$

Hence, condition (6), within the region in which $\hbar\omega_{\lambda=0}/kT \ll 1$ and $\omega_H \ll \omega_{A_0}\eta$, acquires the form

$$\eta \gg \sqrt{kT/\hbar\omega_{A0}SN} = \sqrt{kT/\mu SH_{A0}N}. \qquad (21)$$

However, if $\omega_H \gg \omega_{A_0}\eta$, the following inequality must be satisfied

$$\eta \gg kT/\hbar\omega_H SN = kT/\mu SHN.$$

When $T \to 0$, we have the condition $\eta \gg 1/N$ in place of Eq. (21). For example, condition (21) for $T \sim \Theta$, $S \sim 1$, and $k\Theta/\mu H_{A_0} \sim 10^6$ ($\Theta \sim 10^3$ deg K, $H_{A_0} \sim 10$ oe) is reduced to the requirement that $\eta \gg 10^3/\sqrt{N}$. Then, for $\eta \sim 0.1$, only sufficiently small particles having $N \lesssim 10^8$ fail to fulfill the condition.

The problem of the behavior of systems having Hamiltonians of the type of Eq. (1) in cases in which conditions (6) and (7) are not fulfilled is highly interesting. Here we may expect very peculiar properties, in particular a high susceptibility χ, showing a tendency toward saturation even at not very strong fields. More concretely, we have in mind systems for which the Curie point $\Theta \ll J/k$ (and as a particular case, $\Theta = 0$). Then, in the region $\Theta < T < J/k$, a law of the type $\chi \sim 1/(T - \Theta)$ apparently need not hold true, since the interaction between the spins $\sim J/k$ is still very strong. A system of spins in such a state may be designated as an "incoherent ferromagnetic material" or a "paramagnetic fluid" with ferromagnetic interaction (see also Sec. 3).

2. ANTIFERROMAGNETISM (GENERAL STUDY)

The Hamiltonian of the model being studied of an antiferromagnetic material in an external field **H** and an anisotropy field $\mathbf{H}_A$ has the form

$$\mathcal{H} = \sum_{j,k} J_{jk}\mathbf{S}_j\mathbf{S}_k - \mu\mathbf{H}\sum_{j,k}(\mathbf{S}_j + \mathbf{S}_k) - \mu\mathbf{H}_A\sum_{jk}(\mathbf{S}_j - \mathbf{S}_k). \qquad (22)$$

Here the subscripts j and k denote the spins situated at the points of the two sublattices 1 and 2, while the sign is chosen such that the exchange integral for the antiferromagnetic interaction $J_{jk} > 0$. N is the total number of spins. We shall introduce the operators $\mathbf{S}_{1,2\lambda}$:

$$\mathbf{S}_\mathrm{j} = \left(\frac{2}{N}\right)^{1/2}\sum_\lambda \mathbf{S}_{1,\lambda}\exp(-i\lambda\mathrm{j}),$$

$$\mathbf{S}_\mathrm{k} = \left(\frac{2}{N}\right)^{1/2}\sum_\lambda \mathbf{S}_{2,\lambda}\exp(-i\lambda\mathrm{k}),\quad \mathbf{S}_{1,\lambda} = \left(\frac{2}{N}\right)^{1/2}\sum_\mathrm{j}\mathbf{S}_\mathrm{j}\exp(i\lambda\mathrm{j}), \qquad (23)$$

where

$$\{S^x_{l,\lambda}S^y_{m\mu}\} = i\sqrt{2/N}\,\delta_{lm}S^z_{l,\lambda+\mu} \quad \text{and its cyclic permutations;} \quad l, m = 1, 2;$$

$$\sum_\mathrm{j}\exp\{i(\lambda - \mu)\mathrm{j}\} = (N/2)\,\delta_{\lambda\mu}.$$

Further, the operator (22) acquires the form

$$\mathcal{H} = 4DJ\sum_\lambda \gamma_\lambda\mathbf{S}_{1,\lambda}\mathbf{S}_{2,-\lambda} - \mu\left(\frac{N}{2}\right)^{1/2}\mathbf{H}(\mathbf{S}_{1,0} + \mathbf{S}_{2,0})$$
$$- \mu\left(\frac{N}{2}\right)^{1/2}\mathbf{H}_A(\mathbf{S}_{1,0} - \mathbf{S}_{2,0}),$$
$$2DJ\gamma_\lambda = \sum_{\mathrm{k-j}} J_\mathrm{jk}\exp\{i\lambda(\mathrm{k} - \mathrm{j})\},$$
$$J = (2D)^{-1}\sum_{\mathrm{k-j}} J_\mathrm{jk},\ \gamma_0 = 1,\ \gamma_\lambda = \gamma_{-\lambda}. \qquad (24)$$

Here the fact has been taken into account that we encounter identical terms twice in summing over j and k in Eq. (22). D is the number of dimensions of the simple lattice being studied. Only nearest neighbors are assumed to interact (i.e., 2D = Z = the number of neighbors). We then obtain the equations of motion from Eq. (24):

$$\dot{\mathbf{S}}_{1,\lambda} = \frac{i}{\hbar}\{\mathcal{H}\mathbf{S}_{1,\lambda}\}$$
$$= \omega_0\sqrt{\frac{2}{N}}\sum_\mu \gamma_\mu[\mathbf{S}_{2,-\mu}\mathbf{S}_{1,\lambda+\mu}] + \frac{\mu}{\hbar}[\mathbf{S}_{1,\lambda},\ \mathbf{H} + \mathbf{H}_A],$$
$$\dot{\mathbf{S}}_{2,\lambda} = \omega_0\sqrt{\frac{2}{N}}\sum_\mu \gamma_\mu[\mathbf{S}_{1,-\mu}\mathbf{S}_{2,\lambda+\mu}]$$
$$+ \frac{\mu}{\hbar}[\mathbf{S}_{2,\lambda},\ \mathbf{H} - \mathbf{H}_A],\ \omega_0 = \frac{4DJ}{\hbar}. \qquad (25)$$

We shall now linearize Eqs. (25) in the vicinity of the mean values:

$$\langle S^1_{\mathrm{total}\,z}\rangle = \sum\langle S_{j,z}\rangle = \sqrt{\frac{N}{2}}S^z_{1,0}$$
$$= \frac{N}{2}\langle S_{jz}\rangle = \frac{N}{2}\eta_1 S,$$
$$\langle S^2_{\mathrm{total}\,z}\rangle = \sum_k\langle S_{kz}\rangle = \sqrt{\frac{N}{2}}S^z_{2,0} = -\frac{N}{2}\eta_2 S. \qquad (26)$$

We have obviously made here the common assumption of the theory of antiferromagnetism that both sublattices possess a large mean spin (magnetization) directed along z or in the opposite direction.[3,6,20-23] Here we shall consider, in the absence of an external field H, that $\eta_1 = \eta_2$, i.e., the system as a whole possesses no magnetic moment.* As is known, unlike in the ferromagnetic case, the problem of the quantum-mechanical nature of the original state used in Eq. (26) is not yet very clear,[3] but we shall not take up this important problem now.

After linearization, Eq. (25) may be written as follows (with the fields **H** and $\mathbf{H}_A$ directed along the z axis):

***We may consider analogously the case of ferromagnetism in which the magnetic moments of the sub-lattices are not equal in absolute value, even at H = 0.**

$$\dot{S}^x_{1,\lambda} = (\omega_0 S\eta_2 + \omega_{A0}\eta_2 + \omega_H)\, S^y_{1,\lambda} + \omega_0 S\eta_1\gamma_\lambda S^y_{2,\lambda},$$
$$\dot{S}^x_{2,\lambda} = -(\omega_0 S\eta_1 + \omega_{A0}\eta_1 - \omega_H)\, S^y_{2,\lambda} - \omega_0 S\eta_2\gamma_\lambda S^y_{1,\lambda},$$
$$\dot{S}^y_{1,\lambda} = -(\omega_0 S\eta_2 + \omega_{A0}\eta_2 + \omega_H)\, S^x_{1,\lambda} - \omega_0 S\eta_1\gamma_\lambda S^x_{2,\lambda},$$
$$\dot{S}^y_{2,\lambda} = (\omega_0 S\eta_1 + \omega_{A0}\eta_1 - \omega_H)\, S^x_{2,\lambda} + \omega_0 S\eta_2\gamma_\lambda S^x_{1,\lambda},$$

$$\{S^x_{1,\lambda}S^y_{1,-\lambda}\} = i\eta_1 S, \quad \omega_H = \mu H/\hbar, \quad \omega_{A0} = \mu H_{A0}/\hbar,$$
$$\omega_0 = 4DJ/\hbar, \quad \gamma_\lambda = \gamma_{-\lambda}, \qquad \{S^x_{2,\lambda}S^y_{2,-\lambda}\} = -i\eta_2 S. \quad (27)$$

Here, in the equations for $S^{x,y}_{1,\lambda}$ we have assumed that $(\mu\hbar)H_A = \omega_{A_0}\eta_2$ and in the equations for $S^{x,y}_{2\,\lambda}$, that $(\mu\hbar)H_A = \omega_{A_0}\eta_1$, where $\omega_{A_0} = (\mu\hbar)H_{A_0} = \text{const.}$ Of course this means in essence that we have had to introduce two fields H_A differing for the two sublattices, from Eq. (22) on. By considering the fields H_A to be proportional to $\eta_{1,2}$, we take into account very simply the fact that the anisotropy field H_A itself vanishes when $\eta_{1,2} = 0$. The system (27) is derived from the Hamiltonian

$$\mathcal{H} = \frac{\hbar}{S}\sum_\lambda \frac{1}{2\eta_1}(\omega_0 S\eta_2 + \omega_{A0}\eta_2 + \omega_H)(S^x_{1,\lambda}S^x_{1,-\lambda} + S^y_{1,\lambda}S^y_{1,-\lambda})$$
$$+ \frac{1}{2\eta_2}(\omega_0 S\eta_1 + \omega_{A0}\eta_1 - \omega_H)(S^x_{2,\lambda}S^x_{2,-\lambda} + S^y_{2,\lambda}S^y_{2,-\lambda})$$
$$+ \omega_0 S\gamma_\lambda(S^x_{1,\lambda}S^x_{2,-\lambda} + S^y_{1,\lambda}S^y_{2,-\lambda}). \quad (28)$$

We shall now transform to new variables

$$S^x_{1,\lambda} = (c_{1,\lambda}Q_{1,\lambda} + c_{2,\lambda}Q_{2,-\lambda})\sqrt{\eta_1 S},$$
$$S^y_{1,\lambda} = (c_{1,\lambda}P_{1,-\lambda} - c_{2,\lambda}P_{2,-\lambda})\sqrt{\eta_1 S},$$
$$S^x_{2,-\lambda} = (c_{2,\lambda}Q_{1,-\lambda} + c_{1,\lambda}Q_{2,\lambda})\sqrt{\eta_2 S},$$
$$S^y_{2,-\lambda} = (c_{2,\lambda}P_{1,\lambda} - c_{1,\lambda}P_{2,-\lambda})\sqrt{\eta_2 S},$$
$$\{Q_{l\lambda}P_{m\mu}\} = i\delta_{lm}\delta_{\lambda\mu},$$

where

$$c_{1,\lambda} = c_{1,-\lambda}, \qquad c_{2\lambda} = c_{2,-\lambda}, \qquad c^2_{1,\lambda} - c^2_{2,\lambda} = 1,$$
$$c_{1,\lambda} = \rho_\lambda / \sqrt{\rho^2_\lambda - \gamma^2_\lambda}, \qquad c_{2,\lambda} = -\gamma_\lambda / \sqrt{\rho^2_\lambda - \gamma^2_\lambda},$$
$$\rho_\lambda = (1+\delta)\left(\frac{\eta_1+\eta_2}{2\sqrt{\eta_1\eta_2}}\right) + \left[(1+\delta)^2\left(\frac{\eta_1+\eta_2}{2\sqrt{\eta_1\eta_2}}\right)^2 - \gamma^2_\lambda\right]^{1/2},$$
$$\delta_a \equiv \delta = \frac{\omega_{A0}}{\omega_0 S}.$$

In terms of these new variables, the Hamiltonian acquires the form

$$\mathcal{H} = \frac{\hbar}{2}\sum_\lambda \omega^+_\lambda (Q_{1,\lambda}Q_{1,-\lambda} + P_{1,\lambda}P_{1,-\lambda})$$
$$+ \omega^-_\lambda (Q_{2,\lambda}Q_{2,-\lambda} + P_{2,\lambda}P_{2,-\lambda}),$$

where

$$\omega^+_\lambda = \omega_0 S\,\frac{(1+\delta)(\eta_1\gamma^2_\lambda + \eta_2\rho^2_\lambda) - 2\sqrt{\eta_1\eta_2}\,\gamma^2_\lambda\rho_\lambda}{\rho^2_\lambda - \gamma^2_\lambda} + \omega_H,$$
$$\omega^-_\lambda = \omega_0 S\,\frac{(1+\delta)(\eta_2\gamma^2_\lambda + \eta_1\rho^2_\lambda) - 2\sqrt{\eta_1\eta_2}\,\gamma^2_\lambda\rho_\lambda}{\rho^2_\lambda - \gamma^2_\lambda} - \omega_H. \quad (29)$$

If we assume that $\eta_1 = \eta_2 = \eta$,

$$\omega^\pm_\lambda = \eta\omega_0 S\sqrt{(1+\delta)^2 - \gamma^2_\lambda} \pm \omega_H = \omega_\lambda \pm \omega_H. \quad (29a)$$

In order to express the Hamiltonian as a sum of Hamiltonians of harmonic oscillators, we shall transform to the variables

$$Q_{1,\pm\lambda} = \sqrt{\tfrac{1}{2}}\,(q_{1,\lambda} \pm iq_{2,\lambda}), \qquad P_{1,\pm\lambda} = \sqrt{\tfrac{1}{2}}\,(p_{1,\lambda} \mp ip_{2,\lambda}),$$
$$Q_{2,\pm\lambda} = \sqrt{\tfrac{1}{2}}\,(q_{3,\lambda} \pm iq_{4,\lambda}),$$
$$P_{2,\pm\lambda} = \sqrt{\tfrac{1}{2}}\,(p_{3,\lambda} \mp ip_{4,\lambda}); \quad \{q_{l\lambda}p_{m\mu}\} = i\delta_{lm}\delta_{\lambda\mu}. \quad (30)$$

In terms of these variables, the Hamiltonian operator becomes

$$\mathcal{H} = \sum_{\lambda \geqslant 0}{}' \{\tfrac{1}{2}(q^2_{1,\lambda} + p^2_{1,\lambda} + q^2_{2,\lambda} + p^2_{2,\lambda})\hbar\omega^+$$
$$+ \tfrac{1}{2}(q^2_{3,\lambda} + p^2_{3,\lambda} + q^2_{4,\lambda} + p^2_{4,\lambda})\hbar\omega^-\}.$$

The matrix elements of the operators q and p are equal to:

$$(q_{l,\lambda})_{n,\,n+1} = \sqrt{(n+1)/2} = (q_{l,\lambda})_{n+1,\,n},$$
$$(p_{l,\lambda})_{n,\,n+1} = -(p_{l,\lambda})_{n+1,\,n} = -i\sqrt{(n+1)/2}. \quad (32)$$

The eigenvalues of the energy of the spin waves are:

$$\mathcal{H} = \sum_{\lambda \geqslant 0}{}' (n_{1,\lambda} + \tfrac{1}{2} + n_{2,\lambda} + \tfrac{1}{2})\,\hbar\omega^+_\lambda$$
$$+ (n_{3,\lambda} + \tfrac{1}{2} + n_{4,\lambda} + \tfrac{1}{2})\,\hbar\omega^-_\lambda,$$

or

$$\mathcal{H} = \sum_\lambda (n^+_\lambda + \tfrac{1}{2})\,\hbar\omega^+_\lambda + (n^-_\lambda + \tfrac{1}{2})\,\hbar\omega^-_\lambda. \quad (33)$$

We shall use the following equations as conditions of self-consistency:

$$\langle (S^x_j)^2 + (S^y_j)^2 \rangle = \frac{2}{N}\sum_\lambda \langle S^x_{1,\lambda}S^x_{1,-\lambda}$$
$$+ S^y_{1,\lambda}S^y_{1,-\lambda}\rangle = S(S+1) - \langle (S^z_j)^2 \rangle,$$
$$\langle (S^x_k)^2 + (S^y_k)^2 \rangle = \frac{2}{N}\sum_\lambda \langle S^x_{2,\lambda}S^x_{2,-\lambda}$$
$$+ S^y_{2,\lambda}S^y_{2,-\lambda}\rangle = S(S+1) - \langle (S^z_k)^2 \rangle, \quad (34)$$

which may be written in the form

$$\eta_1 S(G_D + G^{(1)}_{DT}) = S(S+1) - \langle (S^z_j)^2 \rangle,$$
$$\eta_2 S(G_D + G^{(2)}_{DT}) = S(S+1) - \langle (S^z_k)^2 \rangle;$$

$$G_D = \frac{2}{N}\sum_\lambda \frac{\rho^2_\lambda + \gamma^2_\lambda}{\rho^2_\lambda - \gamma^2_\lambda}$$
$$= \frac{2}{N}\sum_\lambda \frac{(1+\delta)(\eta_1+\eta_2)/2\sqrt{\eta_1\eta_2}}{\{(1+\delta)^2[(\eta_1+\eta_2)/2\sqrt{\eta_1\eta_2}]^2 - \gamma^2_\lambda\}^{1/2}};$$

$$G^{(1)}_{DT} = \frac{4}{N}\sum_\lambda \frac{\rho^2_\lambda\overline{n^+_\lambda} + \gamma^2_\lambda\overline{n^-_\lambda}}{\rho^2_\lambda - \gamma^2_\lambda}, \quad G^{(2)}_{DT} = \frac{4}{N}\sum_\lambda \frac{\gamma^2_\lambda\overline{n^+_\lambda} + \rho^2_\lambda\overline{n^-_\lambda}}{\rho^2_\lambda - \gamma^2_\lambda},$$

$$\overline{n^\pm_\lambda} = \left[\exp\frac{\hbar\omega^\pm_\lambda}{kT} - 1\right]^{-1}. \quad (35)$$

For the case of spin $1/2$, to which we shall limit ourselves for simplicity, $(S_j^z)^2 = (S_k^z)^2 = 1/4$, and

$$\eta_1(G_D + G_{DT}^{(1)}) = 1, \qquad \eta_2(G_D + G_{DT}^{(2)}) = 1. \qquad (36)$$

In the absence of a magnetic field, $\eta_1 = \eta_2 = \eta$ for an antiferromagnetic material, Eqs. (35) and (36) coincide, and for spin $S = 1/2$ we have

$$\eta\,(J_D + J_{DT}) = 1, \qquad J_D = \frac{2}{N}\sum\frac{1+\delta}{[(1-\delta)^2 - \gamma_\lambda^2]^{1/2}},$$

$$J_{DT} = \frac{4}{N}\sum\frac{1+\delta}{[(1+\delta)^2 - \gamma_\lambda^2]^{1/2}}(e^{\hbar\omega_\lambda/kT} - 1)^{-1},$$

$$\omega_\lambda = \frac{\eta\omega_0}{2}[(1+\delta)^2 - \gamma_\lambda^2]^{1/2}, \quad \delta = \frac{2\omega_{A0}}{\omega_0}, \quad \omega_0 = \frac{4DJ}{\hbar}. \qquad (37)$$

Obviously, when $T = 0$,

$$\eta(0) = J_D^{-1}. \qquad (38)$$

For two- and three-dimensional lattices of large dimensions, the integral J_D converges even when $\delta = 0$. Then $J_D = (2\pi)^{-D}\int d\lambda\,\sqrt{1-\gamma_\lambda^2}$, and according to Anderson,[20] $J_2 = 1.393$ and $J_3 = 1.156$. Thus, for the two- and three-dimensional cases, respectively, $\eta(0) = 0.72$ and $\eta(0) = 0.865$. The latter result agrees with references 3 and 20, but is more precise. For an infinite linear chain with $\delta \ll 1$, $J_D = J_1 \approx \pi^{-1}\,|\ln\delta\,|$. Hence, the value of $\eta(0)$ is not zero. In fact, as in the ferromagnetic case, there is no long-range order in a linear antiferromagnetic chain. However, the result obtained by neglecting fluctuations seems more natural than that stated by Anderson.[20]

Near the Néel point, when $\eta \ll 1$, we have

$$\eta^2 = A\left(1 - \frac{T}{\Theta}\right), \quad \Theta = \frac{DJ}{kF_a}, \quad F_a = \frac{2}{N}\sum\frac{1+\delta}{(1+\delta)^2 - \gamma_\lambda^2},$$

$$A^{-1} = \frac{2}{3N}\sum(1+\delta)\frac{DJ}{k\Theta} = (1+\delta)\frac{DJ}{3k\Theta}. \qquad (39)$$

For the three-dimensional case, as $\delta \to 0$,

$$F_a = \frac{2}{N}\sum\frac{1}{1-\gamma_\lambda^2} = \frac{1}{(2\pi)^3}\int\frac{d\lambda}{1-\gamma_\lambda^2}$$

$$= \frac{1}{\pi^3}\iiint_0^\pi \frac{du\,dv\,dw}{1 - [(\cos u + \cos v + \cos w)/3]^2}$$

$$= \frac{1}{\pi^3}\iiint_0^\pi \frac{du\,dv\,dw}{1 - (\cos u + \cos v + \cos w)/3} = 1.516. \qquad (40)$$

Hence, the ratio $k\Theta/J = 3J/kF_a = 1.98$, i.e., it is the same as in ferromagnetic materials (see above). Unfortunately, there have been no completely reliable statistical calculations of $k\Theta/J$ for antiferromagnetic materials, at least as known to us. According to the calculations in reference 24, which seem to be the most reliable,[23] $k\Theta/J = 2.004$ for a simple cubic lattice. The necessary condition for the applicability of the self-consistent approximation has a form of the type of Eq. (6), namely

$$N\eta S \gg \sqrt{\langle (S_{1,2\,\mathrm{total}}^{x,y})^2\rangle}.$$

It is easy to see that this condition (for high temperatures with $H = 0$) coincides with criterion (21). As $T \to 0$, for antiferromagnetic materials the following inequality must be satisfied, instead of Eq. (21)

$$\eta \gg 1/N\sqrt{\delta} \qquad (\eta \leqslant 1). \qquad (41)$$

We note that a certain self-consistent treatment in the theory of antiferromagnetism has already been carried out by Ziman[21] for the purpose of deriving corrections to the ordinary theory of spin waves; a paper on this latter subject by Oguchi[25] has recently appeared. In addition, P'u Fu-C'ho[26] recently extended the method of Bogolyubov and Tyablikov[14] to the case of antiferromagnetism with spin $1/2$ in the absence of anisotropy. In the assumptions just given, the result obtained by P'u Fu-C'ho[26] and that derived above [see Eqs. (38) — (41)] agree; this may serve as a test of the correctness of the methods of calculation. The use of different methods here seems essential to us, since the comparison with the results of references 24 and 26 gives us additional confidence in the effectiveness, and in general, in the quite high accuracy of the method of calculation which we have used.

In order to calculate the parallel susceptibility $\chi_{\parallel}$, we must know η_1 and η_2 in the presence of a magnetic field. We shall assume that

$$\hbar\omega_H/kT \leqslant 1, \qquad \eta_1 = \eta + \eta_1', \qquad \eta_2 = \eta + \eta_2',$$

where

$$|\eta_{1,2}'| = O(\hbar\omega_H/kT) \leqslant \eta.$$

Then, we obtain from Eqs. (36) and (37), by expansion in series to an accuracy of first-order terms in $\hbar\omega_H/kT$, η_1', and η_2',

$$\eta_1'(J_D + J_{DT}) + \eta\,[\partial(G_D + G_{DT}^{(1)})/\partial\eta_1]_0\,\eta_1' + \eta\,[\partial(G_D + G_{DT}^{(1)})/\partial\eta_2]_0\,\eta_2'$$

$$\eta R\hbar\omega_H/kT = 0.$$

$$\eta_2'(J_D + J_{DT}) + \eta\,[\partial(G_D + G_{DT}^{(2)})/\partial\eta_1]_0\,\eta_1' + \eta\,[\partial(G_D + G_{DT}^{(2)})/\partial\eta_2]_0\,\eta_2' + \eta R\hbar\omega_H/kT = 0,$$

$$R = \frac{1}{N}\sum_\lambda\left[\left(\coth\frac{\hbar\omega_\lambda}{2kT}\right)^2 - 1\right] = \frac{1}{N}\sum_\lambda\left(\sinh\frac{\hbar\omega_\lambda}{2kT}\right)^{-2},$$

$$\eta\,(J_D + J_{DT}) = 1. \qquad (42)$$

The subscript zero in the derivatives with respect to η_1 and η_2 indicates that they are taken for η_1

$= \eta_2 = \eta$, where

$$\left[\frac{\partial}{\partial\eta_{1,2}}\left(G_{DT}^{(1)} - G_{DT}^{(2)}\right)\right]_0 = \frac{4}{N}\sum\left|\frac{\partial(n^+ - n^-)}{\partial\eta_{1,2}}\right|_0$$

$$= \frac{\hbar}{NkT}\sum_\lambda\left[\left(\coth\frac{\hbar\omega_\lambda}{2kT}\right)^2 - 1\right]\left(\frac{\partial(\omega_\lambda^- - \omega_\lambda^+)}{\partial\eta_{1,2}}\right)_0,$$

$$(\omega_\lambda^- - \omega_\lambda^+) = \omega_0 S(1+\delta)(\eta_1 - \eta_2) - 2\omega_H.$$

Hence, as may be easily seen

$$\eta_1' = -\eta_2', \qquad \eta_1' - \eta_2' = \frac{2\eta R\hbar\omega_H / kT}{(\hbar\omega_0 S / kT)(1+\delta)\eta R + \eta^{-1}},$$

$$S = {}^1/_2, \qquad \hbar\omega_H = \mu H, \qquad \mu = \frac{ge\hbar}{2mc}, \qquad \hbar\omega_0 = 4DJ,$$

$$M_z = (\eta + \eta_1')\frac{N}{2}S\mu - (\eta + \eta_2')\frac{N}{2}S\mu$$

$$= \frac{\mu^2\eta^2 RNH}{\hbar\omega_0(1+\delta)\eta^2 R + 2kT} = V\chi_\parallel H, \qquad (43)$$

where M_Z is the total magnetic moment, V is the volume, and $\chi_\parallel$ is the susceptibility per unit volume.

At low temperatures, when $\hbar\omega_0(1+\delta)\eta^2 R \ll 2kT$,

$$\chi_\parallel = \frac{\mu^2\eta^2}{2kTV}\sum_\lambda \sinh\frac{\hbar\omega_\lambda}{2kT})^{-2}. \qquad (44)$$

For $\eta = 1$, this formula coincides with that derived in the ordinary spin-wave approximation.[21] However, even at low temperatures Eq. (43) is more accurate, since $\eta \approx \eta(0) = J_D^{-1} \neq 1$. Near the Néel point

$$R \approx [2/\eta^2(1+\delta)][k\Theta/2DJ)^2 F_a = {}^1/_2 F_a(1+\delta)\eta^2$$

[see Eq. (39)], and consequently,

$$\chi_\parallel(\Theta) = \frac{\mu^2 N}{8k(1+\delta)F_a V\Theta} = \frac{\mu^2 N}{8D(1+\delta)JV}. \qquad (45)$$

3. ANTIFERROMAGNETISM OF SMALL PARTICLES AND CHAINS. PARAMAGNETIC FLUIDS

The formulas derived in Sec. 2 permit us within certain limits to explain the dependence of the magnetization of the sublattices $M_{1,2}(T)$, the Néel temperature Θ, the susceptibility $\chi_\parallel$, and certain other quantities on the form and dimensions of the system, as well as on various parameters (J, δ, etc). However, we cannot go into detail here on this set of problems, but shall limit ourselves to a few remarks.

In the antiferromagnetic case [in distinction from a ferromagnetic system with the Hamiltonian of Eq. (1)], the magnetization of the sublattices $M_{1,2} = + NS\eta(T)/2$ depends on the form and dimensions of the spin system even at T = 0. This fact is especially clear from Eqs. (37) — (39) and (43) — (44) in the limiting case of very small particles, as is also the fact of the corresponding dependences of the quantities Θ, $\chi_\parallel$, and $\eta(T)$. The point is that in a sufficiently small system, the first term having $\lambda = 0$, which is neglected when we go over to integration, plays the predominant role in the sums. At the same time, if we retain only this first term having $\lambda = 0$, we obtain (for $\delta \ll 1$)

$$J_D = \sqrt{2}/N\sqrt{\delta}, \qquad F_a = 1/N\delta,$$

$$\Theta = DJ/kF_a = 2JN\delta/k = \mu H_{A0}N/2k, \quad \eta(0) = N\sqrt{\delta/2}, \qquad (46)$$

where

$$\delta = 2\omega_{A0}/\omega_0, \qquad \omega_0 = 4DJ/\hbar, \qquad \omega_{A0} = \mu H_{A0}/\hbar.$$

For a large three-dimensional spin system, $\eta(0) = 0.856$, and $F_a = 1.516$; hence it seems that the expressions in (46) might be applied, respectively, under the conditions $N\sqrt{\delta} \ll 1$ [for $\eta(0)$], and $N\delta \ll 1$ (for Θ). However, in fact, there is no region in which the formulas of (46) are applicable, in line with the fact that the inequalities (21) and (41) are not fulfilled. Nevertheless, it follows from (46), just as from a more detailed analysis, that $\eta(0)$ and Θ become less in small particles.* The change in the quantities $\eta(T)/\eta(0)$ and $\chi_\parallel(T)$ with decreasing dimensions of the system is even clearer. In fact, under the condition $NkT\mu H_{A_0}/(\hbar\omega_0)^2 \ll 1$, which is compatible with (41), in the temperature region in which $\hbar\omega_{\lambda=0} = \omega_0\sqrt{\delta/2}\eta \ll kT \ll k\Theta$, we may easily derive (for $N\sqrt{\delta} \gg 1$):

$$J_{DT} = 2kT/\mu H_{A0}N\eta, \qquad \eta(T) = \eta(0)(1 - 2kT/\mu H_{A0}N), \qquad (47)$$

$$\chi_\parallel(T) = (2\mu^2/VkT)(kT/\hbar\sqrt{\omega_0\omega_{A0}})^2. \qquad (48)$$

Thus, within a certain definite region, the difference $1 - \eta(T)/\eta(0)$ and the susceptibility $\chi_\parallel$ differ considerably from the values given by the expressions

$$1 - \eta(T)/\eta(0) \sim (kT/\hbar\omega_0)^2 \text{ and } \chi_\parallel \sim (\mu^2 N/kTV)(kT/\hbar\omega_0)^3,$$

applicable to large systems.[22] The result (45) is also of interest; according to it the value of $\chi_\parallel$ at the transition point for a given J is independent of Θ (for $\delta \ll 1$). Hence it is clear that in the general case in the region $T < \Theta$ (for a given temperature T and for constant values of the other parameters), the susceptibility increases with the decrease in Θ which takes place in small particles, films, and fibers.

---

*Naturally, as the dimensions of the system are decreased, the transition becomes less sharp, and strictly speaking, we cannot speak then of a phase transition. In practice, when $N \gg 1$ this fact is not essential.

Thus, the properties of antiferromagnetic finely-dispersed substances, polymer chains and structures may be appreciably different from those of the ordinarily studied antiferromagnetic substances (above all, it would be expedient to compare the properties of one and the same substance as depending on the dimensions and form of the crystals).

In the region in which the limiting formulas (47) and (48) are valid, the difference between antiferromagnetic specimens of differing forms is obliterated. This is understandable, since the role of the form decreases as all dimensions are decreased. In addition, for sufficiently thin films and fibers, the differences from the three-dimensional case may be very great, since long-range order is generally absent in an infinite chain, as it is also in a two-dimensional system with $\delta = 0$. In fact, according to Eqs. (37) — (39), for an infinite chain, $\eta(0) \approx \pi/|\ln \delta|$, and $\Theta = J\sqrt{2\delta}/k$. However, in this case the approximation itself is not valid, in connection with the increase in fluctuations (see the analogous estimate for ferromagnetic substances at the end of Sec. 1).

However, for finite chains, especially if they are short enough, antiferromagnetic ordering may take place. It is sufficient to say that we cannot make a strict distinction between long-range and short-range order in a finite system. However, with regard to short-range antiferromagnetic order, the latter is observed in a series of substances (e. g., by neutron scattering) even at temperatures considerably above the Néel point. In this regard, we may also refer to the calculation of the short-range order in an infinite chain.[27,28] On the other hand, as has been mentioned, our formulas are inapplicable to sufficiently small particles, as well as to antiferromagnetic systems above the Néel point. This is associated with the use, along with the other magnetic moments, of the anisotropy field $H_A = H_{A_0}\eta$, which vanishes when the mean magnetization of the sublattice equals zero. The fact is that the appearance of short-range order is also associated with a certain anisotropy field H'_A, which apparently is of the order of H_{A_0} for the given small group of spins.

The region of very small particles and chains, as well as the paramagnetism of those systems having thermal transitions $\Theta \ll \Theta_0 \sim J/k$, is nevertheless very interesting. Hence, in spite of the fact that we have no pertinent quantitative results, we shall consider this problem too.

Ordinarily, $\Theta \sim \Theta_0 \sim J/k$, and correspondingly, when $\Theta \ll \Theta_0$ (in particular, when $\Theta = 0$), we may expect that the formula $\chi \sim \text{const}/(T + \Theta)$ will not be valid over the wide temperature range $\Theta < T < \Theta_0 \sim J/k$. (A law of the type $\chi \sim \text{const}/(T + \Theta)$ will probably hold true for $T \gtrsim \Theta_0$.) In the temperature range $\Theta < T \gtrsim \Theta_0$ (this range has no sharp upper boundary), the substance may be called either an "incoherent antiferromagnetic substance" or a "paramagnetic fluid" with antiferromagnetic interaction (the analogous concepts for the ferromagnetic case have already been taken up at the end of Sec. 1; in connection with this subject, the experimental data of Ryan, Pugh, and Smoluchowski[29] may be of interest). As in the case of other fluids, it seems impossible, in general, to construct a consistent and sufficiently exact theory for such a state. Individual special cases may be exceptions (a long chain at low temperatures, etc).* We hope yet to take up this problem in its various aspects, but shall mention at present only the following.

One of the possibilities for a qualitative understanding is the comparison of the properties of fluids with those of a very highly-disperse crystalline medium (the quasi-crystalline model). Individual "crystallites" of such a medium contain a small number of spins each. According to the hypothesis, these crystallites retain their high degree of order, i.e., within them $\eta \sim 1$ (or, say, $\eta \sim 0.1$) and $H'_A = H_{A_0}$. Then, as might be supposed, the lowest excited state of the "crystallite" has an energy of $\hbar\omega_{\lambda=0}$, where

$$\omega_{\lambda=0} = \eta\omega_0 S \sqrt{2\delta} \pm \omega_H \sim \sqrt{\omega_0 \omega_{A0}} \pm \omega_H$$

[see Eq. (29a)]. The susceptibility of such a system is determined by Eq. (48), which had also been derived by us previously.[2] Here, in the region $\hbar\sqrt{\omega_0\omega_{A_0}}/k \ll T \ll \Theta$, the susceptibility increases in proportion to T^2. Then, probably, χ goes through a gentle maximum at $T \sim \Theta_0 \sim J/k$, and beyond this, decreases according to a law of the type $1/(T + \Theta_0)$. We note also that χ depends on H even in fields of the order of $\hbar\sqrt{\omega_0\omega_{A_0}}/\mu$. Such a model is very crude, and the corresponding estimate of χ is not very convincing, but we should expect just this sort of dependence $\chi(T, H)$ in qualitative terms for a "paramagnetic fluid" of the antiferromagnetic type.

In view of what has been said, the attempt which we made[2] to explain the experimental data of Blyumenfel'd et al.[1] retains its content, but refers

---

*We are familiar only with the result for the Ising model.[19,30] In this case, $\chi_{\parallel}$ has a gentle maximum at $kT \sim J$.

now to the region of "incoherent antiferromagnetism." Besides, within the framework of the model which we have studied, the very boundary between the properties of a very finely-dispersed antiferromagnetic system and those of a paramagnetic fluid of the antiferromagnetic type is obliterated to a considerable degree. In particular, internal magnetic fields must be observable (e. g., by the nuclear magnetic resonance method) in "paramagnetic fluids" also, while a strong broadening of the paramagnetic resonance lines will take place as well.

The theoretical and experimental study of "paramagnetic fluids" is of great interest. However, it was possible above, of course, only to pose questions and give some qualitative conceptions which will be necessary in the subsequent analysis. In this regard, it may not be superfluous to emphasize in conclusion that the basic content of this article has not been connected at all with the concept of "paramagnetic fluids", since it has involved the region below the transition temperature.

---

[1] Blyumenfel'd, Kalmanson, and Sheng, Dokl. Akad. Nauk SSSR **124**, 1144 (1959), Soviet Phys.-Doklady **4**, 260 (1959). L. A. Blyumenfel'd, Биофизика (Biophysics) **4**, 515 (1959).

[2] V. L. Ginzburg and V. M. Faĭn, Dokl. Akad SSSR **131**, 785 (1960), Soviet Phys.-Doklady **5**, 328 (1960).

[3] J. Van Kranendonk and J. H. Van Vleck, Revs. Modern Phys. **30**, 1 (1958).

[4] E. M. Lifshitz, JETP **15**, 97 (1945).

[5] C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).

[6] M. Kaganov and V. Tsukernik, JETP **34**, 106 (1958), Soviet Phys. JETP **7**, 73 (1958).

[7] F. J. Dyson, Phys. Rev. **102**, 1217 (1956).

[8] V. L. Ginzburg, Физика твердого тела **2**, 2031 (1960), Soviet Phys.-Solid State **2** in press.

[9] V. Zehler, Z. Naturforsch. **5a**, 344 (1950).

[10] G. S. Rushbrooke and P. J. Wood, Proc. Phys. Soc. **A68**, 1161 (1955).

[11] C. Domb and M. F. Sykes, Proc. Roy. Soc. (London) **A240**, 214 (1957).

[12] G. Heber, Z. Naturforsch. **9a**, 91 (1954).

[13] M. R. Schlafroth, Proc. Phys. Soc. **A67**, 33 (1954).

[14] N. N. Bogolyubov and S. V. Tyablikov, Dokl. Akad. Nauk SSSR **126**, 53 (1959), Soviet Phys.-Doklady **4**, 589 (1959).

[15] S. V. Tyablikov, Ukr. Mat. Zhur. **11**, 287 (1959).

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

[17] C. A. Neugebauer, Phys. Rev. **116**, 1441 (1959).

[18] L. D. Landau and E. M. Lifshitz, Статистическая физика (Statistical Physics), Sec. 144, Gostekhizdat (1951); [Engl. transl. Addison-Wesley, 1958].

[19] G. F. Newell and E. W. Montroll, Revs. Modern Phys. **25**, 353 (1953).

[20] P. W. Anderson, Phys. Rev. **86**, 694 (1952).

[21] J. M. Ziman, Proc. Phys. Soc. **A65**, 540, 548 (1952).

[22] R. Kubo, Phys. Rev. **87**, 568 (1952).

[23] Nagamiya, Yosida, and Kubo, Adv. in Phys. **4**, 1 (1955).

[24] Yin Yuan-li, Phys. Rev. **84**, 721 (1951).

[25] T. Oguchi, Phys. Rev. **117**, 117 (1960).

[26] P'u Fu-C'ho, Dokl. Akad. Nauk SSSR **130**, 1244 and **131**, 546 (1960), Soviet Phys.-Doklady **5**, 128 (1960) and **5**, 321 (1960).

[27] R. Orbach, Phys. Rev. **112**, 309 (1958).

[28] L. R. Walker, Phys. Rev. **116**, 1089 (1959).

[29] Ryan, Pugh, and Smoluchowski, Phys. Rev. **116**, 1106 (1959).

[30] C. Domb, Adv. in Physics, **9**, 150 (1960).

Translated by M. V. King
244