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## Macroscopic theory of spin waves

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A description of magnetic structures by means of macroscopic multipole moments is presented. All types of macroscopically distinct exchange magnetic structures are found for ferromagnets or collinear ferrimagnets (32 types), antiferromagnets (230 types), and noncollinear ferrimagnets (79 types). The general form of the equations that describe long-wave, low-frequency spin waves is elucidated. The examples considered show that in many cases there should exist anomalous branches of the spin oscillations.

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The theory of spin waves is well developed for the case of ferromagnets. On the one hand, there is a microscopic approach<sup>[1,2]</sup> that is based on the Heisenberg model and that enables us to explain the structure of the ground state of a ferromagnet and the spin-wave spectrum over the whole frequency range. On the other hand, the low-frequency, long-wave spin waves can be described macroscopically by means of the Landau-Lifshitz equation.<sup>[3]</sup>

There is a completely different situation in the case of antiferromagnets. Here the microscopic problem can be solved only for spins that are large (i.e., quasi-classical), a property that they in fact, at least in the majority of cases, do not have. The existing phenomenological theory<sup>[4,5]</sup> of low-frequency, long-wave spin-waves leads in many cases to satisfactory results. But this theory is essentially of model type, since the basic quantities of the theory (the sublattice moments) are not macroscopic. They cannot be obtained by macroscopic averaging of any physical quantities.

The present paper proposes a general macroscopic approach to the investigation of low-frequency, long-wave spin waves in any magnetic materials in which the magnetic ordering is the result of the action of exchange forces that appreciably exceed the relativistic interactions. The proposed approach uses no model-type concepts about the state of the magnetic material (localized spins, sublattices, etc.), but is essentially based only on symmetry considerations. The basic

quantities are macroscopic multipole moments, which are obtained from the true microscopic magnetic-moment density by a definite averaging over physically infinitesimal volumes.

The formulation of the macroscopic equations for spin waves is based to a significant degree on macroscopic analysis of the possible types of symmetry of exchange forces in magnetic materials. By means of multipole moments it proves possible to find all the types (their total number, as we shall see, is 373) of macroscopically different magnetic structures.

### 1. MACROSCOPIC MULTIPOLE MOMENTS

We shall first introduce the macroscopic multipole moments for the simplest case, in which they describe some scalar microscopic quantity. Let  $\mathcal{G}$  be the space group of the symmetry of the equilibrium state of the crystal: that is, the group of transformations with respect to which the equilibrium microscopic charge density is invariant. We consider some (in general nonequilibrium) state, described microscopically by a scalar quantity  $f(\mathbf{r})$  (this may be, for example, the deviation of the charge density from its equilibrium value). The problem consists in finding the set of macroscopic quantities that describe this state as fully as is possible.

Any macroscopic quantity  $F(\mathbf{r})$  must be obtained from  $f(\mathbf{r})$  as a result of averaging over macroscopic volumes.

In general this operation can be represented as follows:

$$F(\mathbf{r}) = \int f(\mathbf{r}+\boldsymbol{\rho}) \Phi(\boldsymbol{\rho}) d^3\rho. \quad (1)$$

Here the integration extends over the whole volume;  $\Phi(\mathbf{r})$  is some function that decreases sufficiently rapidly at infinity and that varies appreciably over distances large in comparison with the interatomic distance, but small in comparison with a characteristic length parameter of the macroscopic problem (wavelength etc.) In other respects, the function  $\Phi(\mathbf{r})$  is arbitrary. In order to obtain a macroscopic quantity of general form, it is necessary to use for  $\Phi(\mathbf{r})$  a function that is also of sufficiently general form.

We designate by the symbol  $\mathcal{G}$  an arbitrary transformation of the group  $\mathcal{G}$ , and we consider the state corresponding to the function  $f'(\mathbf{r}) = f(\mathcal{G}^{-1}\mathbf{r})$ . It is clear that any equation describing the properties of the state  $f(\mathbf{r})$  must be invariant with respect to the transformation  $f(\mathbf{r}) \rightarrow f'(\mathbf{r})$ . Any macroscopic equation must therefore be invariant with respect to the transformation  $F(\mathbf{r}) \rightarrow F'(\mathbf{r})$ , where  $F'(\mathbf{r})$  is obtained by substitution of the function  $f'(\mathbf{r})$  for  $f(\mathbf{r})$  in formula (1):

$$F'(\mathbf{r}) = \int f(\mathcal{G}^{-1}(\mathbf{r}+\boldsymbol{\rho})) \Phi(\boldsymbol{\rho}) d^3\rho. \quad (2)$$

Let  $G$  be the group of transformations corresponding to the crystal class. An arbitrary element  $G$  of this group by itself does not, in general, belong to the group  $\mathcal{G}$ . But there always exists a vector  $\mathbf{a}$ , with a length of the order of the interatomic distance, translation along which, in combination with the transformation  $G$ , produces one of the transformations  $\mathcal{G}$ . On substituting this in the expression for  $F'(G\mathbf{r} + \mathbf{a}) \approx F'(G\mathbf{r})$  and making the change of variables  $G^{-1}\boldsymbol{\rho} \rightarrow \boldsymbol{\rho}$  in the integral, we obtain

$$F'(G\mathbf{r}) = \int f(\mathbf{r}+\boldsymbol{\rho}) \Phi(G\boldsymbol{\rho}) d^3\rho. \quad (3)$$

The set of functions  $\Phi(G\boldsymbol{\rho})$  with all possible  $G$  produces the so-called regular representation of the group  $G$ . When expanded in irreducible representations, which corresponds to representation of the function  $\Phi$  in the form of a sum

$$\Phi(\mathbf{r}) = \sum_n \sum_\alpha a_n^\alpha \Phi_n^\alpha(\mathbf{r}), \quad (4)$$

it contains all the irreducible representations, without exception, of the group  $G$  (see [6]). In formula (4) the index  $n$  enumerates the various irreducible representations; the index  $\alpha$  enumerates the functions  $\Phi_n^\alpha$  that produce a given irreducible representation. Substitution of (4) in (1) shows that an arbitrary macroscopic quantity is a linear combination of macroscopic quantities  $F_n^\alpha(\mathbf{r})$  that are determined by the relation

$$F_n^\alpha(\mathbf{r}) = \int f(\mathbf{r}+\boldsymbol{\rho}) \Phi_n^\alpha(\boldsymbol{\rho}) d^3\rho \quad (5)$$

and that, by virtue of (3), transform under the action of transformations of crystal class  $G$  as follows:

$$F_n^{\alpha'}(G\mathbf{r}) = G_n^{\alpha\beta} F_n^\beta(\mathbf{r}), \quad (6)$$

where  $G_n^{\alpha\beta}$  is the matrix of the transformation  $G$  in the  $n$ th irreducible representation. An arbitrary macroscopic equation must be invariant with respect to the transformation (6). In an isotropic medium (fluid), an expansion of the form (4) is an expansion in spherical functions  $Y_l^m(\mathbf{r}/|\mathbf{r}|)$ . The corresponding macroscopic quantities  $F_l^m(\mathbf{r})$  are a generalization to the case of an infinite medium of the usual 2<sup>l</sup>-pole moments [7] that describe a finite system, and they have the meaning of densities of the latter. They were treated by Frenkel [8] from a somewhat different point of view. For crystals, an analogous role is played by the multipole moments  $F_n^\alpha(\mathbf{r})$ . They essentially coincide with the quantities considered in Landau's theory of phase transitions of the second kind, in the capacity of order parameters, in the case in which the transition occurs without change of the elementary cell.

The choice of the macroscopic moments is not unique, because of the arbitrariness of the function  $\Phi(\mathbf{r})$ . But since their law of transformation is independent of the choice of  $\Phi(\mathbf{r})$ , this nonuniqueness is not reflected in any physical consequences. It is an expression of the well-known nonuniqueness of the ordinary multipole moments, which is due to the arbitrariness of choice of the origin of coordinates.

We now consider a magnetically ordered crystal. Let  $\mathbf{m}(\mathbf{r})$  be the microscopic magnetic-moment density. The symmetry of the system is determined by one of the magnetic space groups. These groups contain, as their elements, the purely spatial transformations (rotations, reflections, translations) in combination with the time-reflection transformation  $R$ . This characterization of the symmetry is exact; but for this very reason, in its important approximate symmetry properties are completely lost. In fact, in the properties of magnetic materials a basic role is played by exchange forces, whose symmetry is higher than the symmetry of the weak relativistic interactions. To indicate only the exact symmetry is to lose entirely the information about the much higher symmetry of the exchange forces.

We shall neglect all relativistic interactions and consider the exchange symmetry of the magnetic material. Since the exchange forces depend only on the relative orientations of the spins, in this case, besides the transformation  $R$ , there is permitted an infinite set of new symmetry transformations, consisting of all rotations of the spin space  $U$  (that is, rotations of all the spins through the same angle). In this situation, the general orientation of the spins with respect to the crystallographic axes becomes nominal, and we may consider that the components of the magnetic moment  $m_i(\mathbf{r})$  behave like scalars in all purely spatial transformations, and transform like components of a vector only in rotations of the spin space. The transformation  $R$  plays the role of an inversion of the spin space. The exchange symmetry is determined by prescription of one of the exchange space groups, which contain all those combinations of purely spatial transformations, rotations of the spin space  $U$ , and the transformation  $R$  with respect to which the values of  $m_i(\mathbf{r})$  are invariant. These groups have at present not yet been classified.

The charge-density symmetry group  $\mathcal{G}$  is obtained from the exchange space group by formal identification of the elements  $U$  and  $R$  with the identity transformation. This is the symmetry group generally of all spin scalars in the system.

By the same method by which this was done above for scalars, we introduce macroscopic quantities  $M_{in}^\alpha(\mathbf{r})$  corresponding to each of the components of the magnetic moment  $m_i(\mathbf{r})$ . These quantities differ from  $F_n^\alpha$  in the following respect. Since the function  $f(\mathbf{r})$  in the equilibrium state is invariant with respect to the group  $\mathcal{G}$ , all  $F_n^\alpha$  except that one of them that transforms according to the identity representation of the group  $G$  necessarily vanish in equilibrium. But the magnetic moment  $m_i(\mathbf{r})$  in general is by no means invariant with respect to  $\mathcal{G}$ , even in the equilibrium state. Therefore any one of the quantities  $M_{in}^\alpha$  may have a finite value in equilibrium. In the case in which the transition from a magnetically ordered state to a paramagnetic occurs without change of the elementary cell, the quantities  $M_{in}^\alpha$  essentially coincide with the spin densities introduced by Dzyaloshinskii<sup>[9]</sup> (see also<sup>[10]</sup>).

Prescription of the functions  $M_{in}^\alpha(\mathbf{r})$  gives a complete macroscopic description of the magnetic structure of the body. With their aid it is possible to carry out a classification of all macroscopically different magnetic structures; that is, to determine all the exchange classes, which have the same relation to the exchange space groups that the magnetic classes have to the magnetic space groups. Before proceeding to the solution of this problem, we note the following. The macroscopic moment  $\mathbf{M}_n^\alpha = \{M_{in}^\alpha\}$  that transforms according to the identity representation (we denote it by  $\mathbf{M}^0$ ) is obviously proportional to the usual magnetization of the body and can be so normalized as in general to coincide with it. All the remaining moments give no contribution to the magnetization; therefore they are antiferromagnetic moments. Thus if  $\mathbf{M}^0 \neq 0$ , then the body is a noncollinear ferrimagnet or a ferromagnet, depending on whether there are or are not other nonvanishing moments. (Strictly speaking, in the latter case the body may be a collinear ferrimagnet. But such a ferrimagnet, at least in the sense of macroscopic properties, does not differ from a ferromagnet.) If, however,  $\mathbf{M}^0 = 0$  but there are other moments, then the body is an antiferromagnet. Finally, if all the moments vanish, the body has no magnetic structure at all.

## 2. EXCHANGE CLASSES

We shall elucidate the conditions that must be satisfied by the vectors  $\mathbf{M}_n^\alpha$  in the equilibrium state. We consider the scalar products  $\mathbf{M}_n^\alpha \mathbf{M}_m^\beta$ . They are spin scalars and therefore must be invariant with respect to the group  $G$ . On the other hand, these quantities transform according to the direct product of irreducible representations  $n$  and  $m$ ; therefore they must have the following form:

$$\mathbf{M}_n^\alpha \mathbf{M}_m^\beta = c_n \delta_{nm} \delta_{\alpha\beta},$$

where  $c_n$  is a certain constant. Hence it follows that

different  $\mathbf{M}_n^\alpha$  are perpendicular to each other; and those among them that correspond to a single multidimensional representation have the same length. The maximum number of nonvanishing moments  $\mathbf{M}_n^\alpha$  is consequently equal to three. Thus only the following combinations of irreducible representations can occur: 1) One one-dimensional representation; that is, there is only one nonvanishing moment corresponding to this representation. 2) Two one-dimensional representations or one two-dimensional; here there are two nonvanishing moments, perpendicular to each other, and in the latter case they are equal in absolute value. 3) Three one-dimensional, or a one-dimensional and one two-dimensional, or finally one three-dimensional representation; here there are three nonvanishing moments, they are mutually perpendicular, and in the second case two of them, in the third all three moments are equal in absolute value. By picking out all the combinations of this type in each of the 32 crystal classes  $G$  we obtain all the macroscopically distinct types of magnetic structures; that is, all the exchange classes.

Not all these structures, however, can actually occur. Many of them do not satisfy the following stability criterion, analogous to the well-known criterion in the theory of second-order phase transitions.<sup>[11]</sup> We denote by  $\mathbf{m}_n^\alpha$  the deviations of the macroscopic moments from the equilibrium values, and we assume that there exists an expression, invariant with respect to the group  $G$ , of the form

$$K_\mu^{\alpha\beta} \left( m_n^\alpha \frac{\partial m_m^\beta}{\partial x_\mu} - m_m^\beta \frac{\partial m_n^\alpha}{\partial x_\mu} \right), \quad (7)$$

where  $x_\mu$  are the spatial coordinates. If the moments corresponding to both representations  $n$  and  $m$  are nonzero in equilibrium, then such a structure is unstable. In fact, consider a deviation from equilibrium of the form

$$\mathbf{m}_n^\alpha = [\omega(\mathbf{r}) \mathbf{M}_n^\alpha] \quad (8)$$

for all  $n$  and  $\alpha$ , where  $\mathbf{M}_n^\alpha$  are the equilibrium values of the moments and where  $\omega(\mathbf{r})$  is a slowly varying function of the coordinates. Since at each point of space such a deviation reduces to a rotation of all the moments through the same angle, the local (that is, not containing spatial derivatives) part of the deviation  $\delta E$  of the energy from the equilibrium value vanishes. The principal part of  $\delta E$  is therefore determined by terms of the form (7), linear in the derivatives:

$$\delta E = \sum K_\mu^{\alpha\beta} \left( m_n^\alpha \frac{\partial m_m^\beta}{\partial x_\mu} - m_m^\beta \frac{\partial m_n^\alpha}{\partial x_\mu} \right), \quad (9)$$

where the summation sign means summation over all invariants of the form (7). On substituting from this in (8), we obtain, after simple transformations with allowance for the mutual perpendicularity of the equilibrium moments,

$$\delta E = \sum K_\mu^{\alpha\beta} \frac{\partial \omega}{\partial x_\mu} [\omega [\mathbf{M}_n^\beta \mathbf{M}_n^\alpha]].$$

It is obvious that the expression written is always

capable of taking negative values. Thus a necessary condition for stability of the structure is absence of invariants of the form (7) for all pairs of indices  $n, m$  for which the equilibrium moments are nonzero.

We consider for example the crystal class  $D_3$ . The group  $D_3$  has three irreducible representations: the identity representation  $A_1$  (we denote the corresponding moment by  $M^0$ ); a one-dimensional representation  $A_2$  according to which the coordinate  $z$  transforms (we denote the corresponding moment by  $W$ ); and a two-dimensional  $E$  according to which the coordinates  $x$  and  $y$  transform (we denote the corresponding moments by  $U$  and  $V$ ). Possible combinations of the representations are:  $A_1, A_2, (A_1 A_2), E, (EA_1), (EA_2)$ . The first of these produces a ferromagnetic structure, the third and fifth noncollinear ferrimagnetic, the rest antiferromagnetic. In this case there are four invariants of the form (7):

$$\begin{aligned} & U \frac{\partial V}{\partial z} - V \frac{\partial U}{\partial z}, \quad W \frac{\partial U}{\partial y} - U \frac{\partial W}{\partial y} - W \frac{\partial V}{\partial x} + V \frac{\partial W}{\partial x}, \\ & M^0 \frac{\partial U}{\partial x} - U \frac{\partial M^0}{\partial x} + M^0 \frac{\partial V}{\partial y} - V \frac{\partial M^0}{\partial y}, \quad M^0 \frac{\partial W}{\partial z} - W \frac{\partial M^0}{\partial z}. \end{aligned} \quad (10)$$

The first of these causes instability of all structures containing the two-dimensional representation  $E$ ; the last, of the structure  $(A_1 A_2)$ . Thus in group  $D_3$  there are in all 6 possible structures from the symmetry point of view. But only the following 2 of them are stable:  $A_1, A_2$ .

The Appendix presents the results of a similar analysis of all 32 crystal classes. The first number after the class symbol means the total number of structures possible from the symmetry point of view; the second, the number of stable structures. All the stable structures are then actually enumerated. The symbols for the representations coincide with those adopted in the book of Landau and Lifshitz.<sup>[6]</sup> It must be kept in mind that the representations  $B_1, B_2, B_3$  in class  $D_2$ ;  $B_{1g}, B_{2g}, B_{3g}$  and  $B_{1u}, B_{2u}, B_{3u}$  in class  $D_{2h}$ ;  $B_1, B_2$  in classes  $C_{2v}, C_{4v}, D_4, D_6, C_{6v}$ ;  $B_{1g}, B_{2g}$  and  $B_{1u}, B_{2u}$  in classes  $D_{4h}$  and  $D_{6h}$  are equivalent in the sense that they convert to each other on transformation of the system of coordinates. Replacement of these representations by each other leads to structures equivalent from the symmetry point of view.

In addition, there are still 32 trivial exchange classes, corresponding to crystals without magnetic structure. Thus the total number of macroscopically distinct magnetic structures possible from symmetry considerations is 561. Of these, only the 373 structures enumerated above are stable. Among them are 32 paramagnetic, 32 ferromagnetic, 79 noncollinear-ferrimagnetic, and 230 antiferromagnetic. It cannot be asserted, however, that the unstable structures have no meaning at all. If for some reason the coefficients  $K_{\mu}^{\alpha\beta}$  in formula (9) are anomalously small, then such structures may occur as helicoidal structures (see<sup>[9]</sup>). Each of the 561 structures determines a certain group of transformations (exchange class), consisting of all products of elements of group  $G$ , spin rotations, and the transformation  $R$  with respect to which the given

structure is invariant.

Thus there are in all 561 exchange classes. Each of the exchange structures, if we orient the moments in a definite manner with respect to the crystallographic axes and exclude relativistic interactions, will be characterized by a certain exact symmetry, determined by one of the magnetic classes. The problem arises of determining those magnetic classes by which each of the exchange classes can be characterized when allowance is made for relativistic interactions. We shall not concern ourselves here with this question, but shall restrict ourselves to consideration solely of those properties that are determined by the exchange symmetry.

We note that a formal generalization of the usual magnetic groups, by inclusion in the set of transformations, along with the element  $R$ , of arbitrary spin rotations (but without relation to the exchange approximation), was carried out earlier by a number of authors.<sup>[12-14]</sup> Here, in essence, the topic was a certain accidental symmetry, whose appearance can be caused, for example (see<sup>[13]</sup>), by the properties of one or another model Hamiltonian even with allowance for relativistic interactions.

### 3. GENERAL EQUATIONS FOR SPIN WAVES

Long-wave, low-frequency spin waves can be described macroscopically on the basis of equations of motion for the moments. We introduce the effective fields

$$h^{\alpha}(\mathbf{r}) = -\delta E / \delta \mathbf{m}^{\alpha}(\mathbf{r}), \quad (11)$$

determined by the variational derivatives of the total energy with respect to the deviations  $\mathbf{m}^{\alpha}(\mathbf{r})$  of the moments from equilibrium. Here and hereafter we shall number the moments by a single index  $\alpha$ , so that several moments with "neighboring" indices  $\alpha$  may belong to a single multidimensional representation. In the equilibrium state, all  $h^{\alpha}$  are zero. For small deviations from equilibrium, the time derivatives of  $\mathbf{m}^{\alpha}$  must be linear combinations of the effective fields:

$$\dot{m}_i^{\alpha} = g_{ik}^{\alpha\beta} h_k^{\beta} + g_{i\mu}^{\alpha\beta} \frac{\partial}{\partial x_{\mu}} h_k^{\beta} + g_{i\mu\nu}^{\alpha\beta} \frac{\partial^2}{\partial x_{\mu} \partial x_{\nu}} h_k^{\beta} + \dots, \quad (12)$$

where  $i, k, \dots$  are vector spin indices. Since we are concerned with low-frequency oscillations, we have restricted ourselves to consideration solely of the first time derivatives. The quantities  $g_{ik}^{\alpha\beta}$  must satisfy the usual conditions that follow from invariance for  $t \rightarrow -t$  and conservation of energy (we everywhere neglect dissipation). Invariance with respect to  $t \rightarrow -t$  requires that all the coefficients  $g_{ik}^{\alpha\beta}$  must change sign under the action of  $R$ . For conservation of energy, that is vanishing of the derivative  $\dot{E}$  calculated by means of (11) and (12), it is necessary that the coefficients  $g_{ik}^{\alpha\beta}$  satisfy the following conditions:

$$g_{ik}^{\alpha\beta} = -g_{ki}^{\beta\alpha}, \quad g_{i\mu}^{\alpha\beta} = g_{\mu i}^{\beta\alpha}, \quad g_{i\mu\nu}^{\alpha\beta} = -g_{i\mu\nu}^{\beta\alpha}, \dots, \quad (13)$$

that is, the coefficients of even derivatives are antisymmetric, and of odd derivatives symmetric, with

respect to simultaneous commutation of the indices  $\alpha i$  and  $\beta k$ .

Since in the exchange approximation the spin space is isotropic, the coefficients  $g_{ik}^{\alpha\beta}$  in their dependence on the equilibrium moments can contain only terms proportional to the following combinations of moments:

$$\epsilon_{ikl} M_l^\alpha, [\mathbf{M}^\alpha \times \mathbf{M}^\beta]_i M_k^\gamma, \delta_{ik} (\mathbf{M}^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\beta]),$$

which are spin tensors of the second rank, changing sign under the action of the transformation  $R$ . We have

$$g_{ik}^{\alpha\beta} = \gamma^{\alpha\beta\gamma} \epsilon_{ikl} M_l^\gamma + \gamma^{\alpha\beta\gamma\rho} M_i^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\rho]_k - \gamma^{\beta\alpha\gamma\rho} M_k^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\rho]_i + \delta_{ik} \tilde{\gamma}^{\alpha\beta\gamma\rho} (\mathbf{M}^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\rho]),$$

where the quantities  $\gamma^{\dots}$  and  $\tilde{\gamma}^{\dots}$  are spin scalars, which in transformations of the group  $G$  transform like "tensors," i.e., like products of functions  $\Phi^\alpha \Phi^\beta \Phi^\gamma \dots$ , but are invariant tensors, i.e., do not change their form under the action of the group  $G$ . From the conditions (13) it follows that

$$\gamma^{\alpha\beta\gamma} = \gamma^{\beta\alpha\gamma}, \quad \tilde{\gamma}^{\alpha\beta\gamma\rho} = -\tilde{\gamma}^{\beta\alpha\gamma\rho}.$$

The quantities  $g_{ik}^{\alpha\beta}, \dots$  can be written in similar fashion. As a result the general equations (12), after simple transformations, acquire the following form:

$$\dot{m}_i^\alpha = \Gamma^{\alpha\beta\gamma} [\mathbf{h}^\beta \times \mathbf{M}^\gamma]_i + \Gamma_1^{\alpha\beta\gamma\rho} M_i^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\rho]_k h_k^\beta - \Gamma_2^{\beta\alpha\gamma\rho} M_k^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\rho]_i h_k^\beta + \tilde{\Gamma}^{\alpha\beta\gamma\rho} (\mathbf{M}^\gamma [\mathbf{M}^\alpha \times \mathbf{M}^\rho]) h_i^\beta, \quad (14)$$

where

$$\begin{aligned} \Gamma^{\alpha\beta\gamma} &= \gamma^{\alpha\beta\gamma} + \gamma_i^{\alpha\beta\gamma} \nabla^i + \gamma_2^{\alpha\beta\gamma} \Delta^i, \\ \Gamma_1^{\alpha\beta\gamma\rho} &= \gamma^{\alpha\beta\gamma\rho} + \gamma_i^{\alpha\beta\gamma\rho} \nabla_i + \gamma_2^{\alpha\beta\gamma\rho} \Delta_i, \\ \Gamma_2^{\alpha\beta\gamma\rho} &= \gamma_i^{\alpha\beta\gamma\rho} - \gamma_i^{\beta\alpha\gamma\rho} \nabla_i + \gamma_2^{\alpha\beta\gamma\rho} \Delta_i, \\ \tilde{\Gamma}^{\alpha\beta\gamma\rho} &= \tilde{\gamma}^{\alpha\beta\gamma\rho} + \tilde{\gamma}_i^{\alpha\beta\gamma\rho} \tilde{\nabla}_i + \tilde{\gamma}_2^{\alpha\beta\gamma\rho} \tilde{\Delta}_i; \end{aligned}$$

$\nabla^\alpha, \nabla_{1/2}^\alpha, \tilde{\nabla}^\alpha$  are general linear combinations of all spatial derivatives of odd order, transforming like  $\Phi^\alpha$ ;  $\Delta^\alpha, \Delta_{1/2}^\alpha, \tilde{\Delta}^\alpha$  are combinations, of the same type, of derivatives of even order; the quantities  $\gamma$  are certain invariant tensors, and

$$\gamma_i^{\alpha\beta\gamma} = -\gamma_i^{\beta\alpha\gamma}, \quad \gamma_2^{\alpha\beta\gamma} = \gamma_2^{\beta\alpha\gamma}, \quad \tilde{\gamma}_i^{\alpha\beta\gamma\rho} = \tilde{\gamma}_i^{\beta\alpha\gamma\rho}, \quad \tilde{\gamma}_2^{\alpha\beta\gamma\rho} = -\tilde{\gamma}_2^{\beta\alpha\gamma\rho}.$$

The general equations (14) simplify considerably if, in equilibrium, only one of the moments is nonzero. In this case the last three terms vanish:

$$\dot{\mathbf{m}}^\alpha = \Gamma^{\alpha\beta\gamma} [\mathbf{h}^\beta \times \mathbf{M}^\gamma]. \quad (15)$$

If, in equilibrium, two moments are nonzero, then the last term in (14) vanishes. Let the indices  $a, b, c, \dots$  run through only those two values of  $\alpha$  that correspond to nonvanishing equilibrium moments. Then

$$[\mathbf{M}^a \times \mathbf{M}^b] = |\mathbf{M}^a| |\mathbf{M}^b| \nu \epsilon^{ab},$$

where  $\nu$  is a unit vector normal to the plane in which the equilibrium moments lie, and where  $\epsilon^{ab}$  is the two-

dimensional antisymmetric unit "tensor." The equations for structures with two equilibrium moments take the following form:

$$\dot{\mathbf{m}}^\alpha = \Gamma^{\alpha\beta\gamma} [\mathbf{h}^\beta \times \mathbf{M}^\gamma] + \hat{\Gamma}_1^{\alpha\beta\sigma} \mathbf{M}^\alpha (\mathbf{h}^\beta \nu) - \hat{\Gamma}_2^{\alpha\sigma} \nu [\mathbf{M}^\alpha \times \mathbf{h}^\beta], \quad (16)$$

where

$$\hat{\Gamma}_{1,2}^{\alpha\beta\sigma} = \Pi \Gamma_{1,2}^{\alpha\beta\sigma c} \epsilon^{bc},$$

$\Pi$  is the product of the moduli of the equilibrium moments.

Apropos of equations (14)–(16) it is necessary to make the following comment. In their derivation, an important assumption was that we are concerned with low-frequency oscillations. But a literal application of the equations leads in general to the appearance of an optical branch, i.e., of high exchange frequencies that do not vanish on approach of the wave vector to zero. This means that the procedure for using the equations written above must be as follows. First we neglect all spatial derivatives and find the characteristic frequencies. Those frequencies that are found to be finite in the zeroth approximation are high frequencies, which we have no right to consider. But it can be asserted that linear combinations of the moments  $\mathbf{m}^\alpha$  corresponding to these frequencies, if they produce any, produce only high-frequency oscillations. A certain number of frequencies vanish in the zeroth approximation. To calculate them we must go to the next approximation, i.e., take into account the principal terms with spatial derivatives. Here it is possible simply to set the high-frequency linear combinations of moments equal to zero, i.e., to "freeze" the high-frequency oscillations. Allowance for the degrees of freedom corresponding to them would lead to an unimportant renormalization of certain constants. Along with the small terms containing derivatives, it is possible to take into account also terms of relativistic origin. We shall consider such terms below only in those cases in which their general form can be established without assuming any specific distribution of the moments with respect to the crystallographic axes.

In order to find the relation between the effective fields  $\mathbf{h}^\alpha$  and the moments  $\mathbf{m}^\alpha$ , it is necessary to write an expression for the deviation  $\delta E$  of the energy from the equilibrium value. We shall consider the local part of  $\delta E$ . The exchange energy of the crystal depends only on the scalar products  $\mathbf{M}^\alpha \mathbf{M}^\beta \equiv W^{\alpha\beta}$ . The deviations  $w^{\alpha\beta}$  of these quantities from the equilibrium values are

$$w^{\alpha\beta} = \mathbf{m}^\alpha \mathbf{M}^\beta + \mathbf{m}^\beta \mathbf{M}^\alpha + \mathbf{m}^\alpha \mathbf{m}^\beta. \quad (17)$$

The energy  $\delta E$  can be written in the form

$$\delta E = F^{\alpha\beta} w^{\alpha\beta} + 1/2 F^{\alpha\beta\gamma\delta} w^{\alpha\beta} w^{\gamma\delta}, \quad (18)$$

where the invariant tensors  $F^{\dots}$  satisfy the conditions

$$F^{\alpha\beta} = F^{\beta\alpha}, \quad F^{\alpha\beta\gamma\delta} = F^{\beta\alpha\gamma\delta} = F^{\alpha\beta\delta\gamma} = F^{\delta\gamma\alpha\beta}. \quad (19)$$

Furthermore, the terms in formula (18) that are linear in  $\mathbf{m}^\alpha$  must vanish. Hence we have

$$F^{\alpha\beta}M^\beta=0 \quad (20)$$

for all  $\alpha$ .

The nonlocal part of the energy  $\delta E$ , to the second order in the derivatives, is

$$\delta E = \frac{1}{2}K^{\alpha\beta\gamma}(\mathbf{m}^\alpha\partial^\gamma\mathbf{m}^\beta - \mathbf{m}^\beta\partial^\gamma\mathbf{m}^\alpha) + \frac{1}{2}a^{\alpha\beta\gamma\delta}\partial^\gamma\mathbf{m}^\alpha\partial^\delta\mathbf{m}^\beta, \quad (21)$$

where  $\partial^\alpha$  is a general linear combination of first spatial derivatives, transforming like  $\Phi^\alpha$ ;  $K^{\alpha\beta\gamma}$  and  $a^{\alpha\beta\gamma\delta}$  are invariant tensors, satisfying the conditions

$$K^{\alpha\beta\gamma} = -K^{\beta\alpha\gamma}, \quad a^{\alpha\beta\gamma\delta} = a^{\beta\alpha\gamma\delta} = a^{\alpha\beta\delta\gamma}. \quad (22)$$

Below we shall consider several characteristic examples of application of the general equations.

#### 4. EXAMPLES

1. We shall consider the antiferromagnetic structures that have been studied most, experimentally and theoretically:  $A_{2g}$  and  $A_{2u}$  in class  $D_{3d}$ . The first of these is realized, for example, in  $\text{FeCO}_3$  and  $\text{MnCO}_3$ , the second in  $\text{Cr}_2\text{O}_3$ . The moments corresponding to the one-dimensional representations  $A_{1g}$ ,  $A_{2g}$ ,  $A_{1u}$ ,  $A_{2u}$  we shall denote by  $\mathbf{M}$ ,  $\mathbf{W}$ ,  $\mathbf{P}$ ,  $\mathbf{Z}$  respectively. The pairs of moments that transform according to the two-dimensional representations  $E_g$  and  $E_u$  we shall denote by  $(\mathbf{U}, \mathbf{V})$  and  $(\mathbf{X}, \mathbf{Y})$  respectively. The deviations of the moments from the equilibrium values we shall everywhere denote by the corresponding lower-case letters. In the substances under consideration, the transition from the antiferromagnetic state to the paramagnetic occurs without change of the elementary cell; therefore the macroscopic moments essentially coincide with the spin densities of reference<sup>[10]</sup>.

In the structure  $A_{2u}$ , the equilibrium moment  $\mathbf{Z}$  is nonzero. The equations of motion (15) in the zeroth approximation take the following form:

$$\begin{aligned} \dot{\mathbf{m}} &= \gamma_0[\mathbf{h}^x \times \mathbf{Z}], \quad \dot{\mathbf{z}} = \gamma_0[\mathbf{h}^m \times \mathbf{Z}], \quad \dot{\mathbf{p}} = \gamma_1[\mathbf{h}^w \times \mathbf{Z}], \\ \dot{\mathbf{w}} &= \gamma_1[\mathbf{h}^p \times \mathbf{Z}], \quad \dot{\mathbf{u}} = \gamma_2[\mathbf{h}^y \times \mathbf{Z}], \quad \dot{\mathbf{y}} = \gamma_2[\mathbf{h}^u \times \mathbf{Z}], \\ \dot{\mathbf{v}} &= -\gamma_2[\mathbf{h}^x \times \mathbf{Z}], \quad \dot{\mathbf{x}} = -\gamma_2[\mathbf{h}^v \times \mathbf{Z}], \end{aligned} \quad (23)$$

where  $\gamma_0 = \gamma^{mzz} = \gamma^{zmm}$ ,  $\gamma_1 = \gamma^{pww} = \gamma^{wpp}$ ,  $\gamma_2 = \gamma^{uyz} = \gamma^{yuz} = -\gamma^{vzx} = -\gamma^{xvz}$ . The tensor  $F^{\alpha\beta}$  always has only diagonal components, of which in the present case, by virtue of (20), only  $F^{zz}$  vanishes. Then it follows from equations (23) that the pairs of variables  $(\mathbf{p}, \mathbf{w})$ ,  $(\mathbf{u}, \mathbf{y})$ , and  $(\mathbf{v}, \mathbf{x})$ , which are coupled to each other, correspond to oscillations with high exchange frequencies. In the determination of the low-frequency oscillations, all of these may be set equal to zero. Consequently we are left with just the first two equations (23), in which the following expressions may be used for the effective fields:

$$\begin{aligned} \mathbf{h}^m &= -F_m\mathbf{m} - K^{mz}\partial z/\partial z, \\ \mathbf{h}^z &= -\alpha_m z + K^{mz}\partial m_z/\partial z - \alpha_{uv}k_\mu k_\nu z, \end{aligned} \quad (24)$$

these take account of the relativistic anisotropy energy.

Here  $F_m = 2F^{mm}$ ,  $\alpha_{ik}$  is the anisotropy tensor, and  $\alpha_{uv}k_\mu k_\nu$  is an invariant quadratic form composed of components of the wave vector  $\{k_\mu\} = \mathbf{k}$ . The terms containing spatial derivatives are obtained from the general formula (21). Substitution of (24) in the first two equations (23) leads to a frequency spectrum that coincides with the result of the usual two-sublattice model.

A different situation arises in structure  $A_{2g}$ , where the moment  $\mathbf{W}$  is nonzero in equilibrium. Here, in the zeroth approximation, a finite result is obtained only for the following derivatives:

$$\dot{\mathbf{w}} = -\gamma_0 F_m[\mathbf{m} \times \mathbf{W}], \quad \dot{\mathbf{p}} = \gamma^{pzw}[\mathbf{h}^z \times \mathbf{W}], \quad \dot{\mathbf{z}} = \gamma^{pzw}[\mathbf{h}^p \times \mathbf{W}], \quad (25)$$

where  $\gamma_0 = \gamma^{mww}$ . Since now the only vanishing diagonal component of the tensor  $F^{\alpha\beta}$  is  $F^{ww}$ , only the two variables  $\mathbf{p}$  and  $\mathbf{z}$  must be frozen. All the remaining time derivatives vanish by virtue of the  $D_{3d}$  symmetry. To calculate them, we must take into account in the equations the terms that are linear in the spatial derivatives:

$$\begin{aligned} \dot{\mathbf{u}} &= \gamma_1 \frac{\partial}{\partial z}[\mathbf{h}^x \times \mathbf{W}] + \gamma_2 \left( \frac{\partial}{\partial x}[\mathbf{h}^x \times \mathbf{W}] - \frac{\partial}{\partial y}[\mathbf{h}^y \times \mathbf{W}] \right), \\ \dot{\mathbf{v}} &= \gamma_1 \frac{\partial}{\partial z}[\mathbf{h}^y \times \mathbf{W}] - \gamma_2 \left( \frac{\partial}{\partial x}[\mathbf{h}^y \times \mathbf{W}] + \frac{\partial}{\partial y}[\mathbf{h}^x \times \mathbf{W}] \right), \\ \dot{\mathbf{x}} &= -\gamma_1 \frac{\partial}{\partial z}[\mathbf{h}^u \times \mathbf{W}] - \gamma_2 \left( \frac{\partial}{\partial x}[\mathbf{h}^u \times \mathbf{W}] - \frac{\partial}{\partial y}[\mathbf{h}^v \times \mathbf{W}] \right) \\ &\quad + \gamma_3 \frac{\partial}{\partial y}[\mathbf{h}^m \times \mathbf{W}], \end{aligned} \quad (26)$$

$$\begin{aligned} \dot{\mathbf{y}} &= -\gamma_1 \frac{\partial}{\partial z}[\mathbf{h}^v \times \mathbf{W}] + \gamma_2 \left( \frac{\partial}{\partial x}[\mathbf{h}^v \times \mathbf{W}] + \frac{\partial}{\partial y}[\mathbf{h}^u \times \mathbf{W}] \right) \\ &\quad - \gamma_3 \frac{\partial}{\partial x}[\mathbf{h}^m \times \mathbf{W}], \end{aligned}$$

$$\dot{\mathbf{m}} = \gamma_0[\mathbf{h}^w \times \mathbf{W}] + \gamma_3 \left( \frac{\partial}{\partial x}[\mathbf{h}^y \times \mathbf{W}] - \frac{\partial}{\partial y}[\mathbf{h}^x \times \mathbf{W}] \right),$$

where

$$\begin{aligned} \gamma_1 &= \gamma_1^{uxwz} = \gamma_1^{vzwz}, \quad \gamma_2 = \gamma_1^{uxxz} = -\gamma_1^{uywv} = -\gamma_1^{vzwz} = -\gamma_1^{vzwx}, \\ \gamma_3 &= \gamma_1^{myxz} = -\gamma_1^{mxyv}. \end{aligned}$$

As will be seen below, in all low-frequency oscillatory modes the amplitude of oscillations of  $\mathbf{w}$  significantly exceeds the amplitudes of oscillations of all the other variables. Therefore in the right sides of equations (26) it is necessary to take into account those terms quadratic in the spatial derivatives that contain the variable  $\mathbf{w}$ . Such terms occur because of the terms in the expression for the effective fields  $\mathbf{h}^x$  and  $\mathbf{h}^y$  that are linear in the derivatives:

$$\mathbf{h}^x = -F_x\mathbf{x} + K_1 \frac{\partial \mathbf{w}}{\partial y}, \quad \mathbf{h}^y = -F_y\mathbf{y} - K_1 \frac{\partial \mathbf{w}}{\partial x},$$

where  $F_x = 2F^{xx} = 2F^{yy}$ ,  $K_1 = K^{wyz} = -K^{wzy}$ . In the expressions for  $\mathbf{h}^u = -F_u\mathbf{u}$  and  $\mathbf{h}^v = -F_v\mathbf{v}$  ( $F_u = 2F^{uu} = 2F^{vv}$ ), there are no such terms. They occur also in the expression for  $\mathbf{h}^w$ :

$$\mathbf{h}^w = \mathbf{h}_0^w + K_2 \left( \frac{\partial \mathbf{y}}{\partial x} - \frac{\partial \mathbf{x}}{\partial y} \right),$$

where

$$h_{0i}^w = -\alpha_{ik} w_k + a_{\mu\nu} \frac{\partial^2}{\partial x_\mu \partial x_\nu} w_i$$

is the effective field calculated without allowance for the possibility of a deviation of the other variables from equilibrium, and completely analogous to formula (24).

On taking account of everything that has been said and on expressing the spatial derivatives in terms of the components of the wave vector, we get

$$\begin{aligned} \dot{\mathbf{m}} &= \gamma_0 [\mathbf{h}_0^w \times \mathbf{W}] + i(\gamma_0 K_2 - \gamma_3 F_x) (k_x [\mathbf{y} \times \mathbf{W}] - k_y [\mathbf{x} \times \mathbf{W}]) \\ &\quad + \gamma_3 K_2 (k_x^2 + k_y^2) [\mathbf{w} \times \mathbf{W}], \\ \dot{\mathbf{u}} &= -iF_x \{ (\gamma_1 k_x + \gamma_2 k_x) [\mathbf{x} \times \mathbf{W}] - \gamma_2 k_y [\mathbf{y} \times \mathbf{W}] \} \\ &\quad - K_2 (\gamma_1 k_x k_y + 2\gamma_2 k_x k_y) [\mathbf{w} \times \mathbf{W}], \\ \dot{\mathbf{v}} &= -iF_x \{ (\gamma_1 k_x - \gamma_2 k_x) [\mathbf{y} \times \mathbf{W}] - \gamma_2 k_y [\mathbf{x} \times \mathbf{W}] \} \\ &\quad + K_2 (\gamma_1 k_x k_x + \gamma_2 k_y^2 - \gamma_2 k_x^2) [\mathbf{w} \times \mathbf{W}], \\ \dot{\mathbf{x}} &= iF_u \{ (\gamma_1 k_x + \gamma_2 k_x) [\mathbf{u} \times \mathbf{W}] - \gamma_2 k_y [\mathbf{u} \times \mathbf{W}] \} - i\gamma_3 F_m k_y [\mathbf{m} \times \mathbf{W}], \\ \dot{\mathbf{y}} &= iF_u \{ (\gamma_1 k_x - \gamma_2 k_x) [\mathbf{v} \times \mathbf{W}] - \gamma_2 k_y [\mathbf{v} \times \mathbf{W}] \} + i\gamma_3 F_m k_x [\mathbf{m} \times \mathbf{W}]. \end{aligned} \quad (27)$$

We shall consider the range of not very small values of the wave vector ( $ak^2 \gg \alpha$ ), in which the anisotropy constants may be neglected in the expression for  $\mathbf{h}_0^w$ . If instead of  $\mathbf{w}$  we introduce the new variable  $\mathbf{w}' = k\mathbf{w}$ , then Eqs. (27) together with the first of equations (25) will constitute a closed system of linear equations, with coefficients that are homogeneous functions of the first order of the frequency  $\omega$  and the components of the wave vector. The characteristic equation is cubic in  $\omega^2$ ; therefore there are three distinct frequencies  $\omega_{1,2,3}^I(\mathbf{k})$  each of which is a homogeneous function of the first order of the components of the wave vector. Let  $\xi, \eta$  be mutually perpendicular unit vectors lying in the plane perpendicular to  $\mathbf{W}$  (for what follows, it is convenient to choose them as the principal axes of the anisotropy tensor  $\alpha_{ik}$ ). The complete system of equations splits into two groups of equations for the variables  $(m_\xi, w'_\eta, x_\eta, y_\eta, u_\xi, v_\xi)$  and  $(m_\eta, w'_\xi, x_\xi, y_\xi, u_\eta, v_\eta)$ ; the coefficients of these two groups of equations transform into each other under the substitution  $\omega \rightarrow -\omega$ . To each of the frequencies  $\omega_{1,2,3}^I(\mathbf{k})$  correspond two types of oscillations, one for each group of variables.

These six types of oscillations, in the range of small values of the wave vector ( $ak^2 \ll \alpha$ ), behave differently in antiferromagnets of the easy-axis type ( $\text{FeCO}_3$ ) and of the easy-plane type ( $\text{MnCO}_3$ ).

In the first case, the anisotropy tensor has the form  $\alpha_{ik} = \alpha \delta_{ik}$ , and the four variables  $(m_\xi, w'_\eta)$  and  $(m_\eta, w'_\xi)$  correspond to oscillations with finite frequency  $\omega_0^2 = \gamma_0^2 W^2 \alpha F_m$ . The remaining four types of oscillations have significantly lower frequencies. Therefore to determine them, we may set  $\mathbf{m} = \mathbf{w}' = 0$  in the last four equations (27). The corresponding characteristic equation is quadratic in  $\omega^2$ , so that there are two frequen-

cies  $\omega_{1,2}^{II}(\mathbf{k})$ , homogeneous functions of the first order of  $\mathbf{k}$ ; to each of them correspond two types of oscillations, one for each of the groups of variables  $(x_\eta, y_\eta, u_\xi, v_\xi)$  and  $(x_\xi, y_\xi, u_\eta, v_\eta)$ .

In the second case, the anisotropy tensor may be supposed to be  $\alpha_{ik} = \alpha \xi_i \xi_k$  (the direction  $\xi$  coincides with a third-order axis). One type of oscillations (oscillations of the variables  $m_\eta, w'_\xi$ ) corresponds to a finite frequency  $\omega_0$ . The remaining five types have frequencies that are linear functions of  $|\mathbf{k}|$ . It is clear that the equations for the variables  $(m_\xi, w'_\eta, x_\eta, y_\eta, u_\xi, v_\xi)$  in this case are the same as in the range  $ak^2 \gg \alpha$ . Therefore this group of variables produces three types of oscillations with frequencies  $\omega_{1,2,3}^{II}(\mathbf{k})$ . The equations for the variables  $(x_\xi, y_\xi, u_\eta, v_\eta)$  coincide with the equations in the range  $ak^2 \ll \alpha$  for the easy-axis case. These variables produce the remaining two types of oscillations with frequencies  $\omega_{1,2}^{II}(\mathbf{k})$ .

We note that the sublattice model, which has been applied to the structure  $A_{2x}$  (see<sup>[51]</sup>), describes only two of the six types of oscillations. It is necessary, however, to emphasize that these two branches, in the experimentally investigated substances with group  $D_{3d}$  ( $\text{FeCO}_3, \text{MnCO}_3$ ), are described by the sublattice model with quite high accuracy. Because this model corresponds to classical localized spins, and in the localized picture the variables  $\mathbf{u}, \mathbf{v}, \mathbf{x}, \mathbf{y}$  are absent in the substances under consideration (see<sup>[101]</sup>), the coefficients  $F_u$  and  $F_x$  that enter in the general equations (27) are apparently anomalously large; and this leads, as is seen from (27), to an increase of the frequencies of the anomalous branches, and in particular to an increase of the purely relativistic gaps corresponding to them. Nor must the extreme possibility be excluded, that the anomalous branches may disappear altogether, if the large value of  $F_u$  and  $F_x$  is more important than the smallness of relativistic effects in comparison with exchange.

2. In the example given above, all branches of the oscillations have the linear dependence of frequency on wave vector that is characteristic of antiferromagnets. We shall now consider an antiferromagnetic structure in group  $D_3$ , where there are unusual branches of the low-frequency oscillations. In equilibrium let the antiferromagnetic moment  $\mathbf{W}$  (representation  $A_2$ ) be non-zero, whereas  $\mathbf{M}^0 = \mathbf{U} = \mathbf{V} = 0$ , where  $\mathbf{U}$  and  $\mathbf{V}$  are the moments that transform according to representation  $E$ . The deviations of the moments from equilibrium we shall denote by the corresponding lower-case letters. By setting up the equations of motion for  $\mathbf{m}^0$  and  $\mathbf{w}$  according to the general rules, it is easy to show that they couple to each other in the usual manner, so that the standard antiferromagnetic branches of the oscillations arise. In addition to these, however, there exist in the present case other branches with significantly lower frequencies. To determine them, therefore, we may set  $\mathbf{m}^0 = \mathbf{w} = 0$  and consider oscillations of the values of  $\mathbf{u}$  and  $\mathbf{v}$  alone. Then by virtue of the  $D_3$  symmetry, there are no terms with  $\gamma^{\alpha\beta\gamma}$  and  $\gamma_1^{\alpha\beta\gamma\delta}$  in the expression for  $\Gamma^{\alpha\beta\gamma}$ , so that the following equations are obtained:

$$\begin{aligned}\dot{\mathbf{u}} &= \gamma_2(\Delta^u[\mathbf{h}^u \times \mathbf{W}] - \Delta^v[\mathbf{h}^v \times \mathbf{W}]), \\ \dot{\mathbf{v}} &= -\gamma_2(\Delta^v[\mathbf{h}^u \times \mathbf{W}] + \Delta^u[\mathbf{h}^v \times \mathbf{W}]),\end{aligned}\quad (28)$$

where  $\gamma_2 = \gamma_2^{uuuu} = -\gamma_2^{vvvv} = -\gamma_2^{uvuv} = -\gamma_2^{vuvu}$ . The general linear combinations  $\Delta^u, \Delta^v$  in this symmetry are

$$\Delta^u \approx 2 \frac{\partial^2}{\partial x \partial y} - a \frac{\partial^2}{\partial z \partial y}, \quad \Delta^v \approx \frac{\partial^2}{\partial x^2} - \frac{\partial^2}{\partial y^2} + a \frac{\partial^2}{\partial z \partial x},$$

where  $a$  is an arbitrary constant. The tensor  $F^{\alpha\beta}$  has the components  $F^{uu} = F^{vv} = F/2$ ; therefore the effective fields  $\mathbf{h}^u$  and  $\mathbf{h}^v$  may be supposed to be  $-\mathbf{F}\mathbf{u}$  and  $-\mathbf{F}\mathbf{v}$  respectively. With allowance for this, we get from equations (28) the following spectrum of oscillations:

$$\omega^2 = F^2 W^2 \gamma_2^2 \{ (k_x^2 - k_y^2 + a k_x k_z)^2 + (2k_x k_y - a k_z k_y)^2 \}. \quad (29)$$

Thus in antiferromagnets there can exist spin waves of the ferromagnetic type, with a quadratic dependence of frequency on wave vector. These oscillations, if they exist, make an important contribution to the thermodynamics of the crystal. For example, the specific heat of the crystal, because of these oscillations, should be proportional to  $T^{3/2}$ , and not, as is usual, to  $T^3$ . The gap that may appear in the energy spectrum (29) upon allowance for relativistic interactions should be purely relativistic, as in ferromagnets, not semi-relativistic, as in the usual spectra of antiferromagnets. We note in this connection that in an antiferromagnetic structure with group  $C_{3v}$  (this group is isomorphic to  $D_3$ ) and with a single moment  $\mathbf{W}$ , corresponding to representation  $A_2$ , there should exist, along with the usual branches, anomalous ones with a linear dependence of frequency on wave vector—in which, however, the gap may be only purely relativistic. When the frequencies of the anomalous and usual branches differ greatly, the equations for  $\mathbf{u}$  and  $\mathbf{v}$  in group  $C_{3v}$  are analogous to equations (28), but in the first of these there is an additional term  $\gamma_1 \nabla^0[\mathbf{h}^v \times \mathbf{W}]$ , and in the second  $-\gamma_1 \nabla^0[\mathbf{h}^u \times \mathbf{W}]$ , where  $\gamma_1 = \gamma_1^{uvuv} = -\gamma_1^{vuvu}$ . Otherwise all the calculations are identical to the preceding. On taking into account that in group  $C_{3v}$

$$\nabla^0 \approx \frac{\partial}{\partial z}, \quad \Delta^u \approx 2 \frac{\partial^2}{\partial x \partial y} + a \frac{\partial^2}{\partial z \partial x}, \quad \Delta^v \approx \frac{\partial^2}{\partial x^2} - \frac{\partial^2}{\partial y^2} + a \frac{\partial^2}{\partial z \partial y}$$

we get for the frequencies of the anomalous branches the following expression:

$$\omega^2 = F^2 W^2 \{ \gamma_1^2 k_x^2 + \gamma_2^2 (k_x^2 + k_y^2)^2 \}. \quad (30)$$

At temperatures less than the width of the semirelativistic gap of the ordinary oscillations, the anomalous branches make the main contribution to the specific heat; this contribution is proportional to the square of the temperature.

3. We shall consider the ferrimagnetic structure ( $A_g, B_g$ ) in the class  $C_{2h}$ . This group has only the one-dimensional representations  $A_g, B_g, A_u, B_u$ , which we shall number by values of the index  $\alpha$  equal respectively to 0, 1, 2, 3. In equilibrium the moments  $\mathbf{M}^0$  and  $\mathbf{M}^1$  are nonzero. Equations (16), without allowance for terms with derivatives, have in the present case the

following form:

$$\begin{aligned}\dot{\mathbf{m}}^0 &= \gamma^{000}[\mathbf{h}^0 \times \mathbf{M}^0] + \gamma^{011}[\mathbf{h}^1 \times \mathbf{M}^1] + \hat{\gamma}^{010} \mathbf{M}^0(\mathbf{h}^1 \nu) - \hat{\gamma}^{100} \nu(\mathbf{M}^0 \mathbf{h}^1) \\ &\quad + \hat{\gamma}^{001} \mathbf{M}^1(\mathbf{h}^0 \nu) - \hat{\gamma}^{001} \nu(\mathbf{M}^1 \mathbf{h}^0), \\ \dot{\mathbf{m}}^1 &= \gamma^{101}[\mathbf{h}^0 \times \mathbf{M}^1] + \gamma^{110}[\mathbf{h}^1 \times \mathbf{M}^0] + \hat{\gamma}^{100} \mathbf{M}^0(\mathbf{h}^0 \nu) - \hat{\gamma}^{010} \nu(\mathbf{M}^0 \mathbf{h}^0) \\ &\quad + \hat{\gamma}^{111} \mathbf{M}^1(\mathbf{h}^1 \nu) - \hat{\gamma}^{111} \nu(\mathbf{M}^1 \mathbf{h}^1), \\ \dot{\mathbf{m}}^2 &= \gamma^{231}[\mathbf{h}^3 \times \mathbf{M}^1] + \gamma^{220}[\mathbf{h}^2 \times \mathbf{M}^0] + \hat{\gamma}^{230} \mathbf{M}^0(\mathbf{h}^3 \nu) - \hat{\gamma}^{320} \nu(\mathbf{M}^0 \mathbf{h}^3) \\ &\quad + \hat{\gamma}^{221} \mathbf{M}^1(\mathbf{h}^2 \nu) - \hat{\gamma}^{221} \nu(\mathbf{M}^1 \mathbf{h}^2), \\ \dot{\mathbf{m}}^3 &= \gamma^{330}[\mathbf{h}^3 \times \mathbf{M}^0] + \gamma^{321}[\mathbf{h}^2 \times \mathbf{M}^1] + \hat{\gamma}^{320} \mathbf{M}^0(\mathbf{h}^2 \nu) - \hat{\gamma}^{230} \nu(\mathbf{M}^0 \mathbf{h}^2) \\ &\quad + \hat{\gamma}^{331} \mathbf{M}^1(\mathbf{h}^3 \nu) - \hat{\gamma}^{331} \nu(\mathbf{M}^1 \mathbf{h}^3),\end{aligned}\quad (31)$$

where the quantities  $\hat{\gamma}^{\alpha\beta\alpha}$  are related to  $\gamma^{\alpha\beta\gamma\sigma}$  in the same way that  $\hat{\Gamma}_{1,2}^{\alpha\beta\alpha}$  are related to  $\Gamma_{1,2}^{\alpha\beta\gamma\sigma}$ ; that is,  $\hat{\gamma}^{\alpha\beta\alpha} = \gamma^{\alpha\beta\alpha\beta} \epsilon^{bc\pi}$ . The tensor  $F^{\alpha\beta}$  has two nonvanishing components  $F^{22} = F_2/2$ ,  $F^{33} = F_3/2$ , and the local part of the energy is

$$\begin{aligned}\delta E &= 1/2 A (\mathbf{m}^0 \mathbf{M}^0)^2 + 1/2 B (\mathbf{m}^1 \mathbf{M}^1)^2 + C (\mathbf{m}^2 \mathbf{M}^0) (\mathbf{m}^1 \mathbf{M}^1) \\ &\quad + 1/2 D (\mathbf{m}^2 \mathbf{M}^1 + \mathbf{m}^1 \mathbf{M}^2)^2 + 1/2 E (\mathbf{m}^2 \mathbf{M}^0)^2 + 1/2 G (\mathbf{m}^3 \mathbf{M}^1)^2 \\ &\quad + H (\mathbf{m}^2 \mathbf{M}^0) (\mathbf{m}^3 \mathbf{M}^1) + 1/2 P (\mathbf{m}^2 \mathbf{M}^0)^2 + 1/2 Q (\mathbf{m}^3 \mathbf{M}^1)^2 \\ &\quad + R (\mathbf{m}^2 \mathbf{M}^0) (\mathbf{m}^3 \mathbf{M}^1) + 1/2 F_2 (\mathbf{m}^2)^2 + 1/2 F_3 (\mathbf{m}^3)^2,\end{aligned}$$

where  $A, B, \dots, R$  are certain constants of exchange origin. On calculating the effective fields of the zeroth approximation by means of the last formula and on substituting them in equations (31), we get

$$\begin{aligned}(\mathbf{m}^0 \nu) &= \lambda_1 (\mathbf{m}^0 \mathbf{M}^1 + \mathbf{m}^1 \mathbf{M}^0), \quad (\mathbf{m}^1 \nu) = (A \lambda_2 + C \lambda_3) (\mathbf{m}^2 \mathbf{M}^0) \\ &\quad + (C \lambda_2 + B \lambda_3) (\mathbf{m}^1 \mathbf{M}^1), \\ (\dot{\mathbf{m}}^2 \nu) &= F_2 \left( \gamma^{220} \frac{M_0}{M_1} - \hat{\gamma}^{221} \right) (\mathbf{m}^2 \mathbf{M}^1) - F_3 \left( \gamma^{231} \frac{M_1}{M_0} - \gamma^{230} \right) (\mathbf{m}^1 \mathbf{M}^0), \\ (\dot{\mathbf{m}}^3 \nu) &= F_3 \left( \gamma^{330} \frac{M_0}{M_1} - \hat{\gamma}^{331} \right) (\mathbf{m}^3 \mathbf{M}^1) - F_2 \left( \gamma^{321} \frac{M_1}{M_0} - \hat{\gamma}^{320} \right) (\mathbf{m}^2 \mathbf{M}^0), \\ (\dot{\mathbf{m}}^2 \mathbf{M}^0) &= F_3 M_0^2 \left( \gamma^{231} \frac{M_1}{M_0} - \hat{\gamma}^{230} \right) (\mathbf{m}^1 \nu), \\ (\dot{\mathbf{m}}^1 \mathbf{M}^0) &= F_2 M_0^2 \left( \gamma^{321} \frac{M_1}{M_0} - \hat{\gamma}^{320} \right) (\mathbf{m}^2 \nu), \\ (\dot{\mathbf{m}}^2 \mathbf{M}^1) &= -F_2 M_1^2 \left( \gamma^{220} \frac{M_0}{M_1} + \hat{\gamma}^{221} \right) (\mathbf{m}^2 \nu), \\ (\dot{\mathbf{m}}^3 \mathbf{M}^1) &= -F_3 M_1^2 \left( \gamma^{330} \frac{M_0}{M_1} + \hat{\gamma}^{331} \right) (\mathbf{m}^3 \nu),\end{aligned}\quad (32)$$

where  $M_0, M_1$  are the moduli of the equilibrium moments, and where

$$\begin{aligned}\lambda_1 &= (\gamma^{000} M_0 M_1 - \gamma^{011} M_0 M_1 + \hat{\gamma}^{100} M_0^2 + \hat{\gamma}^{001} M_1^2) D, \\ \lambda_2 &= -\gamma^{011} M_0 M_1 + \hat{\gamma}^{010} M_0^2, \quad \lambda_3 = \gamma^{110} M_0 M_1 + \hat{\gamma}^{111} M_1^2.\end{aligned}$$

The time derivatives of all the remaining quantities vanish in the zeroth approximation. From Eqs. (32) it is evident that the oscillations of the variables  $(\mathbf{m}^2 \nu)$  and  $(\mathbf{m}^3 \nu)$ , together with certain linear combinations of the pairs of variables  $(\mathbf{m}^2 \mathbf{M}^1)$ ,  $(\mathbf{m}^3 \mathbf{M}^0)$  and  $(\mathbf{m}^3 \mathbf{M}^1)$ ,  $(\mathbf{m}^2 \mathbf{M}^0)$  respectively, occur with a high exchange frequency. These variables must therefore be frozen in the calculation of the low-frequency oscillations.

From Eqs. (31) without use of the zeroth approximation for the effective fields, we find

$$\begin{aligned}(\lambda_2 A + \lambda_3 C) (\mathbf{m}^2 \mathbf{M}^0) + (C \lambda_2 + B \lambda_3) (\mathbf{m}^1 \mathbf{M}^1) &= 2 \hat{A} (\mathbf{h}^1 \nu), \\ \lambda_1 (\mathbf{m}^2 \mathbf{M}^1 + \mathbf{m}^1 \mathbf{M}^2) &= 2 \hat{B} (\mathbf{h}^0 \nu),\end{aligned}\quad (33)$$

where  $\hat{A} = \frac{1}{2} (A \lambda_2^2 + 2C \lambda_2 \lambda_3 + B \lambda_3^2)$ ,  $\hat{B} = \lambda_1^2 / 2D$ . On the right



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## Magneto-resonance investigations of antiferromagnetic FeCl<sub>3</sub>

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The temperature dependence of the line width is investigated for ESR at  $T > T_N$ , for NMR of the Fe<sup>57</sup>, Cl<sup>35</sup> and Cl<sup>37</sup> nuclei at  $T < T_N$  and for AFMR in magnetically ordered FeCl<sub>3</sub>. The critical exponent that describes the broadening of the ESR line as the temperature approaches  $T_N = 9^\circ\text{K}$  is found to vary between 2.5 for  $T > 22^\circ\text{K}$  and 0.56 for  $T < 20^\circ\text{K}$ . The NMR spectrum of Fe<sup>57</sup> corresponds to  $H_{\text{hf}}(T \rightarrow 0) = 487$  kOe whereas the NMR spectrum of the Cl<sup>35</sup> and Cl<sup>37</sup> nuclei is almost continuous in the 20-55 MHz frequency range. The temperature dependence of the NMR spectrum is not described by a Brillouin function. The AFMR spectrum was observed in the 28 to 57 GHz frequency range. The frequency and field dependences of the AFMR spectral lines do not agree with those expected for a uniaxial antiferromagnet. A discussion of the results shows that they are consistent with the complex helicoidal magnetic structure of FeCl<sub>3</sub> obtained by neutron-diffraction measurements.

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FeCl<sub>3</sub> crystals belong to the BiI<sub>3</sub> structure type (space group  $R\bar{3}$ )<sup>[1]</sup> and are similar in structure to the layered ferromagnet CrBr<sub>3</sub>. There are indications that FeCl<sub>3</sub> can go over at  $T < 250^\circ\text{K}$  into a different structural form,<sup>[2]</sup> but it is stated in<sup>[3]</sup>, where the magnetic structure was investigated by neutron diffraction, that the crystal structure of FeCl<sub>3</sub> in the magnetically-ordered state corresponds to that described in<sup>[1]</sup> (Fig. 1) with slightly modified lattice parameters. An investigation of the susceptibility<sup>[4]</sup> and of the Mössbauer effect<sup>[5]</sup> have yielded the antiferromagnetic ordering temperature  $T_N = 9.1^\circ\text{K}$ , whereas a value  $T_N = 15^\circ\text{K}$  is reported in<sup>[3,6]</sup>.

It is reported in<sup>[3]</sup> that the magnetic structure of FeCl<sub>3</sub> is complicated and such that the spins of the Fe<sup>3+</sup> ions that are closest to one another along the hexagonal axis are oppositely directed and lie in the crystallographic plane (1450) (hexagonal Miller indices), but the directions of the spins in the nearest adjacent planes of the (1450) type are rotated relative to one another through an angle  $2\pi/15$ . Thus, the antiferromagnetism vector forms a helical structure with a helix axis along the  $[14\bar{5}0]$  directions and with a period equal to  $15d$ , where  $d$  is the distance between the nearest planes of the (1450) type. Measurements of the Mössbauer effect on Fe<sup>57</sup><sup>[7]</sup> yield a value  $H_{\text{hf}} = 487 \pm 15$  kOe for the hyperfine field. The measurements of the temperature dependence of  $H_{\text{hf}}$  in these studies are not accurate enough, and measurements in external magnetic fields<sup>[5]</sup> do not allow us to state whether a helical magnetic structure is present or not, owing to the small values of the employed fields.

Taking into account the contradictory character of

the information on the magnetic properties of the FeCl<sub>3</sub> crystals, and also the interest in the helicoidal magnetic structures, we have carried out a group of investigations of FeCl<sub>3</sub> crystals, including the following: a) electron spin resonance (ESR) at  $T > T_N$ , b) nuclear magnetic resonance (NMR) in the magnetically ordered state, and c) antiferromagnetic resonance (AFMR).

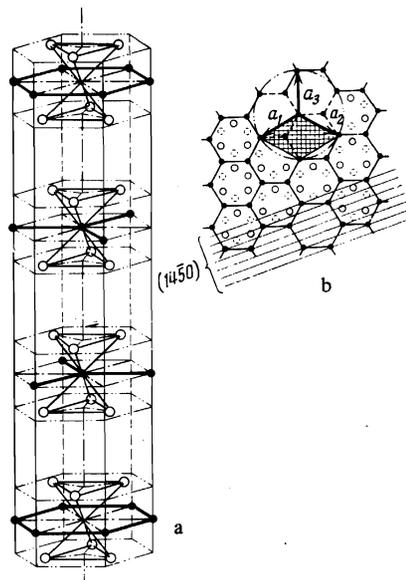


FIG. 1. a)  $R\bar{3}$  crystal structure of FeCl<sub>3</sub>; b) projection of the FeCl<sub>3</sub> "sandwich" on the (0001) plane: ●—Fe in the (00001) plane, ○—Cl above the (0001) plane, dashed circle—Cl below the (0001) plane. Shaded part—area of unit cell.  $a_1, a_2, a_3$  axes of the chosen coordinate system. Straight lines—traces of the intersection of the (0001) plane with planes of the (1450) type.