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# Wave Turbulence Under Parametric Excitation

Applications to Magnets

With 69 Figures

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### 2 The General Properties of Magnetodielectrics

University and college departments of physics traditionally do not include advanced courses in magnetism in their curriculum. Therefore we offer the reader this short review chapter presenting necessary data on magnetically ordered dielectrics. This saves the reader the necessity of referring to many books on magnetism [2.1–13].

### 2.1 Classification of Substances by Their Magnetic Properties

All substances have more or less pronounced magnetic properties in the sense that their properties change to some degree under the influence of a magnetic field. One main parameter quantifying this influence is the magnetic susceptibility  $\chi$ , the derivative of the magnetization M with respect to the strength of the magnetic field H. All substances fall into one of four groups according to the magnitude and sign of  $\chi$ :

#### 2.1.1 Diamagnets

Diamagnets have a magnetic susceptibility which is negative and small compared to unity  $|\chi| \simeq 10^{-6}$ . Atoms with no magnetic moment of their own are usually diamagnetic. Diamagnetism can be explained by the Lenz rule according to which a current arising in the system of charges (electron shells of atoms) placed into the magnetic field tends to reduce this field. Therefore their resulting magnetic moment is directed opposite to the applied field.

#### 2.1.2 Superconductors

From the viewpoint of their magnetic properties, superconductors constitute a special group. They eject the magnetic field into which they are placed when its strength is below some critical value. This phenomenon is called the *Meissner effect*. It results in *strong diamagnetism* when  $\chi = -1/4\pi$ .

#### 2.1.3 Paramagnets

These are substances with a low positive susceptibility. They comprise ions with uncompensated magnetic moments of electrons  $\mu$  that have a spin nature:  $\mu = \mu_B S$  where  $\mu_B$  is the Bohr magneton, S is the spin of an atom (ion). Such ions are termed paramagnetic. Examples of such paramagnetic ions are elements with incomplete d-shells (elements of the Ferrum group: Fe, Ni, Co, Mn), f-shells (rare earth elements Sm, Dy, Yb, Lu, Tm), etc. The magnetic moment of the paramagnets is a result of two competing factors, the external magnetic field and temperature. The first factor tends to produce parallel orientation of the magnetic moment of ions: under the influence of the second one the magnetic moment of ions becomes chaotic. At high temperatures the resulting magnetic moment is small  $\chi \simeq n\mu/T$ (here n is the concentration of paramagnetic atoms with moment  $\mu$ , T is the temperature in energy units). At room temperature  $\chi \simeq 10^{-3}$ . This picture is observed if the interaction between the magnetic moments of the ions is sufficiently small compared with the temperature. If the temperature is decreased the substance passes to a magnetically ordered state.

#### 2.1.4 Magnetically Ordered Substances (Magnets)

These have many typically magnetic properties and are characterized in particular by an additional type of collective excitations, spin waves, whereby the magnetic moments oscillate with respect to the ordered orientation. Nonlinear properties of spin waves will be the main object of study in this book. Spin wave properties are, with other magnetic properties, largely determined by the type of magnetic ordering, or, in other words, by the magnetic structure of magnets. Let us consider the types of magnetic structure.

1 Ferromagnets - FM. According to modern classification these are substances in which the magnetic moments of all the atoms have parallel orientation. This results in a macroscopic magnetic moment equal to the sum of the magnetic momentum of all the atoms. Without the external magnetic field such a fully ordered state often proves to be thermodynamically unstable and the FM breaks into domains, i.e. macroscopic regions in which the moments of the electrons are parallel. The resultant moment of the sample then approximates zero. Then the ferromagnet (e.g. iron) is said to be not magnetized, or in the multidomain state. To attain a one-domain state, an external magnetic field should be applied. Lest this state be broken without the external magnetic field, the motion of the domain walls and the rotation of magnetic moments inside the domains must be specially prevented. For more details on the domain structure see [2.4, 9]. In the general case magnetic ordering is accompanied by the appearance of several magnetic sublattices, each of which is a group of ions with similar magnetic moments.

To be more exact, magnetic ions of the same sublattice are translationally invariant, i.e. can be replaced by each other via an integer number of elementary translations of the crystal lattice. The number of magnetic sublattices is in this case determined by the number of magnetic atoms (ions) in an elementary cell allowing for the magnetic order. Such a magnetic cell may comprise an additional elementary crystal cell. From this viewpoint FMs are substances with a single magnetic sublattice.

- 2 Ferrimagnets. These are substances with several magnetic sublattices (with magnetization  $M_i$ ). Interaction between the sublattices results in an orientation such that the total magnetic moment  $M = \sum_{i} M_{i}$  is nonzero. In the simplest case there are two collinear sublattices with different magnetic moments  $M_1 \neq M_2$  with antiparallel orientation so that  $M = M_1 - M_2 \neq 0$ . For three or more sublattices the orientation is not necessarily collinear. A classical example of a ferrimagnet is the Yttrium-Iron Garnet (YIG) Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> with twenty magnetic sublattices.
- 3 Antiferromagnets AFM. Unlike the ferrimagnet, the sum of magnetic moments of the AFM lattices is equal to zero. The simplest case of an AFM has two equivalent antiparallel sublattices  $M_1 = -M_2$ . Antiferromagnetic ordering is typical, for instance in oxides MnO, FeO, CoO, and fluorides  $MnF_2$ ,  $CoF_2$ ,  $FeF_2$ .
- 4 Antiferromagnets with weak ferromagnetism. Unlike pure AFMs the sublattices of these substances are weakly uncollinear owing to a specific relativistic Dzyaloshinsky-Moria interaction [2.9, 16]. Since such interaction is significantly weaker than the exchange interaction, the resultant magnetic moment M is much less than the magnetization of each sublattice. Examples of such a structure are  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, NiF<sub>2</sub>, MnCO<sub>2</sub> and CoCO<sub>2</sub>.
- 5 Helicoidal structures or helicomagnets. These substances are characterized by much more complicated ordering and cannot be described in terms of magnetic sublattices. If there is one magnetic atom per elementary crystal cell (in the paramagnetic phase) such structure may be represented, say, as a "stationary wave of the magnetic moment"

$$\mu_{jz} = \mu_z , \quad \mu_{jx} = \mu \cos(qR_j) , \quad \mu_{jy} = \mu \sin(qR) .$$
 (2.1.1)

Here  $\mu_i$  is the ion magnetization in the cell  $R_i$ , q is the wave vector of the spiral incommensurable with the period of the reciprocal lattice. In the socalled simple S-S spiral  $\mu_z = 0$ , and the resultant magnetic moment of the crystal is equal to zero. Such an ordering is antiferromagnetic. If  $\mu_z \neq 0$  the spiral is called conical or ferromagnetic. There are more complicated spirals in which  $\mu_z$  depends periodically on the ion number. Helicoid magnetic

structure at low temperature is typical for many rare earth metals. Such structures have also been observed in alloys of MnAu<sub>2</sub>, the compound MnI<sub>2</sub>, etc.

6 Magnetic glasses. This is a wide class of substances, which are of great interest for theoreticians and promising for practical applications. Magnetic glasses can be called congealed paramagnets with some magnetic structure but without long-range magnetic ordering. Their sample average magnetic moment is zero, but thermodynamic average of the magnetic moment of each paramagnetic atom differs from zero. As in magnetically ordered substances, magnetic glasses may support the propagation of long spin waves. There is an extensive literature on magnetic glasses and amorphous magnets (see, for example, [2.14]).

In conclusion of this section it must be noticed that the above classification is fairly schematic since it is always difficult to fit reality into formulae. For instance, there is no strict borderline between AFMs with weak ferromagnetism and ferrimagnets, between helimagnets with the spiral period in commensurable with the crystallographic period and multi-sublattice AFMs. For more details on these problems – types of magnetic ordering, its relation to crystal symmetry, etc. – see [2.9, 12], and the works by Landau [2.15] and Dzyalochinsky [2.16].

#### 2.2 Nature of Interaction of Magnetic Moments

It is common knowledge that the strongest interaction determining the type of magnetic structure is usually the *exchange interaction*. It is electrostatic in nature and is due to the Pauli principle which states that no two electrons can exist in identical quantum states. The origin of the exchange interaction can best be illustrated by the simple example of the hydrogen molecule.

#### 2.2.1 Exchange Interaction in the Hydrogen Molecule

The Hamiltonian  $\mathcal{H}$  of this system consists of the unperturbed Hamiltonians of the two atoms a and b,  $\mathcal{H}_a$  and  $\mathcal{H}_b$ , and the Hamiltonian of their interaction

$$\mathcal{H}_{ab} = e^2 \left[ \frac{1}{R_{ab}} + \frac{1}{R_{12}} - \frac{1}{R_{a2}} - \frac{1}{R_{b1}} \right], \tag{2.2.1}$$

where  $R_{ab}$  is the distance between the protons,  $R_{12}$  denotes the distance between the electrons,  $R_{a2}$  and  $R_{b1}$  is the distance between electron of the atom a and the proton of the atom b and vice versa. Since  $\mathcal{H}_{ab}$  does not depend on the spin variables  $S_1$ ,  $S_2$  the multiplication form of the wave function of the molecule must be sought. In accordance with the Pauli principle the wave function  $\Psi$  must be antisymmetric under exchange of electrons. As in the state with total spin of the system S=1, the electron spins are parallel, the spin term of  $\Psi$  will be symmetrical. Its coordinate term  $\varphi$  will therefore be antisymmetric. Accordingly, in the state with antiparallel spins, S=0, the function  $\varphi$  will be symmetric. To first order in the perturbation theory in the interaction Hamiltonian  $\mathcal{H}_{ab}$  we construct the functions of the zeroth approximation  $\varphi_{\pm}(\mathbf{R}_1,\mathbf{R}_2)$  using the wave functions of the hydrogen atoms  $\varphi(\mathbf{R}_{ci})$  composed of the *i*-th electron (i=1,2) and *c*-th nucleus (c=a,b):

$$\varphi_{\pm}(\mathbf{R}_1, \mathbf{R}_2) = \frac{\left[\varphi(\mathbf{R}_{a1})\varphi(\mathbf{R}_{b2}) \pm \varphi(\mathbf{R}_{a2})\varphi(\mathbf{R}_{b1})\right]}{2\sqrt{1-\delta^2}}, \qquad (2.2.2)$$

where  $\delta$  is the overlap integral. In this approximation the energy  $E_+$  of hydrogen molecule in the state with S=1 and the energy  $E_-$  of the molecule with S=0 have the following forms:

$$E_{\pm} = \int \varphi_{\pm}^{2}(\mathbf{R}_{1}, \mathbf{R}_{2}) \,\mathcal{H}_{ab} \, d\mathbf{R}_{1} d\mathbf{R}_{2} = [A(R_{ab}) \pm B(R_{ab})]/(1 + \delta^{2}) , (2.2.3)$$

$$A(R_{ab}) = \int \mathcal{H}_{ab} \varphi^{2}(\mathbf{R}_{a1})\varphi^{2}(\mathbf{R}_{b2}) d\mathbf{R}_{1} d\mathbf{R}_{2} ,$$

$$B(R_{ab}) = \int \mathcal{H}_{ab} \varphi(\mathbf{R}_{a1})\varphi(\mathbf{R}_{b1})\varphi(\mathbf{R}_{a2})\varphi(\mathbf{R}_{b2}) d\mathbf{R}_{1} d\mathbf{R}_{2} .$$

$$(2.2.4)$$

Computation using these formulas shows that the function  $E_{+}(R_{ab}) > 0$  and decreases monotonically as the distance between the nuclei  $R_{ab}$  increases and the function  $E_{-}(\mathbf{R}_{ab})$  has a sharp minimum under some  $R_{ab} = R_0$ . It is important that  $E_{-}(R_{ab}) < 0$ . This means that two hydrogen atoms can form a molecule only in a state with antiparallel spins. The quantummechanical explanation of the homopolar chemical bond presented in our book was given by Heitler and London in 1927 [2.17]. As can readily be seen from (2.2.4), the function  $A(R_{ab})$  determines the Coulomb energy of two atoms on the assumption that the first electron "belongs" only to the nucleus a, and the second one belongs to the nucleus b. As the distance between the atoms increases this energy diminishes as  $1/R_{ab}$ . The function  $B(R_{ab})$  is a nondiagonal matrix element of the electrostatic energy  $\mathcal{H}_{ab}$ between the state  $\langle a1, b2 |$  in which the first electron belongs to the atom a and the second electron belongs the atom b, and the state  $|a2,b1\rangle$  in which the electrons exchange their positions. Since the function  $B(R_{ab})$  is non-zero only due to the overlap of electron shells it decreases exponentially as the distance increases.

This simple example shows that the pure quantum effect of electron exchange leads to an interaction between the atoms determined by the total spin of the atom S in spite of the fact that the Hamiltonian of the interaction is independent of the spin variables. The two expressions (2.2.3) for the

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binding energy  $E_{\pm}$  in the state with total spin S=0 and S=1 may be written uniformly using the spin variables  $S_1, S_2$ .

$$E_{+} = E - JS_{1}S_{2}$$
,  $E = 3E_{-}/4 + E_{+}/4$ ,  $J = E_{-} - E_{+}$ . (2.2.5)

Here it has been considered that the values of the operator  $S_1S_2$  are given by 3/4 in the singlet state (when  $S_1 = 1$ ) and 1/4 in the triplet state (when S = 0). The spin-dependent term in (2.2.5) is called the *Hamiltonian of exchange interaction* and is denoted as

$$\mathcal{H}_{\text{ex}} = -JS_1S_2 \ . \tag{2.2.6}$$

The function  $J(R_{12})$  is called the exchange integral. If J < 0 (as in our example) the antiparallel spin orientation is desirable, then the exchange interaction is said to be antiferromagnetic. Under J > 0 parallel orientation of spins is established and the exchange is called ferromagnetic.

The operator (2.2.6) is commonly known as the Heisenberg Hamiltonian, though in this form it was first obtained by Dirac and first used in the theory of magnetism by Van Flek. But it was Heisenberg who showed that interactions resulting in magnetic ordering had quantum-mechanical nature due to the Pauli principle. The original idea that spontaneous magnetization is due to the specific interaction between the magnetic atoms was first suggested by Weiss in 1907. His estimation of the temperature of transition based on the magnetic dipole interaction, however, was 0.25 K, which is less than its real value by a factor of 10<sup>4</sup>. But in spite of such a failure he prove to be an incorrigible optimist, saing that "this difficulty must be treated not as an argument against this theory of molecular field, but as an incentive for looking for new ideas in the theory of the atomic structure". Such ideas were indeed formulated 20 years later!

#### 2.2.2 Interatomic Exchange

As a rule, magnetic atoms have several valence electrons, therefore the question of exchange interaction arises. This case differs from the above-mentioned in the first place in the fact that binding energy includes only interaction between the electrons

$$\mathcal{H}_{ab} = e^2 / R_{12} \tag{2.2.7}$$

and in the second place due to the fact that the coordinate terms of various quantum-mechanical states of electrons of a single atom m and  $n - \varphi_m(\mathbf{R})$  and  $\varphi_n(\mathbf{R})$  are orthogonal. Therefore their overlap integral equals zero and the expression (2.2.3) is reduced:

$$E_{\pm} = A \pm B \ . \tag{2.2.8}$$

Then from (2.2.4, 5) follows a simple expression for J:

$$J = 2B = \int \rho^*(\mathbf{R}_1)(e^2/R_{12})\rho(\mathbf{R}_2) d\mathbf{R}_1 d\mathbf{R}_2 ,$$
  

$$\rho(\mathbf{R}) = \varphi_m(\mathbf{R})\varphi_n^*(\mathbf{R}) .$$
(2.2.10)

Employing the Fourier transform we can obtain

$$J = \frac{e^2}{2\pi^2} \int \frac{|\rho(\mathbf{k})|^2}{k^2} d\mathbf{k} . \tag{2.2.11}$$

Thus, the interatomic exchange is a ferromagnetic one, and it brings about parallel spin orientation of the valence electrons of one atom. Hence follows the well-known  $Hund\ rule$ : among all the states of an atom with the same configuration (the number of valence electrons) the state with the maximum possible spin will have the lowest energy value. The exchange integral (2.2.10) has no small factor (due to the overlap of the wave functions). Its order of magnitude approximates the Coulomb energy of the two electrons on the same center, i.e. it has an atomic scale of  $1\ eV$ . This contribution to the energy is usually much greater than all the others and the magnetic moments of the valence electrons may be considered rigidly connected to one another. They are manifested in interatomic interactions as a single big magnetic moment with respective atomic spin S > 1/2.

#### 2.2.3 Interatomic Exchange of Large Spins

Consider the exchange interaction of two atoms in the state with a total spin  $S_a > 1/2$  and  $S_b > 1/2$ . The wave functions are antisymmetric under rearrangement of N electrons on the atom a and N electrons on the atom b. Using these, a completely antisymmetric wave function must be constructed. Here each of the allowed types of rearrangement symmetry of the space wave function is associated with a certain value of the total spin S (for details see, for instance, [2.12]). Consequently, each S is associated with its own energy value of the two-atom system E(S). The total spin of the system may take  $2S_b + 1$  values (for definiteness we take  $S_a > S_b$ ) from  $S_a - S_b$  to  $S_a + S_b$  and accordingly the energy of the two atoms can assume 2S + 1 values. They can be described by an effective spin Hamiltonian having  $2S_b + 1$  constant:

$$E(S) = E - J_{ab}^{(1)} S_a S_b - J_{ab}^{(2)} (S_a S_b)^2 - \dots - J_{ab}^{(2S)} (S_a S_b)^{2S_b} . \quad (2.2.12)$$

This implies that the Hamiltonian of the exchange interaction of two multielectron atoms, generally speaking, can have no simple Heisenberg form (2.2.6). It must be emphasized, though, that to the first approximation in the overlap of the wave functions of atoms a and b (when only the interatomic exchange of a simple electron pair is allowed for) all the  $J_{ab}^{(n)}$  except  $J_{ab}^{(1)}$  are equal to zero (Nedlin [2.18]). For this reason, the Heisenberg approximation (2.2.6) can generally be used also for the exchange interaction between multielectron atoms. Only in some rare cases the term proportional to  $J_{ab}^{(2)}$  (called the biquadratic exchange) has to be taken into account.

#### 2.2.4 Indirect Exchange Interactions

It must be noted that in magnetic dielectrics, paramagnetic ions usually are not nearest neighbor, but are separated by a diamagnetic ion (oxygen, fluoride). Therefore their wave functions do not overlap and the above discussed direct exchange interaction is absent.

Nevertheless, exchange interaction arises in higher orders of the perturbation theory due to the overlap integrals between the wave functions of the paramagnetic and diamagnetic ions. One of the mechanisms of such an exchange suggested by Kramers in 1934 and Anderson in 1963 (see, e.g. [2.9, 11, 12) is due to the mixing of states of the magnetic and nonmagnetic ions. Indeed, electrons are not fully localized on the ions even in ion crystals. Therefore there is a non-zero probability of the transition of one of the electrons belonging to the diamagnetic ion c (it will be denoted 1c) to the neighboring paramagnetic ion a. Since the initial Hamiltonian of the system is independent of spins, this transition will not be accompanied by an overturn of the spin. As a result, the spin of the electron 1c remains antiparallel to the spin of the second, already unpaired electron of the ion c (electron 2c). Thus, two exchange interactions (namely the exchange of the unpaired electron of the ion a with the incoming 1c and the exchange of the electron of the other paramagnetic ion b with the unpaired electron 2c in the ion c) result in the dependence of the total energy of the three ions a-c-b on the relative orientation of the spins  $S_a$  and  $S_b$ . This suggests that the exchange interaction of the paramagnetic ions a and b separated by the diamagnetic ion c does exist. Such interaction was named the super-exchange or indirect exchange. There are other mechanisms of indirect exchange corresponding to different terms of the perturbation theory for the energy of the three ions a - c - b. (See, for instance, [2.12, 17, 20], etc.).

Obviously, the calculation of the exchange integrals for particular crystals will be too cumbersome a task requiring computer processing, data bases on ion spectroscopy for a reasonable choice of the initial wave functions, etc. All this, of course, is beyond the scope of our work. Henceforth the exchange integral  $J_{ab}$  between the ions of the ions a and b of the crystal will be taken to be a phenomenological constant whose value could be experimentally obtained. Studying nonlinear properties of the spin waves in the present book we shall proceed from the Heisenberg approximation of the exchange Hamiltonian of the magnetic:

$$\mathcal{H}_{\rm ex} = \sum J_{ab} S_1 S_2 \ . \tag{2.2.13}$$

All of the above should not be understood as the derivation of the formula for  $\mathcal{H}_{ex}$ . We intended to illustrate the only following two facts: firstly, that the exchange interaction has a simple nature: it is simply the part of the Coulomb interaction between the electrons due to the Pauli principle;

secondly, that the actual calculation of the exchange integrals in real crystals is not only difficult but unnecessary for the investigation of nonlinear properties of the spin wave.

#### 2.2.5 Relativistic Interactions

The exchange interaction is the strongest, but not the only interaction between the magnetic ions of the magnets. Of fundamental importance are also relativistic interactions: dipole-dipole interactions between the magnetic moments of the electrons and the spin-orbit interaction between the spin and orbital moments of the electron. In the first place, these result in an effective anisotropy energy which determines the energetically advantageous directions of magnetization about the crystallographic axes. In the second place, they lead to processes where the number of the spin waves is not conserved. This results in the complete thermodynamic equilibrium in the system. Later, these cases will be discussed in more detail. Now we shall only write down and consider the expression for the energy of those interactions. We start from the magnetic dipole-dipole interaction:

$$\mathcal{H}_{dd} = 2\mu_{B}^{2} \sum_{a,b} \frac{1}{R_{ab}^{5}} \left[ (\boldsymbol{S}_{a} \boldsymbol{S}_{b}) R_{ab}^{2} - 3(\boldsymbol{S}_{a} \boldsymbol{R}_{ab}) (\boldsymbol{S}_{b} \boldsymbol{R}_{ab}) \right]. \tag{2.2.14}$$

Here, as above,  $\mathbf{R}_{ab} = \mathbf{R}_a - \mathbf{R}_b$  is the distance between the atoms a and b. The energy of the dipole–dipole interaction calculated per atom is  $4S^2\mu^2/a$ , where a denotes the lattice constant. The corresponding value of the magnetic ordering temperature is of the order of 0.25 K. It is much less than the experimentally observed Curie temperature due to exchange interaction, which is normally between 10 and 1000 K.

A further type of relativistic interaction is the *spin-orbit interaction* whose operator has the form:

$$\mathcal{H}_{so} = \lambda(LS) \ . \tag{2.2.15}$$

Here L and S designate the operators of the mechanical and spin atomic moments, and  $\lambda$  is the coupling parameter. The order of magnitude of  $\lambda \simeq (v/c)^2 E_a$ , where v is the characteristic velocity of an electron in an atom, c is the velocity of light, and  $E_a$  is the atomic energy, of the order of one Rydberg, i.e. 10 eV. Ordinarily,  $\lambda \simeq 10^{-2}$  eV. The energy (2.2.15) is determined by orientation of S relative to L. On the other hand, the energy of an ion in the "crystal" electric field depends upon the orientation of L relative to the lattice, since different L are associated with different wave functions. Therefore, in the second order of the perturbation theory with respect to the Hamiltonian (2.2.15) the energy of the ion becomes dependent on its spin orientation about the crystallographic axes. It can be described by the effective spin Hamiltonian

Here  $\lambda_{ij}^{(2)} \simeq \lambda^2/\Delta$ , where  $\Delta$  is the splitting energy of the levels with different L by the crystal field. Since  $\Delta \simeq 1$  eV,  $\lambda^{(2)} \simeq 10^{-4}$  eV  $\simeq 1$  K. If the ground state of the ion is the singlet (L=0), then the value of  $\lambda^{(2)}$  is lower still, because anisotropy arises due to the admixture of states with L=0 in the ground state. As a result, in various crystals  $\lambda^{(2)} \simeq 0.01-1$  K, which corresponds to the effective field of the crystallographic anisotropy  $\mathcal{H}_a \simeq 0.1 \div 10$  kOe. In uniaxial crystals:

$$\lambda_{ij}^{(2)} = \lambda_1 \delta_{ij} + \lambda_2 \delta_{zz} .$$

For  $\lambda_2 < 0$  the anisotropy field tends to orient spins along the z-axis. Then the crystal is said to have the anisotropy of the "easy-axis type". At  $\lambda_2 > 0$  the anisotropy orients the spins perpendicular to the z-axis. This is referred to as anisotropy of the easy-plane type.

If the nearest neighbors of the ion have cubic symmetry then  $\lambda_2 = 0$  and  $\mathcal{H}_{\text{eff}}^{(2)} = \text{const.}$  Then there is no crystallographic anisotropy in the second order of the perturbation theory in the spin-orbit interaction (2.2.15). It arises only in the fourth order. In this case

$$\mathcal{H}_{\text{eff}}^{(4)} = \sum_{ijkl} \lambda_{ijkl}^{(4)} S_i S_j S_k S_l , \qquad (2.2.17)$$

where  $\lambda^{(4)} \simeq \lambda^4/\Delta^3 \ll \lambda^{(2)} \ll \lambda$ . The corresponding field of the cubic anisotropy will be less still than the field of a uniaxial anisotropy:  $H_a$  is of the order of single to tens of Oersteds. Depending on the sign of some combination of constants in (2.2.17) the easy axis will be either the direction [111] or [100].

In conclusion it must be noted that allowing for the overlap of the wave functions of the ions as well as for the spin-orbit interaction results in the effective Hamiltonian of the form of (2.2.16) where spins belong to the neighboring atoms a and b, i.e.  $\mathcal{H}_D \simeq S_{ai}S_{bj}$ . As has already been noted, such an interaction can lead to weakly noncollinear sublattices [2.20].

2.3 Energy of

## 2.3 Energy of Ferromagnets in the Continuum Approximation

Recall that in the spin system of the magnetodielectrics the strongest interaction is the exchange interaction which provides for the very existence of magnetic order. The main part of this interaction is described by the Heisenberg Hamiltonian  $\mathcal{H}_{\rm ex}$  (2.2.13). In the dynamics of long spin waves the dipole–dipole interaction  $\mathcal{H}_{\rm dd}$  (2.2.14) is also of great importance. The interaction of the magnetic moments of electrons with a homogeneous external magnetic field  $\boldsymbol{H}$  must also be taken into account:

$$\mathcal{H}_m = -2\mu_{\rm B} \sum_a (\boldsymbol{H} \boldsymbol{S}_a) \ . \tag{2.3.1}$$

In the previous section it has been shown that the spin-orbit interaction results in the emergence of the effective energy of the magnetic anisotropy. In uniaxial crystals (see (2.2.16)):

$$\mathcal{H}_a^{(2)} = \lambda^{(2)} \sum_{a} (S_{a,z})^2 . \tag{2.3.2}$$

In ferromagnets with cubic symmetry (see (2.2.17)):

$$\mathcal{H}_a^{(4)} = \lambda^{(4)} \sum_{a} \left[ (S_{a,x})^4 + (S_{a,y})^4 + (S_{a,z})^4 \right]. \tag{2.3.3}$$

These expressions contain only the terms of the second and fourth orders of the perturbation theory in the spin-orbit interaction. Sometimes still weaker terms of sixth order are taken into account. In uniaxial hexagonal crystals such terms bring about the anisotropy in the basal plane perpendicular to the 6-fold rotational axis of symmetry.

The total Hamiltonian of the ferromagnet spin system contains, generally speaking, all the above-mentioned terms:

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{dd}} + \mathcal{H}_m + \mathcal{H}_a . \tag{2.3.4}$$

This expression does not contain the interaction of the spin subsystem of a crystal with its other subsystems – phonons, excitons, etc.

We now turn to a discussion of the classical approximation for the ferromagnetic energy assuming the magnetic moments of electrons on the neighboring sites to be almost parallel. Formally, the procedure must be as follows. First, the classical expression for the ferromagnetic energy must be written, which corresponds to the Hamiltonian (2.3.4). Second, the notion of the averaged density of the magnetic moment M(r) must be introduced. To this end the magnetic moment of the electron must be "spread" over the unit cell. Third, we must change from a summation over the sites to an integration over space.

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This procedure can be most easily performed for the part of the Hamiltonian containing a single summation over sites. It is from (2.3.1-3) that the well-known expressions for  $W_m$  and  $W_a$  are most readily obtained:

$$W_m = -\int \boldsymbol{H} \boldsymbol{M}(\boldsymbol{r}) d\boldsymbol{r} , \qquad W_a^{(2)} = \frac{\lambda^{(2)} v_0}{4\delta \mu_B^2} \int M_z^2(\boldsymbol{r}) d\boldsymbol{r} , \qquad (2.3.5a, b)$$

$$W_a^{(4)} = K_4 \int [M_x^4(\mathbf{r}) + M_y^4(\mathbf{r}) + M_z^4(\mathbf{r})] d\mathbf{r}, \quad K_4 = (\lambda^4 v_0^3 / 16 \mu_{\rm B}^4) . (2.3.6)$$

M(r) here denotes the density of the magnetic moment,  $K_4$  is the cubic constant of the 4-th order anisotropy, and  $v_0$  designates the volume of the unit cell. The transformation of (2.2.13) for the exchange energy proves somewhat more complicated. Here only the final expression for  $W_{\rm ex}$  will be shown. The reader can do the necessary calculations independently, or can see it, for instance, in [2.8]. The result is

$$W_{\rm ex} = \frac{\alpha_{ik}}{2} \int \frac{\partial M_j}{\partial x_i} \frac{\partial M_j}{\partial x_k} d\mathbf{r} , \quad \alpha_{ik} = \frac{1}{8\mu_{\rm B}^2} \sum_{\mathbf{n}} J(r_{\mathbf{n}}) r_i(\mathbf{n}) r_k(\mathbf{n}) . \quad (2.3.7)$$

Here  $r_j(\mathbf{n})$  is the *i*-projection (i=x,y,z) of the  $\mathbf{r}$ -coordinate of the magnetic atoms  $\mathbf{n}$  in the lattice. Expressions (2.3.5-7) were derived under the assumption that the distance l over which the exchange interval is essentially changed is small in comparison with the distance at which an essential change  $\mathbf{M}(\mathbf{r})$  takes place.

Note that the expression (2.3.7) for the exchange energy has a great generality. To a great extent it is independent of model assumptions: spin magnitude, dependence of the exchange integral on the distance between atoms, etc. It may be treated as a phenomenological expression for the nonhomogeneous exchange energy. Indeed if it is assumed, first, that this energy is independent of magnetization orientation about the crystallographic axis, second, that inversion is an element of crystal symmetry and, third, that there exists a quadratic dependence of the energy on magnetization, then from symmetry considerations the expression (2.3.6) can be obtained. The proof of this simple statement will not be given here. The reader can find it in [2.8]. We shall only note that the first of the above assumptions is general and follows from the invariance of the exchange interaction with respect to the rotation of all spins. The second assumption is based on the fact that such inversion is characteristic of most magnetically ordered dielectrics. As for the last assumption, strictly speaking it is correct only for the case S = 1/2 when the exchange interaction operator has the Heisenberg form (2.2.13). Under S > 1/2 the approximation (2.2.13) sometimes proves inadequate and the additional term must be allowed for, i.e., the so-called "biquadratic exchange" whose operator is given by (2.2.12). There is a corresponding additional term in the energy of the inhomogeneous exchange:

$$W_{\rm ex}^{(4)} = \frac{\bar{\alpha}_{ik}}{2} \int M_l \frac{\partial M_l}{\partial x_i} M_m \frac{\partial M_m}{\partial x_k} d\mathbf{r} , \qquad (2.3.8)$$

$$\bar{\alpha}_{ik} = \frac{1}{8\mu_{\rm B}^4} \sum_{a} J^{(2)}(a) a_i a_k \ . \tag{2.3.9}$$

The expression (2.3.9) is obtained under the assumption that only the "nearest neighbor" interactions are essential. Than  $J^{(2)}(\boldsymbol{a})$  is biquadratic excange integral  $J^{(2)}(\boldsymbol{r})$  (2.2.12). For the general case the expression (2.3.8) can be obtained from symmetry considerations, thus taking the exchange interaction to be invariant with respect to the rotations of  $\boldsymbol{M}$  and the presence of inversion.

Now we consider the energy of dipole-dipole interactions. The following macroscopic energy corresponds to the Hamiltonian  $\mathcal{H}_{dd}$  (2.2.14):

$$W_{\rm dd} = \frac{v_0^2}{2} \sum_{a \neq b} \frac{1}{R_{ab}^5} \left[ (\boldsymbol{M}_a \cdot \boldsymbol{M}_b) R_{ab}^2 - 3(\boldsymbol{M}_a \cdot \boldsymbol{R}_{ab}) (\boldsymbol{M}_b \cdot \boldsymbol{R}_{ab}) \right] . (2.3.10)$$

Passing to integration in (2.3.10) is not so trivial, though the resulting expression for  $W_{\rm dd}$  is simple:

$$W_{\rm dd} = -\frac{1}{2} \int \left[ \mathbf{M} \cdot \mathbf{H}_m + \frac{4\pi M^2}{3} + a_{ik} M_i M_k \right] d\mathbf{r} ,$$
 (2.3.11)

$$a_{ik} = v_0 \sum_{b} \frac{\partial^2 r}{\partial R^i_{ab} \partial R^k_{ab}} \left(\frac{1}{R_{ab}}\right). \tag{2.3.12}$$

Here  $H_m$  is the static magnetic field due to the magnetic moment M(r). Explicit and detailed derivation of these expressions is presented in [2.8]. Therefore here we shall discuss only the physical meaning of these expressions. The first term in (2.3.11) is a well-known expression of the magnetic energy under continuous space distribution of the magnetic dipoles. The others term account for the discrete structure of the magnets. They are due to the difference between the true value of the magnetic field in dipole locations and the averaged value  $H_m$ . The second term in (2.3.11) approximately allows for this difference, as it is done in the calculation of Lorentz-Lorenz correction to the permittivity of the crystal(see [2.18]). Specifically, in calculating the magnetic field H acting upon the dipole, the dipole is taken to be inside the spherical cavity (the Lorentz sphere) in the continuum with a continuous distribution of the magnetization M. It can be shown (see, e.g., [2.3]) that

$$H = H_m + 4\pi M/3 . (2.3.13)$$

Taking this into account, it becomes clear that the first two terms in (2.3.11) are simply the energy of the dipole interaction  $-M \cdot H/2$ . Finally, the last

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term in (2.3.11) allows for the difference between the real crystal discreteness and the approximate model using the Lorentz sphere. From symmetry considerations in cubic crystals  $a_{ij} = a\delta_{ij}$ . On the other hand, it follows from (2.3.12) that  $\text{Tr}\{a_{ij}\}=0$ , and, as a result, a=0. This means that for cubic crystals the results of the approximate model with the Lorentz sphere are exact. In uniaxial crystals at the same time  $a_{ik}=a_i\delta_{ik}$ ,  $a_z\neq a_x=a_y$ , and the last term in (2.3.11) does not become zero. It can be represented as:

$$-\frac{a_z - a_x}{2} \int M_z^2(\mathbf{r}) d\mathbf{r} - \frac{a_x}{2} \int M^2(\mathbf{r}) d\mathbf{r} . \qquad (2.3.14)$$

The form of the first term coincides here with the expression (2.3.5) for  $W_a^{(2)}$  and therefore represents the dipole–dipole contribution to the energy of crystallographic anisotropy. Henceforth this term will be considered to have been accounted for in (2.3.5) by corresponding renormalization of the coefficient  $\lambda^{(2)}$ . The remaining terms in (2.3.11) and (2.3.14) proportional to  $M^2(\mathbf{r})$  represent the constant contribution to the energy W which does not depend on the orientation of M and is of no interest for us. Therefore in (2.3.11) we can retain only the first term.

For this term to be expressed only in terms of magnetization the following equations of magnetostatics must be solved

$$\operatorname{curl} \boldsymbol{H} = 0 , \quad \operatorname{div} \boldsymbol{B} = 0 . \tag{2.3.15}$$

We will be interested in the following two cases:

- 1. The Plane Wave:  $M(r) = \langle M \rangle + [m_k \exp(ikr) + \text{c.c.}]$ , the wavelength of the spin wave  $2\pi/k$  being much less than the sample size.
- 2. The sample is shaped as an ellipsoid and M(r) = M = const. As is well known, (2.3.15) has exact solutions in both cases. In the first case

$$\boldsymbol{H}(\boldsymbol{r}) = [\boldsymbol{H}_m(\boldsymbol{k}) \exp(i\boldsymbol{k}\boldsymbol{r}) + \text{c.c.}], \quad \boldsymbol{H}(\boldsymbol{k}) = -4\pi \boldsymbol{k} (\boldsymbol{k} \cdot \boldsymbol{m}_{\boldsymbol{k}})/k^2.$$

In the second case  $\boldsymbol{H}_{m,i} = -4\pi N_{ik} M_k$ ,  $\text{Tr}\{N_{ij}\} = 1$ , where  $N_{ij}$  is the tensor of demagnetizing factors, depending on the shape of the ellipsoid. Using these results the energy of the dipole-dipole interaction (2.3.11) can be expressed in terms of  $\boldsymbol{M}$ . To this end, represent  $\boldsymbol{M}(\boldsymbol{r})$  as an expansion in terms of plane waves

$$M(r) = \langle M \rangle + \sum_{k} m(k) \exp(ik \cdot r)$$
 (2.3.16)

Here we take M(k) = 0 at k = 0, and the mean magnetization value in the sample is given by the formula

$$\langle \boldsymbol{M} \rangle = \frac{1}{V_s} \int \boldsymbol{M}(\boldsymbol{r}) d\boldsymbol{r} ,$$
 (2.3.17)

where  $V_s$  is the volume of the sample. Substituting the expansion (2.3.16) into the formulae to (2.3.11) and using the expression for  $\boldsymbol{H}(\boldsymbol{r})$  and for  $\boldsymbol{H}_{m,i}$  we obtain

$$W_{\rm dd} = 2\pi V N_{ik} \langle M_i \rangle \langle M_k \rangle + 2\pi V \sum_{\mathbf{k}} [\mathbf{k} \cdot \mathbf{m}(\mathbf{k})] [\mathbf{k} \cdot \mathbf{m}(-\mathbf{k})] / k^2 . \quad (2.3.18)$$

Recall that this expression was derived under the assumption that the minimum linear dimension of the sample is much greater than  $2\pi/k$ . This means that we do not allow for the Walker modes which are magnetostatic eigenmodes of magnetization oscillations with characteristic dimension on the order of the sample size (see, for instance, Sect. 11 in [2.8]). The only Walker mode taken into account by the first term in (2.3.18) is the uniform precession of magnetization (UP). This limitation of (2.3.18) is, however, not very important since in most experiments treated any modes below the Walker modes (except for UP) are not excited.

Summing up the present section we may say that the energy of the ferromagnet W is the sum

$$W = W_m + W_a + W_{\rm ex} + W_{\rm dd} , \qquad (2.3.20)$$

where  $W_m$  is the energy of interaction with the external field,  $W_a$  is the energy of magnetic anisotropy,  $W_{\rm ex}$  is the exchange energy and  $W_{\rm dd}$  is the energy of the dipole–dipole interaction, with expressions given by respectively by (2.3.5–9, 18). These expressions will be used for the calculation of the spin waves' dispersion law and the functions characterizing their interaction.

### 2.4 Magnetic and Crystallographic Structure of Some Magnets

We shall not give a detailed description or classification of magnetic crystals. The reader is referred to the specialized treatises [2.22], reviews [2.11], chapters in books [2.9]. In the present book we give only the structures of those magnets that are most often employed in experiments.

#### 2.4.1 Crystals with Spinel Structure

Such crystals are of special interest, since they were used for the first verification of the Néel theory of ferromagnetism, which generalized the Weiss theory of the molecular field to the case of several magnetic sublattices. The spinel structure is typical of dioxides of transition metals having the chemical formula  $A^{2+}B_{2}^{3+}O_{4}$ . Here  $A^{2+}$  and  $B^{3+}$  stand for two- and three-charge ions of the metals. Spinels are also formed by almost densely packed oxygen.

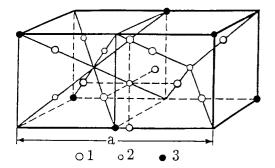


Fig. 2.1. Two of eight octants of the elementary cell of spinels: (1) oxygen, (2) metal in the octahedral interstices, (3) metal in the tetrahedral interstices

The ions of a metal are located in octahedral and tetrahedral interstices surrounded respectively by six and four oxygen ions (Fig. 2.1). The unit cell of the spinel consists of eight basic formula units (AB<sub>2</sub>O<sub>4</sub>) and comprises 32 oxygen atoms forming 16 octahedric and 8 tetrahedric interstices. They are designated respectively by the letters d and a. The cell a is a cube of edge length  $a_0 = 8.5 \,\text{Å}$ ) consisting of eight octants (small cubes  $a_0/2$  on edge) of two types, differing in the arrangements of their ions. The octants of different types have a common edge. In the so-called *normal spinels* the cations A are located in the tetrahedral sites and the cations B are in the octahedral sites. This is characteristic, for instance, of the ferrites  $\text{ZnFe}_2\text{O}_4$ ,  $\text{CdFe}_2\text{O}_4$ .

In the *inverted spinels* the cations A and half of the cations B are located in octahedrons and the other half of the cations B are in tetrahedrons. This refers, for example, to manganese, lithium, and nickel ferrites. The ferrite MgFe<sub>2</sub>O<sub>4</sub> has an intermediate (between the normal and the inverted spinels) type of cation localization.

If both metals A and B are paramagnetic the spinel is as a rule ferrimagnetic with predominating intersublattice exchange  $|J_{AB}| \gg |J_{AA}|, |J_{BB}|$ , with the antiferromagnetic sign. In this case the magnetizations of the sublattices A and B are antiparallel.

Normal spinels ZnFe<sub>2</sub>O<sub>4</sub> and CdFe<sub>2</sub>O<sub>4</sub> where the magnetic ions occupy only equivalent (octahedric) positions are antiferromagnetic. This is due to the antiferromagnetic sign of the B–B exchange subdividing the ensemble of the sites B into two magnetic sublattices.

#### 2.4.2 Crystals with Garnet Structure

The Yittrium–Iron Garnet (YIG) is a wonderful phenomenon. Its role in the physics of magnets is comparable to the part germanium played in the physics of semiconductors and quartz in acoustics of crystals. This can be explained by the following. Firstly, it has a high Curie temperature  $T_c=560~\rm K$  which enables one to experiment at room temperature. On the other hand, although the unit cell consists of 80 atoms, each of which must assume its unique "correct" position, the technology of YIG crystal growing has been developed so well that the sound damping decrement is less than in quartz crystals. Finally it has the narrowest known line of ferromagnetic resonance and the smallest damping decrement of the spin waves. All this makes it indispensable not only in SHF engineering but also in the experimental physics of magnets studying new effects and phenomena.

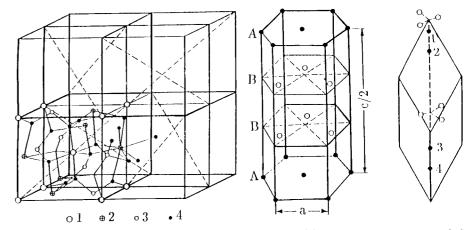


Fig. 2.2. (left) The elementary cell of YIG: (1), (2) and (3) are the positions a, c and d respectively, (4) are the oxygen ions

Fig. 2.3. (middle) One half of the CsMnF<sub>3</sub> elementary cell (A and B are the Mn ions); (right) an elementary cell of Fe<sub>2</sub>O<sub>3</sub> and Cr<sub>2</sub>O<sub>3</sub>: (1), (2), (3) and (4) are the ions of Fe or Cr

The YIG unit cell is a half-cube  $a_0 = 12.38$  Å on edge consisting of 4 identical octants, i.e. small cubes  $a_0/2$  on edge comprising one formula unit (Fig. 2.2). The arrangement of atoms in the octant has no cubic symmetry. In the unit cell, however, the orientation of octants makes it practically cubic of the  $O_h$  symmetry group. The apices and centers of every octant are occupied by Fe<sup>3+</sup> cations surrounded by octahedrons (the so-called a-site). Their coordinates in the first octant can be written as (0,0,0); (0,1/2,1/2); (1/2,0,1/2); (1/2,1/2,1/2,1/2); (1/4,1/4,1/4). A unit cell comprises 8 a-sites. There are also 12 tetrahedral sites (12d) occupied by

Fe<sup>2+</sup> anions and 12 dodecahedral sites (12c), occupied by Y anions. Their coordinates in the first octant of the site (12d) are (3/8, 0, 1/4); (1/4, 3/8, 0); (0, 1/4, 3/8); and in the site (12c)(1/8, 0, 1/4); (1/4, 1/8, 0); (0, 1/4, 1/8).

The oxygen anions occupy the common sites. The sites of all the 40 atoms in a unit cell (of  $Y_3 Fe_2^{3+} Fe_3^{2+} O_{12}$ ) are considered, e.g., in [2.12]. Thus, there are two non-equivalent sites of paramagnetic ferrum anions in YIG, i.e. a and d. Therefore, in a rough approximation YIG is sometimes considered a ferrimagnet with 2 sublattices. This approximation, however, has no field of applicability (Kolokolov et al. [2.21, 22]). At T < 150 K only one branch of the spin wave spectrum is excited and YIG can be considered a ferromagnetic. At large T the excitations of at least four or more spectrum branches must be taken into account.

In addition to YIG there are rare earth ferrogarnets  $R_3Fe_5O_{12}$ , where R=Sm,Dy,Yb,Lu,Tm. Their rare earth sublattices are also magnets and the number of magnetic sublattices is larger than in YIG.

#### 2.4.3 Crystals with Hexagonal Structure

Such crystals are characterized by the space group  $D_{6h}^4$ . First we shall describe the antiferromagnet (AFM) CsMnF<sub>3</sub>, which has been the object of very interesting experiments by Prosorova, Kotyuzhansky et al. studying the parametric excitation of the spin waves. Its unit cell contains 6 formula units. Mn ions occupy two non-equivalent sites A (2 ions) and B (4 ions) centrally positioned in octahedrons formed by fluorine ions (see Fig. 2.3). Site A: (0, 0, 1/2). Sites B are : (1/3, 2/3, i); (1/3, 2/3, -i); (2/3, 1/3, 1/2 + i)i); (2/3, 1/3, -i). Thus, the Mn layers A and B alternate as ABBABBA. In every plane the ordering is ferromagnetic; moments are in the plane. The moments of all neighboring planes are antiparallel. Thus, CsMnF<sub>3</sub> is a pure AFM, unlike its compounds RbNiF<sub>3</sub>, TlNiF<sub>3</sub>, CsFeF<sub>3</sub> which have the same structure, but are ferrimagnets. In these crystals intensive antiferromagnetic A-B exchange and a weak ferromagnetic B-B exchange are observed. Therefore these crystals may be treated as quasi-two-dimensional ones formed by triples, i.e. by groups of three strongly coupled BAB-planes: (BAB)(BAB)... All the moments are in the basal plane; magnetic ordering of the planes is of the type (+-+)(+-+)(+-+).... In the structure under consideration, the crystallographic surrounding of each magnetic ion has a six-fold symmetry axis. Anisotropy in the basal plane arises only in the sixth order of perturbation theory in the spin-orbit interaction and thus appears rather small: H=1 Oe. Also, uniaxial anisotropy (of the easy-plane type) arising in the second order of perturbation theory is rather big – the field of anisotropy is of the order of 10 kOe.

#### 2.4.4 Crystals with Rhombohedral Structures

The two most remarkable representatives are  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> and Cr<sub>2</sub>O<sub>3</sub>. Dzyaloshinsky's theory, stating the connection of weak ferromagnetism arising in AFMs with magnetic ordering symmetry was verified employing these crystals by authors of [2.12, 17]. The space group of these crystals is  $D_{3d}^6$ . The unit cell comprises two formula units, i.e. four magnetic ions located on the axis of the 3-fold rotation symmetry (Fig. 2.3, right). They are antiferromagnetically ordered. Under a certain sequence of alternating spin signs in a unit cell weak ferromagnetism is possible, only when the moments are in the basal plane [2.12]. These conditions exist in MnCO<sub>3</sub> at temperatures 950 > T > 250 K. Indeed, this substance has been experimentally proven to have a weak ferromagnetic moment within the above mentioned temperature range. As for Cr<sub>2</sub>O<sub>3</sub>, the spin alternation in it is of another kind and in full accordance with the theory that it contains no magnetic moment  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> (AFM with anisotropy of the easy-plane type) is also a rhombohedric crystal. Its Néel temperature is 32 K. This magnetic is employed in experiments on the parametric excitation of spin waves. Unlike CsMnF<sub>3</sub> it is characterized by weak ferromagnetism.

No detailed description of the other magnetic structures will be given. We shall only name some crystals of perovskite structure (CaTiO<sub>3</sub>) whose composition can be represent as ABX where X is oxygen or fluorine, and A and B designate metals [2.12]. These are YFeO<sub>3</sub>, KMnF<sub>3</sub>, RbCoF<sub>3</sub>. Magnetic crystals with a structure of rock salt are worth mentioning, i.e. AFM MnO, FeO, CoO, NiO, and ferromagnets EuO, EuS, EuSe (at T < 2.8 K). It must be emphasized that in the long list of the named crystals only the three europium compounds are simple ferromagnets with one magnetic lattice.