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## New physical phenomena caused by magnetoelectric and antiferroelectric interactions in magnets

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Abstract. The review examines the dynamics of a special class of magnetic substances (magnetically ordered crystals) in which the magnetic atoms do not coincide with the symmetry center (provided that such a center exists in the crystal). Research into the magnetoelectric and antiferroelectric interactions in such magnets has led to a new section in the spin dynamics of both the electrical and nuclear subsystems. The review is based primarily on the latest works (2001 – 2004) done by theoretical physicists from the Ural region. Several results of the pioneering works (1988 – 1990) of Ukrainian physicists, who opened this new section in spin dynamics, are also discussed. All this research has made it possible to compare the predicted effects for different crystal systems and thus has provided a more meaningful arrangement of the desired experiments.

### 1. Introduction

Magnetically ordered substances are lumped together as magnets irrespectively of the type of their ordering: ferromagnetic (FM), antiferromagnetic (AFM), or ferrimagnetic (FIM). It must be immediately noted that after the well-

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known works of Dzyaloshinskii [1] it proved convenient and to a certain extent even important in the physical and symmetrical aspects to describe some magnets not in terms of the magnetizations  $\mathbf{M}_{\nu}(\mathbf{r})$  ( $\nu=1,2,3,\ldots$ ) of their sublattices, but in terms of the vector of total local magnetization  $\mathbf{M}(\mathbf{r})$  and the vectors (or vector) of antiferromagnetism,  $\mathbf{L}(\mathbf{r})$ . By way of illustration, for a two-sublattice magnetic substance ( $\nu=1,2$ ), the following relations hold true:

$$\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2 \,, \tag{1}$$

$$\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2. \tag{2}$$

Small linear oscillations of magnetic moments are described by spin waves, or magnons. The first theoretical investigation of spin waves (for ferromagnets) was carried out more than 70 years ago by F Bloch [2]. Dozens of monographs and reviews and, possibly, thousands of papers on the subject have appeared since Bloch's paper was published in 1930. An example is a very thorough book written by the present authors in collaboration with colleagues [3] that appeared rather recently (in 2001). However, and this may seem remarkable, the vast literature contains almost no mention of an entire section of magnetodynamics. We are speaking of the dynamic phenomena in magnetic substances whose crystallochemical structure exhibits the property of spatial inversion, i.e., has a center of symmetry  $\overline{1}$ , with the center implementing a nonidentical permutation of magnetic atoms (into each other and not into themselves) of the same position of multiple points.

It is significant that the present review is devoted not to particular problems of magnetodynamics of secondary importance. Instead we focus on new important aspects of the problem that have not found sufficient coverage in such problem-oriented review journals as *Physics – Uspekhi*.

Here are only three main groups of the above-mentioned phenomena that are treated in this review.

First, we discuss a new unusual type of spin wave. A characteristic feature of waves of this kind is that only the antiferromagnetic vector (or vectors) L oscillates [see Eqn (2)], while the magnetization vector **M** defined by equation (1) is at rest. In 2001, one of the present authors (E A T) proposed a special name for such waves, antimagnons (see Ref. [4]). In contrast to other types of magnons (ferromagnons, quasiferromagnons, and quasiantiferromagnons), in which the components of the vector M that are capable of being excited by an alternating magnetic field  $\mathbf{H}(t)$  also belong to the group of oscillation variables, antimagnons can usually be excited only by an alternating electric field  $\mathbf{E}(t)$ . Another characteristic feature of antimagnons is that their natural frequency often has an exchange origin; this, however, is true only of collinear and weakly (relativistically) noncollinear magnetic structures and, generally speaking, is not true of exchangenoncollinear structures (see Section 7).

The excitation of antimagnons and sometimes of other types of magnons (see below) is caused by respective linear (in the field E) magnetoelectric (ME) and antiferroelectric (AFE) interactions of the following forms

$$s_{ijk}M_iL_iE_k$$
, (3)

$$f_{ijk}L_{1i}L_{2j}E_k \quad (\mathbf{L}_1 \neq \mathbf{L}_2). \tag{4}$$

The specific form of the tensors  $s_{ijk}$  and  $f_{ijk}$  is determined by the requirement that expressions (3) and (4) must be invariant with respect to the symmetry elements of the corresponding *space* group of the crystal. The common convention of summation over the twice repeating indices is applied here.

The static (and quasistatic) ME effect (exposed by D N Astrov [5] in 1960), i.e., induction of magnetization M by an electric field, and the inverse effect, namely, induction of polarization P by a magnetic field, have been studied for a long time (e.g., see Refs [6, 7] and the review in Ref. [3]). However, it turned out that the dynamic manifestations of the ME interaction (plus the AFE interaction) are much richer (see the preprint [8]), and the present review is devoted to precisely these manifestations.

Suppose that one of the two magnetic vectors (**M** or **L**) is at rest and determines the ground state (with the lowest possible energy) we are interested in. Then an electric field  $\mathbf{E}(t) \propto \exp{(-\mathrm{i}\omega t)}$  will 'sway' the second vector (within the framework of linear response), with the possibility of the above *purely* AFM oscillations, or antimagnons, being excited in the FM phase.

The second group of phenomena caused by ME and AFE interactions are the waves of antimagnons and other electroactive spin waves coupled with electromagnetic waves, i.e., a magnon-photon resonance in a field  $\mathbf{E}(t)$  of a certain frequency.

If we are dealing with antimagnons, both groups of phenomena belong to a fairly high (exchange) frequency range (however, see Section 7). But there is also a third, low-frequency, group of phenomena related to oscillations of  $\mathbf{M}$  or  $\mathbf{L}$  vectors caused by an electric field  $\mathbf{E}(t)$ . What we have in mind is the excitation of oscillations of nuclear magnetic moments in the magnets by a field  $\mathbf{E}(t)$  of the appropriate frequency  $\omega \approx \omega_n$  [the nuclear magnetic resonance (NMR)

frequency]. The reason for such excitation is that the oscillations of M and L generated by such a field E(t) because of interaction (3) or (4) contribute to the hyperfine field (to its permanent and varying parts), thus leading to a new resonance effect, the nuclear magnetoelectric resonance (NMER) [8], as it is called. Such excitation of NMR by an electric field, predicted theoretically in Refs [8, 9], has not so far been observed in experiments.

A number of other effects related to antimagnons and their interactions (3) and (4) have been studied (or at least mentioned as problems) in Ref. [8], with a consistent transfer from simple cases to more complex ones. It must be noted, however, that it would, generally speaking, be incorrect to claim that the research covered in the present review began with paper [4] published in 2001. Actually, the idea underwent considerable modifications. The thing is that in 1988 several articles appeared in leading Soviet physics journals [10-13] and in a collection of works [14], written by the well-known Ukrainian physicists D A Yablonskii, V N Krivoruchko, V V Eremenko et al. These articles contained some of the main ideas and results we discussed earlier in this review. which, it would seem, could serve as a good starting point for the above-mentioned section of the dynamics of magnetic substances. What was really remarkable and unexpected was that this did not happen: we know of no works of other researchers on this topical problem, and Refs [10-14] were scarcely cited in other works. Here we will not discuss the reason for this. Suffice it to note that the author of Ref. [4] 'rediscovered' some of the ideas and results almost a decade later. With the articles that followed, the author of Ref. [4] and his coauthors were 'lucky' only in the sense that the subject of their research and the methodical approach they used (which is, of course, not that important) differed from those used in Refs [10-14], where only antiferromagnetic structures, such as α-Fe<sub>2</sub>O<sub>3</sub>, Cr<sub>2</sub>O<sub>3</sub>, orthoferrites, and others (with four or more magnetic sublattices) were studied. Contrary to this, the author of Ref. [4] began, as it turned out, with the simplest ferromagnetic structures with two magnetic sublattices (v = 2), and only later did he study more complicated structures (FM, AFM, FIM) with v > 2. Moreover, the area of research widened and finally became a new section of magnetodynamics, which we would like to describe in this review more or less concisely and in plain words.

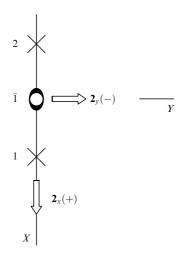
Due to the large volume of material, in citing this or that original paper we are forced to limit ourselves to its simplest part (to an extent that will help judge the important aspects of the topic). For uniaxial crystals, only the easy-axis case is usually described, although in the works cited the results are generally given for the easy-plane state, too.

It is advisable to begin our consideration with the simplest case of a two-sublattice centrally antisymmetric (CAS) ferromagnet, although the corresponding paper [4] was not the first in this area of research.

## 2. A two-sublattice ferromagnet with magnetoelectric interaction

### 2.1 The magnetic structure, the functions describing it, and their transformation table

Let us examine a two-sublattice ferromagnet with the coordinates of the magnetic atoms being 1(x,0,0) and 2(-x,0,0). In Fig. 1, these atoms are depicted as  $\times$ 's. The



**Figure 1.** Two sites (denoted by  $\times$ 's) for position 2i of group *Pmmm*. Also shown are the symmetry elements  $\overline{1}$ ,  $2_x$ , and  $2_y$  — the generators of the group. Note that  $\overline{1}$  and  $2_y$  permute the atoms 1 and 2, while the axis  $2_x$  passing through the atoms leaves each in place.

space symmetry of the corresponding crystal (it is commonly assumed that the magnetic and chemical unit cells coincide) may, for one thing, be described by the Fedorov group **Pmmm**  $\equiv D_{2h}^1$  of a rhombic system. In Fig. 1, this group is represented by three independent symmetry elements (the generators of the group): the symmetry center — space inversion  $\overline{1}$  — and two simple (nonscrew) two-fold axes  $\mathbf{2}_{x} \| X$  and  $\mathbf{2}_{y} \| Y$  [15]. Being elements of a space group, they break down into two types according to their permutation properties (in relation to the selected position of the atoms). The  $2_x$  axis which passes through both atoms permutes each atom into itself:  $\mathbf{2}_x 1 = 1$  and  $\mathbf{2}_x 2 = 2$ , and this is known as an identity permutation. We label such elements by a 'plus' sign in parentheses:  $\mathbf{2}_x \equiv \mathbf{2}_x(+)$ . The elements  $\overline{\mathbf{1}}$  and  $\mathbf{2}_y$  (see Fig. 1) perform a nonidentity permutation (interchanging of the atoms):

$$\overline{\mathbf{1}}(-)1 = 2$$
,  $\overline{\mathbf{1}}(-)2 = 1$ ;  $\mathbf{2}_{\nu}(-)1 = 2$ ,  $\mathbf{2}_{\nu}(-)2 = 1$ . (5)

To distinguish between these elements from the previous ones in their permutation properties, we label them with a 'minus' sign in parentheses.

On the whole, allowing for the permutations (5) of the atoms into positions of multiple points, we can write the elements adopted as the generators of the symmetry group as follows:

$$\overline{\mathbf{1}}(-)\mathbf{2}_{x}(+)\mathbf{2}_{v}(-). \tag{6}$$

We call this the permutation code of the position of the atoms. Sometimes the elements with a 'plus' or a 'minus' sign are said to be 'even' and 'odd', respectively [10-14].

 $^1$  Usually, an international system of notation is used for the symmetry elements and the symbols of the symmetry group: numbers for symmetry axes (including the center of symmetry), and letters for symmetry planes. The same numbers are used to enumerate the atoms in the unit cells, while letters denote other variables, and both may be present in the formulas at the same time. Therefore, to avoid confusion, we use bold type for the symmetry elements (e.g.,  $\overline{1}$  instead of  $\overline{1}$ , m instead of m, etc.). Of course, to a certain extent this violates the common system of notation and may draw unfavorable criticism from the vigilant reader, but we were forced to do this to make the text easier to read.

The set of elements (generators) written in this way in the single-position case under consideration provides all the information about the space group  $(D_{2h}^1)$  we are interested in, which is necessary if we want to find (in terms of the symmetrically arranged vectors  $\mathbf{M}$ ,  $\mathbf{L}$ ,  $\mathbf{E}$ , etc.) the invariant form of the thermodynamic potential, the constitutive tensors, and the like. The elements  $\overline{\mathbf{I}}$ ,  $\mathbf{2}_x$ , and  $\mathbf{2}_y$  act on the vectors  $\mathbf{M}$ ,  $\mathbf{L}$ , and  $\mathbf{E}$  similar to point-group elements, for example:

$$\overline{\mathbf{1}}\mathbf{M} = \mathbf{M}, \quad \overline{\mathbf{1}}\mathbf{L} = \mathbf{L}, \quad \mathbf{2}_{x}L_{y} = -L_{y}, \quad \mathbf{2}_{y}L_{y} = L_{y}.$$
 (7)

If, however, we allow for permutations (5), coded in expression (6) and arising from the need to use a space symmetry group, we arrive at certain alterations of these transformations (this is usually associated with an additional change in sign). For instance, if we allow for relationship (5), we get, accordingly,  $\overline{\mathbf{I}}(-)\mathbf{L} = -\mathbf{L}$  and  $\mathbf{2}_y(-)L_y = -L_y$ . At the same time, one finds  $\mathbf{2}_x(+)L_y = -L_y$  [as in Eqn (7)], and so forth, as for an ordinary point group. The components of the fields  $\mathbf{E}$  and  $\mathbf{H}$  are not affected by the permutation.

It is convenient to tabulate what we have just said about the vectors  $\mathbf{M}$ ,  $\mathbf{L}$ ,  $\mathbf{E}$ , and  $\mathbf{H}$  (see Table 1), in which the numbers +1 and -1 determine whether the sign of the function changes (-1) or does not change (+1) as the symmetry element acts on the function.

**Table 1.** Transformation of the basis vectors **M** and **L**, position 2i of group  $D_{2h}^1$ .

$\Gamma_n$	Dynamical variables	$\overline{1}(-)$	$2_{x}(+)$	$2_{y}(-)$	Static and alternating fields
$\Gamma_1$	$M_{\scriptscriptstyle X}$	+1	+1	-1	$H_{\scriptscriptstyle X}$
$\Gamma_2$	$M_y$	+1	-1	+1	$H_y$
$\Gamma_3$	$M_z$	+1	-1	-1	$H_z$
$\Gamma_4$	$L_{\scriptscriptstyle X}$	-1	+1	+1	
$\Gamma_5$	$L_y$	-1	-1	-1	$E_z$
$\Gamma_6$	$L_z$	-1	-1	+1	$E_y$
		-1	+1	-1	$E_{\scriptscriptstyle X}$

The axial vectors **M** and **H** are transformed in like manner, but the polar vector **E** is transformed differently.

Thus, all six dynamical variables  $(M_x, M_y, M_z, L_x, L_y, L_z)$  that describe the behavior of the magnetic substance occupy different rows  $(\Gamma_1 - \Gamma_6)$  in the table and all transform differently. This is a very important fact, and it is usually said in group-theoretical analysis that the variables  $(M_i, L_j)$  break down into one-dimensional irreducible representations. To each representation, i.e., to each row  $\Gamma_n$ , there corresponds a certain magnetic structure (phase) in which in the ground state only one function out of the all six components of the row in question is not equal to zero. If we were to deal with the sublattice variables  $M_{1i}$  and  $M_{2i}$  (i = x, y, z), in each row there would be a linear combination of these variables.

Thus, it turns out that the vectors **M** and **L** are basis vectors. This is a vivid demonstration of the effectiveness of using the vectors of antiferromagnetism and total magnetization.

Table 1 contains all the information needed to study the problems we are interested in here: we are able to write down invariant expressions for the thermodynamic potential  $\Phi$ , for the constitutive tensors determined by the quantities in Table 1, and so forth.

Moreover, Table 1 makes it possible to single out the sets of oscillation spin-wave variables corresponding to independent oscillation modes (even before the expression for the thermodynamic potential and the equations of motion have been written). Obviously, and we demonstrate this in Sections 2.2 and 4.2, such an approach simplifies the calculations substantially. To obtain a set of spin-wave variables for a phase  $\Gamma_m$  entering Table 1, we must select in this table the rows  $\Gamma_n$  and  $\Gamma_{n'}$  whose products (pairwise from the numbers +1 and -1 that are present in these rows) yield the row  $\Gamma_m$ . Symbolically, this can be represented as follows [3, 16]:

$$\Gamma_n \times \Gamma_{n'} = \Gamma_m. \tag{8}$$

For instance, for the FM structure we are primarily interested in, namely, the phase  $\Gamma_1(M_x)$ , we find two modes  $\Gamma_{23}(M_y,M_z)$  and  $\Gamma_{56}(L_y,L_z)$ . Note that, being eigenmodes, they belong to independent pairs of the oscillation variables  $(M_y,M_z)$  and  $(L_y,L_z)$ . It is these modes that we investigate in the present review.

### 2.2 Thermodynamic potential, equations of motion, and their solution with allowance for ME interaction

Let us first write down the thermodynamic potential density  $\Phi(\mathbf{r})$  for the case of a homogeneous medium in uniform fields. Here, we distinguish at once between two types of interaction in the M, L system: the exchange interaction and the relativistic interaction [17]. The former is determined solely by the angles between the magnetic moments proper (but not by the angles between the magnetic moments and the crystallographic axes), with the result that invariant expressions for this interaction can consist only of scalar products of the vectors M and L, including various powers of these vectors (beginning with the square of a vector). The second interaction is magnetically anisotropic or relativistic (since its coefficients must contain a small relativistic parameter v/c, where v and c are the electron speed in the atom and the speed of light). Actually, today we know that such a decomposition of interactions is not always justified (see the literature cited in Ref. [3]), but here we consider only the simplest case. We also allow for the interaction with the fields **H** and **E**.

Limiting ourselves to the bilinear approximation (in **M** and in **L**) and using Table 1, we find an expression for the potential density that is invariant under transformations belonging to Table 1:

$$\Phi(\mathbf{r}) = \frac{1}{2} \left[ (A_M \mathbf{M}^2 + A_L \mathbf{L}^2) + (K_{M1} M_y^2 + K_{M2} M_z^2) \right. \\
+ (K_{L1} L_y^2 + K_{L2} L_z^2) \right] - M_x H_x \\
- \frac{1}{2M_0} \left[ (s_1 L_y E_y + s_2 L_z E_z) M_x \right. \\
+ (s_3 M_x L_x + s_4 M_y L_y + s_5 M_z L_z) E_x \\
+ (s_6 M_y E_y + s_7 M_z E_z) L_x \right]. \tag{9}$$

Here,  $A_M$  and  $A_L$  are the parameters of the exchange interaction,  $K_{M1(2)}$  and  $K_{L1(2)}$  are the magnetocrystalline anisotropy constants, and  $s_1, \ldots, s_7$  are the appropriate components of the tensor  $s_{ijk}$  in expression (3).

According to Landau's concept, the phase transition into a magnetically ordered state usually proceeds along a single representation. Hence, below we limit ourselves to the analysis of the phases corresponding to one of the irreducible representations.

We decided to start with the phase  $\mathbf{M}^0 \parallel X$  [the representation  $\Gamma_1(M_x^0)$ ]. Hereinafter the 'zero' is usually an upper index and corresponds to the ground state (i.e., the state with the lowest energy). In the ground state, the other components of  $\mathbf{M}$  and  $\mathbf{L}$  are equal to zero, provided there are no fields and no other external interactions:

$$M_x = M_y^0 = 2M_0$$
,  $M_y^0 = M_z^0 = 0$ ;  $L^0 = 0$ ,

where  $M_0$  is the nominal length of the sublattice magnetization vector. We simplify the problem by adopting the very popular equal-modulus model in which it is assumed that  $\mathbf{M}_1^2 = \mathbf{M}_2^2 = M_0^2$  or (in terms of  $\mathbf{M}$  and  $\mathbf{L}$ )

$$\mathbf{M}^2 + \mathbf{L}^2 = (2M_0)^2$$
,  $\mathbf{L}\mathbf{M} = 0$ . (10)

In the ground state, the quantities contained in Eqn (9) must obey the following inequalities:  $A_M < 0$ ,  $A_L > 0$ ,  $K_{M1(2)} > 0$ , and  $K_{L1(2)} > 0$ . Only then will the FM state of interest to us with  $\mathbf{M}^0 \parallel X$  be realized.

In the linear oscillation theory we must then put  $\mathbf{M} = \mathbf{M}^0 + \Delta \mathbf{M}$  and  $\mathbf{L} = \mathbf{L}^0 + \Delta \mathbf{L}$ , thus separating the oscillation part from the ground state and isolating in  $\boldsymbol{\Phi}$  [see Eqn (9)] the part  $\boldsymbol{\Phi}_2$  that is quadratic in  $\Delta \mathbf{M}$  and  $\Delta \mathbf{L}$ , with allowance for formulas (10). Here, the first equality in formulas (10) can be used to exclude the first exchange term (the one with the coefficient  $A_M$ ) from the right-hand side of expression (9). Moreover, the same relation suggests that the field component in expression (9) also contributes through  $M_X$  to  $\boldsymbol{\Phi}_2$ , by virtue of the fact that

$$M_x \approx 2M_0 - \frac{\Delta M_y^2 + \Delta M_z^2 + \Delta L_y^2 + \Delta L_z^2}{4M_0}.$$

Bearing in mind the aforesaid, we find that  $\Phi_2$  breaks down into two independent parts: the purely ferromagnetic part

$$\Phi_{2M} = \frac{1}{2} \left( K_{M1} + \frac{H_x}{2M_0} \right) \Delta M_y^2 + \frac{1}{2} \left( K_{M2} + \frac{H_x}{2M_0} \right) \Delta M_z^2 \quad (11)$$

(containing only  $\Delta M$ ), and the purely antiferromagnetic part

$$\Phi_{2L} = \frac{1}{2} \left( A_1 + \frac{H_x}{2M_0} \right) \Delta L_y^2 + \frac{1}{2} \left( A_2 + \frac{H_x}{2M_0} \right) \Delta L_z^2 
- s_1 \Delta L_y E_y - s_2 \Delta L_z E_z$$
(12)

(which incorporates only  $\Delta L$ ), where

$$A_1 = A_L - A_M + K_{L1}$$
,  $A_2 = A_L - A_M + K_{L2}$ .

This result, namely, the separation of the oscillation part  $\Phi$  into two independent parts  $\Phi_{2M}$  and  $\Phi_{2L}$ , agrees with the representations  $\Gamma_{23}(M_y, M_z)$  and  $\Gamma_{56}(L_y, L_z)$  for the FM phase  $\Gamma_1(M_x^0)$ , obtained earlier [on the basis of formula (8)].

Using the relationship  $\Phi_2 = \Phi_{2M} + \Phi_{2L}$ , we need only solve the equations of motion with allowance for the electric field  $E_y, E_z \propto \exp(-i\omega t)$ . If conditions (10) are met, the equations of motion are the Landau–Lifshitz (LL) equations, which for the two-sublattice cases take the form [18]

$$\dot{\mathbf{M}} = \gamma \left( \mathbf{M} \times \frac{\partial \Phi_2}{\partial \mathbf{M}} + \mathbf{L} \times \frac{\partial \Phi_2}{\partial \mathbf{L}} \right), \tag{13}$$

$$\dot{\mathbf{L}} = \gamma \left( \mathbf{M} \times \frac{\partial \Phi_2}{\partial \mathbf{L}} + \mathbf{L} \times \frac{\partial \Phi_2}{\partial \mathbf{M}} \right), \tag{14}$$

where the dot over indicates a time (t) derivative, and  $\gamma$  is the absolute value of the gyromagnetic ratio. In the linear approximation, the first equation proves to be an equation for  $\Delta M_y$  and  $\Delta M_z$ , and it can be solved if we take into account expression (11) for  $\Phi_{2M}$ , while the second equation is an equation for  $\Delta L_y$  and  $\Delta L_z$  in which we took into account expression (12) for  $\Phi_{2L}$ .

2.2.1 The magnetoactive ferromagnon mode  $\Gamma_{23}(\Delta M_y, \Delta M_z)$  in the  $\Gamma_1(M_x^0)$  phase. Ferromagnons. The solutions of equation (13) differ in no way from the solutions for an ordinary (single-sublattice) rhombic ferromagnet (e.g., see Ref. [3]) and are ferromagnetic spin waves with a natural frequency (their gap)

$$\omega_M = \gamma \sqrt{(2M_0 K_{M1} + H_x)(2M_0 K_{M2} + H_x)} . \tag{15}$$

Another name for these waves is homogeneous ferromagnons. Since expression (11) for  $\Phi_{2M}$  does not contain ME interactions, ferromagnons do not interact with the electric field  $\mathbf{E}(t)$ . They are excited by an alternating magnetic field  $\mathbf{H}(t) \perp X$ , whose frequency is close to  $\omega_M$ .

2.2.2 The electroactive antimagnon mode  $\Gamma_{56}(\Delta L_y, \Delta L_z)$  in the ferromagnetic  $\Gamma_1(M_x^0)$  phase. The fact that our magnetic substance constitutes a two-sublattice one suggests that there is one more branch of spin waves that represent oscillations of the vector  $\mathbf{L}$  (2), with the total local FM vector  $\mathbf{M}$  (1) remaining constant. Here, equation (14), together with expression (12), yields

$$\Delta L_i = \beta_{ij} E_j \quad (i, j = y, z) \,, \tag{16}$$

where  $\beta_{ij}$  is the electric antimagnon susceptibility with the following nonzero components

$$\beta_{yy} = s_1 \frac{2M_0}{2M_0 A_2 + H_x} \beta(\omega) , \quad \beta_{zz} = s_2 \frac{2M_0}{2M_0 A_1 + H_x} \beta(\omega) ,$$
  
$$\beta_{zy} = is_1 \frac{2\gamma M_0 \omega}{\omega_z^2} \beta(\omega) , \quad \beta_{yz} = -\frac{s_2}{s_1} \beta_{zy} , \qquad (17)$$

$$\beta(\omega) = \frac{\omega_L^2}{\omega_7^2 - \omega^2} \,.$$

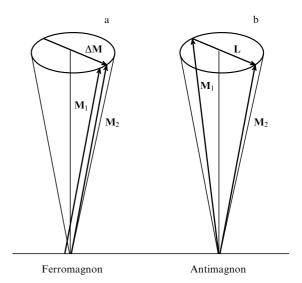
The quantity

$$\omega_L^2 = \gamma^2 (2M_0 A_1 + H_x)(2M_0 A_2 + H_x) \tag{18}$$

determines the frequency of antimagnon resonance (the homogeneous case) which is an addition to ferromagnon resonance corresponding to frequency (15) (see Fig. 2). As Fig. 2b and formulas (16) and (17) clearly show, for the vector  $\mathbf{L} = \Delta \mathbf{L}$  there is elliptic (for the rhombic case) precession about the axis  $X \parallel \mathbf{M}^0$ , similar to the precession for the vector  $\Delta \mathbf{M} = \mathbf{M} - \mathbf{M}^0$  (Fig. 2a).

## 2.3 Spatial dispersion. Heat losses due to antimagnon excitation by field $\mathbf{E}(t)$

Spatial dispersion (the dependence of  $\omega_M$  and  $\omega_L$  on the wave vector **k**) can also be taken into account by introducing inhomogeneous terms into the initial potential  $\Phi$  (9), terms with spatial derivatives of **M** and **L**. Note that the following



**Figure 2.** Two types of precession of vectors  $M_1$  and  $M_2$ : (a) the ferromagnon type  $\Delta M = M - M^0$ , and (b) the antimagnon type  $\Delta L \equiv L$ .

result has been obtained for small k's [8]:

$$\omega_M(k) \approx \omega_M(0) + D_M k^2,$$

$$\omega_L(k) \approx \omega_L(0) + D_L k^2,$$
(19)

where  $\omega_M(0) = \omega_M$  is the FM resonance frequency (15),  $\omega_L(0) = \omega_L$  is the antimagnon resonance frequency (18), while  $D_M$  and  $D_L$  are constants. In both cases, the dispersion law is quadratic and, generally,  $D_M$  and  $D_L$  are matrices of rhombic symmetry.

As for the minimum frequency (the gap in the spectrum), here the situation is quite different:  $\omega_M$  usually resides in the microwave frequency range, while  $\omega_L$  [the exchange interaction contributes the most to this frequency; see formula (18)] usually lands in the optical (IR or, possibly, the submillimeter) range. Moreover, one must not forget that ferromagnons are excited by a magnetic field, while antimagnons are excited by an electric field.

In analyzing resonance phenomena in the field  $\mathbf{E} = \mathbf{E}(t)$ , one must bear in mind heat losses Q due to antimagnon excitation. To evaluate these losses, it is convenient to first represent the ME interaction [the last two terms in  $\Phi$  (9)] as  $-P_y E_y - P_z E_z$ , where  $P_y = s_1 L_y$  and  $P_z = s_2 L_z$  are the components of the vector  $\mathbf{P}$  of effective polarization related to  $\mathbf{I}$ .

Absorption can then be written in the well-known form [19]

$$Q = -\overline{\mathbf{P}} \frac{\mathrm{d}\mathbf{E}}{\mathrm{d}t},\tag{20}$$

where the line above the right-hand side indicates averaging over the time  $t \gg 2\pi/\omega$ .

Let  $E_z = 0$ . The cofactors in the quadratic expression (20) must be taken in the real-valued form, for example:

$$\operatorname{Re} P_{y}(t) = \frac{1}{2} s_{1} \left[ \beta_{yy} E_{y} \exp \left(-i\omega t\right) + \beta_{yy}^{*} E_{y}^{*} \exp \left(i\omega t\right) \right].$$

Reasoning in a similar manner, we arrive at  $Re E_y$ , with the result that

$$Q_{E_y} = \frac{1}{4} s_1 i\omega(\beta_{yy}^* - \beta_{yy}) |E_y|^2 = \frac{1}{2} s_1 \omega \beta_{yy}'' |E_y|^2.$$

<sup>&</sup>lt;sup>2</sup> What we mean here is that the entire vector  $\mathbf{M}$ , and not only its length  $|\mathbf{M}|$ , is constant.

Here, to obtain the imaginary part of the susceptibility  $\beta$  we must take dissipation into account in Eqn (17). In the simplest case, this is achieved through the substitution  $\omega \to \omega + i\Gamma$  ( $\Gamma$  is the half-width of the resonance curve).

Similar results are also obtained for  $E_z \neq 0$  and  $E_y = 0$ . But if  $E_z \neq 0$  and  $E_y \neq 0$ , the contributions of the components  $E_y$  and  $E_z$  mix in a nonadditive way.

#### 2.4 Transfer to tetragonal symmetry

The above results can be extended to some tetragonal magnets — one must only make sure that a four-fold axis, either  $\mathbf{4}(+)$  or  $\mathbf{4}(-)$ , which complements the rhombic group P*mmm*  $(D_{2h}^1)$  to the respective tetragonal group, is directed along the symmetry axis  $\mathbf{2} \equiv \mathbf{2}(+)$  in such a way that the following condition is satisfied:

$$\mathbf{4}_{i}^{2}(\pm) = \mathbf{2}_{i}(+). \tag{21}$$

In tetragonal crystals, the symmetry axis  $\bf 4$  is usually assumed to be the coordinate Z-axis, so that in code (6) it is advisable to perform a cyclic permutation of the coordinates:

$$x \to z \to v \to x$$
. (22)

Then the code assumes the form  $\overline{\mathbf{1}}(-)\mathbf{2}_x(-)\mathbf{2}_z(+)$ , and adding to it  $\mathbf{4}_z(+)$  or  $\mathbf{4}_z(-)$  according to Eqn (21), we arrive at two tetragonal groups with the following position codes:

I. 
$$\overline{\mathbf{1}}(-)\mathbf{2}_x(-)\mathbf{4}_z(+)$$
, II.  $\overline{\mathbf{1}}(-)\mathbf{2}_x(-)\mathbf{4}_z(-)$ . (23)

Clearly, the bilinear thermodynamic potential  $\Phi$  can be used in the previous form (9), but two additional conditions must be added: first, the cyclic permutation (22) must be taken into account and, second, the coefficients must be corrected so that they incorporate the fact that  $\Phi$  is invariant with respect to the symmetry axes  $\mathbf{4}_z(+)$  and  $\mathbf{4}_z(-)$ . This leads to  $A_1 = A_2 = A_L - A_M + K_L \equiv J$ , since the anisotropy constants in the tetragonal case are equal:  $K_{L1} = K_{L2} \equiv K_L$ .

Here, the antimagnon resonance frequency (18) (in the geometry similar to that used in Section 2.2.2) assumes the form  $\omega_L = \gamma (2M_0J + H_z)$ .

As for the ME constants  $s_1$  and  $s_2$ , in contrast to the situation examined in the previous sections (the group  $D_{2h}^1$ ), they prove to be coupled, and this coupling is different for the cases I and II in Eqn (23):

$$s_1 = s_2 = s$$
 for I (4(+)),  
 $s_1 = -s_2 = s$  for II (4(-)). (24)

Bearing in mind the aforesaid, we can easily write down the remaining appropriate formulas from Section 2.2, as applied to the tetragonal case. But since the system is now cylindrically symmetric (in the bilinear approximation), it is convenient to employ the circularly polarized variables  $L_{\pm} = L_x \pm i L_y$  and  $E_{\pm} = E_x \pm i E_y$ .

For both cases in Eqn (23), the results in terms of these variables have the form

I. 
$$L_{\pm} = \beta_{\pm} E_{\pm}$$
, II.  $L_{\pm} = \beta_{\pm} E_{\mp}$ , (25)

where the upper (lower) indices  $(\pm)$  on the right-hand sides correspond to the upper (lower) indices on the left-hand sides, and

$$\beta_{\pm} = \frac{2s\gamma M_0}{\omega_L \pm \omega} \ . \tag{26}$$

Thus, the difference between the cases of positions  $\mathbf{4}_z(+)$  and  $\mathbf{4}_z(-)$  is that antimagnon resonance for the  $L_-$  oscillations is excited by a circularly polarized field  $E_-$  or  $E_+$  for  $\mathbf{4}_z(+)$  and  $\mathbf{4}_z(-)$ , respectively.

#### 2.5 Coupled antimagnon-electromagnetic waves

If we consider the field **E** in the ME interaction to be the electric field in an electromagnetic wave, it is the cause for coupling between antimagnons and the electromagnetic waves (light), thus leading to changes in the optical characteristics of matter. As a first approximation, we can ignore the spatial dispersion of antimagnons and assume in formula (19) that

$$\omega_L(k) \approx \omega_L(0) = \omega_L$$
 (27)

Next, we take into account relationships (22) and (24) and represent the ME interaction [the last two terms on the right-hand side of equation (9)] in the form

$$\Phi_{\rm ME} = -s(P_+E_- + P_-E_+)$$
,

where  $P_+$  and  $P_-$  are the effective polarizations caused by the electric field  $E_\pm$  of the electromagnetic wave through the  $L_+$  oscillations:

$$P_{+} = sL_{+}, \quad P_{-} = sL_{-} \text{ for I},$$
  
 $P_{+} = sL_{-}, \quad P_{-} = sL_{+} \text{ for II}.$  (28)

The field  $E_{\pm}$  for waves with the wave vector  $\mathbf{k} \parallel Z$  satisfies the wave equation [3, 8]

$$n^2 E_{\pm} = D_{\pm}(E_{\pm}) \,. \tag{29}$$

Here,  $n = kc/\omega$  is the refractive index, and  $D_{\pm}$  is the generalized induction [3]

$$D_{\pm} = \varepsilon_{\perp} E_{\pm} + 4\pi P_{\pm} \,, \tag{30}$$

where  $P_{\pm}$  has been defined in Eqn (28), and  $L_{\pm}$  in Eqns (25) and (26);  $\varepsilon_{\perp}$  is the transverse permittivity. If we ignore spatial dispersion of antimagnons, equations (25)–(30) yield

$$n_{+}^{2} = \varepsilon_{\perp} + 4\pi s \beta_{\pm} \quad \text{for} \quad \mathbf{4}_{z}(+) \,, \tag{31}$$

$$n_{+}^{2} = \varepsilon_{\perp} + 4\pi s \beta_{\pm} \quad \text{for} \quad \mathbf{4}_{z}(-) \,. \tag{32}$$

The difference  $n_+^2 - n_-^2$  determines Faraday rotation of the polarization plane (e.g., see Ref. [20, p. 96]) of electromagnetic waves (light), caused by the interaction of these waves and antimagnons. As a result, taking equations (31), (32), and (26) into account, we find that the Faraday angle per unit length travelled by the wave for the cases  $\mathbf{4}(+)$  and  $\mathbf{4}(-)$ 

$$\varphi_1 = \frac{1}{2}(k_+ - k_-) = \mp \frac{4\pi s^2 \gamma M_0 \omega^2}{cn(\omega_T^2 - \omega^2)}.$$
 (33)

The results for 4(+) and 4(-) differ only in sign [a 'plus' and 'minus', respectively, in formula (33)]. The effect has a resonance at the frequency

$$\omega = \omega_L = \gamma (2M_0J + H_z) \,,$$

which can be attained by varying the external magnetic field  $H_z$ .

Allowing for antimagnon dissipation [this is done by replacing  $\omega$  with  $\omega + i\Gamma$  in formula (33)] results in the function  $\varphi_1$  becoming complex-valued. This means that an electromagnetic wave that is linearly polarized at the input at frequencies  $\omega = \omega_L$  acquires, in addition to Faraday rotation, ellipticity related to what is known as dichroism [20]. Both the Faraday rotation and dichroism undergo a rapid change near the resonance frequency. Here, of course, one must bear in mind that this singularity is proportional to the square of the small parameter s.

It is also worth noting that magneto-optical effects caused by ME interaction in multisublattice antiferromagnets (nonreciprocal rotation of the plane of polarization of light, for one thing) have been thoroughly studied in experiments (e.g., see Refs [21, 22]), while what we examined was the FM phase. In Section 4.3.1, we will discuss the dynamics of the four-sublattice  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> and Cr<sub>2</sub>O<sub>3</sub> antiferromagnets with the ME interaction taken into account.

# 3. A two-sublattice antiferromagnet with magnetoelectric interaction. Quasiantiferromagnons and their excitation by an electric field

The existence of the above phenomena is related to a not infrequent feature of the position of multiple points occupied by magnetic atoms. The symmetry center  $\overline{\bf 1}$  must not be a closed element for this position or, in other words, it must not permute the atoms in an identical manner, i.e.,  $\overline{\bf 1} \equiv \overline{\bf 1}(-)$  in the notation adopted here. We have discussed the properties of the simplest FM structure for this case: the presence of a new type of spin wave, or antimagnons which correspond only to the oscillations of the AFM L vector and are excited not by a magnetic field (as ordinary magnons are) but only by an electric field  ${\bf E}(t)$  at frequencies of an exchange origin.

But what will happen if the magnetic moments in a twofold position are ordered not in a ferromagnetic way (as happened above) but in an antiferromagnetic way? Suppose that we are examining a typical antiferromagnet, the phase  $\Gamma_6(L_z^0)$  from Table 1, in which  $\mathbf{L}^0 \| Z (L_z^0 = 2M_0)$  and  $\mathbf{M}^0 = 0$  (of course, other AFM phases, such as  $\Gamma_4(L_x^0)$  or  $\Gamma_5(L_v^0)$ , can also be analyzed, but the result will be the same). The general form of the thermodynamic potential remains almost the same [see Eqn (9)]. Only the ME interaction and the relationship between the exchange constants will change  $(A_M > 0 \text{ and } A_L < 0)$ . Table 1 suggests that in AFM phases, including  $\Gamma_6$ , there are no antimagnons, since otherwise rule (8) will not be satisfied. It might be well to point out that here this is essentially related to the fact that the magnetic substance is of the twosublattice type. The reader will recall that in Refs [10–12, 14] only four-sublattice antiferromagnets were examined, and for these the above statement is invalid.

At the same time, rule (8) applied to the ground AFM state  $\Gamma_6(L_z^0)$  from Table 1 yields

$$\Gamma_2(M_v) \times \Gamma_4(L_x) = \Gamma_1(M_x) \times \Gamma_5(L_v) = \Gamma_6(L_z^0)$$

i.e., there are two quasiantiferro-modes,  $\Gamma_{24}(M_y, L_x)$  and  $\Gamma_{15}(M_x, L_y)$ , each of which contains one component of the **M** vector. As noted earlier, antimagnons, i.e., oscillations in which **M** does not take part, are not present in two-sublattice AFM phases. The magnetoelectric terms in the potential  $\Phi$ 

become

$$- s_1 \frac{L_z^0}{2M_0} M_x E_z - s_2 \frac{L_z^0}{2M_0} M_z E_x$$

$$\equiv -s_1 M_x E_z - s_2 M_z E_x, \qquad (34)$$

where the factors  $L_z^0/(2M_0) = 1$  were introduced only to verify the invariance of  $\Phi$ . From Eqn (34) we see that, say, for  $\Gamma_{15}$  the ME interaction, i.e., the term with  $s_1$ , incorporates only one oscillation variable  $(M_x)$  of one of the  $\Gamma_{15}(M_x, L_y)$  modes, which thus can only be excited by an electric field  $\mathbf{E} \parallel Z$ .

Combining what we have stated above, we can easily find the linear response of  $M_x$  to  $E_z \propto \exp(-i\omega t)$  from Eqn (13):

$$M_{x} = \frac{s_{1}(2\gamma M_{0})^{2} K_{L2}}{\omega_{EK}^{2} - \omega^{2}} E_{z},$$

where

$$\omega_{EK}^2 = 2\gamma^2 H_E H_K \tag{35}$$

determines the exchange-relativistic frequency of the quasiantiferro-mode  $\Gamma_{15}$ , which incorporates the exchange field

$$H_E = 2M_0(A_M - A_L + K_{M1})$$

[with the anisotropic correction; see Eqn (9)] and the magnetic anisotropy field  $H_K = 4M_0K_{L2}$ . Both modes  $\Gamma_{15}$  and  $\Gamma_{24}$  can be excited by an alternating magnetic field  $[\mathbf{H}(t) \parallel Z]$  and  $\mathbf{H}(t) \parallel Y$ , respectively], but we will focus only on the first mode, since it also can be excited by an electric field  $\mathbf{E} \parallel Z$ , and this excitation occurs at very low frequencies (in the microwave range), much lower than the frequencies at which antimagnons are excited.

Thus, even quasiantiferromagnons may be electroactive, provided that the appropriate thermodynamic potential contains the ME interaction covering the variables of this mode.

## 4. Tetragonal and other four-sublattice antiferromagnets, magnetoelectric and antiferroelectric phenomena

## **4.1 Introductory remarks, thermodynamic potential, and the equations of motion. Tetragonal antiferromagnets** In Sections 2 and 3, we used the simplest two-sublattice model

(FM and AFM) to examine a number of new dynamical phenomena related to the ME and AFE interactions. Unfortunately, we were unable to indicate the specific magnetic substances in which experimenters should look for these predicted phenomena. But such magnets do exist (trirutiles, rare-earth phosphates, vanadates, etc. [10, 14, 23–27]), although they belong to more complicated v-sublattice structures ( $v \ge 4$ ). So far no experiments revealing the presence of such phenomena have been conducted. Nevertheless, we would like to point out the specific substances and the features of the effects predicted in them in order to indicate the area in which the experimenters should look for these effects.

First, we should mention tetragonal four-sublattice antiferromagnets with the trirutile structure: space group  $P4_2/mnm$   $(D_{4h}^{14})$ , magnetic-atoms position 4e (Fe, Cr, V, etc.)

in which the symmetry center  $\overline{\bf 1}$  permutes each atom in a nonidentical manner (into each other and not into itself) [3, 25, 27]. In addition to this property of the position of magnetic atoms, notice that among trirutiles there are easy-axis structures and easy-plane antiferromagnets. Among the first is Fe<sub>2</sub>TeO<sub>6</sub> ( $T_{\rm N}=217~{\rm K}$ ) which has proved to be the simplest substance (see Ref. [26]) to demonstrate some of the properties of trirutiles we are interested in. Other trirutiles, including the easy-plane trirutiles Cr<sub>2</sub>TeO<sub>6</sub>, Cr<sub>2</sub>WO<sub>6</sub>, and V<sub>2</sub>WO<sub>6</sub>, have been examined in Refs [25, 27].

In the single-position antiferromagnet in question, the four sublattice magnetizations  $\mathbf{M}_{\nu}$  ( $\nu = 1, 2, 3, 4$ ) can determine, in addition to the vector of total local magnetization  $\mathbf{M}$ , three more AFM-basis vectors  $\mathbf{L}_a$ ,  $\mathbf{L}_b$ , and  $\mathbf{L}_c$ :

$$\mathbf{M} = \mathbf{M}_{1} + \mathbf{M}_{2} + \mathbf{M}_{3} + \mathbf{M}_{4},$$

$$\mathbf{L}_{a} = \mathbf{M}_{1} + \mathbf{M}_{2} - \mathbf{M}_{3} - \mathbf{M}_{4},$$

$$\mathbf{L}_{b} = \mathbf{M}_{1} - \mathbf{M}_{2} + \mathbf{M}_{3} - \mathbf{M}_{4},$$

$$\mathbf{L}_{c} = \mathbf{M}_{1} - \mathbf{M}_{2} - \mathbf{M}_{3} + \mathbf{M}_{4}.$$
(36)

For the generators of the group  $D_{4h}^{14}$ , it has proved convenient to take the elements  $\overline{\bf 1}$ ,  ${\bf 2}_{1x}$ , and  ${\bf 4}_{2z}$ , and combine them to arrive at the position code by specifying their permutation properties in the following form

$$\overline{\mathbf{1}} \begin{pmatrix} 1-2\\3-4 \end{pmatrix} \mathbf{2}_{1x} \begin{pmatrix} 1-4\\2-3 \end{pmatrix} \mathbf{4}_{2z} \begin{pmatrix} 1-3\\2-4 \end{pmatrix}. \tag{37}$$

Here, dashes connect the atom numbers that are permuted by the respective symmetry element. Position code (37) makes it possible to build a table of transformations of all the components of basis vectors (36), similar to Table 1. Such a table takes into account not only the point transformations of the components of the vectors in formulas (36) (rotations and reflections) but also the permutation of atoms. We will not build such a table here, but it does differ from Table 1. The difference emerges because the table is based on that of the rhombic crystal for the group Pnmm which is a subgroup of  $P4_2/mnm$  if the former is complemented up to the latter with the element  $\mathbf{4}_{2z}$  (see Ref. [23]). We note only that, according to Eqns (36) and (37), two vectors in formulas (36),  $\mathbf{M}$  and  $\mathbf{L}_a$ , are centrally symmetric (CS), while the other two,  $L_b$  and  $L_c$ , are centrally antisymmetric (CAS) vectors. What this means is that the thermodynamic potential  $\Phi$  may contain in addition to an ME interaction  $s_{ijk}M_iL_{bj}E_k$  (or  $M_iL_{cj}E_k$ ) of type (3) also an AFE interaction  $f_{ijk}L_{ai}L_{bj}E_k$  (or  $L_{ai}L_{cj}E_k$ ) of type (4) [10-14] with coefficients that ensure the invariance of these expressions with respect to the elements (generators) of group (37).

The existence of an AFE interaction in a system of four (or more) basis vectors (in which at least three are antiferromagnetic vectors) is the reason for additional phenomena (compared to the characteristic phenomena involving two-sublattice magnets) related to the interaction of magnons with an alternating electric field  $\mathbf{E}(t)$ . This is true even for those magnetic substances where there can be no static ME effect, because such an effect would be forbidden according to symmetry considerations (a well-known example, which however refers to another crystal system, is the absence of a static ME effect in hematite). The new dynamical phenomena in magnetic substances with four sublattices (instead of two) also manifest themselves in NMR and in the region of coupled magnon-electromagnetic waves. The possibilities of

exciting magnons by the  $\mathbf{E}(t)$  field become more numerous: not only antimagnons can be excited, but in some cases even low-frequency (exchange-relativistic or even purely relativistic) magnons can be excited.

The phenomena proved to be so numerous that because of the space allocated to the present review we are able to mention only a small fraction of the new dynamical phenomena caused by the field  $\mathbf{E}(t)$ . What is important is that the prediction of these phenomena refers to specific substances (as it does in Refs [10-14]) that have been studied in other areas of the physics of magnetic phenomena, so that there is hope that in the future these phenomena will be discovered in experiments.

Thus, going to the theory, we write down the expression for the thermodynamic potential (per unit volume in the homogeneous state). We are talking about Fe<sub>2</sub>TeO<sub>6</sub>, but if the ME and AFE interactions are ignored, the following expression can also be used to describe other trirutiles and, in particular, easy-plane states [26]:

$$\Phi = \frac{1}{2} A_M \mathbf{M}^2 + \frac{1}{2} \sum_n A_n \mathbf{L}_n^2 + \frac{1}{2} K_M (M_x^2 + M_y^2)$$

$$+ \frac{1}{2} \sum_n K_n (L_{nx}^2 + L_{ny}^2) + r(L_{bx} L_{cy} + L_{by} L_{cx})$$

$$+ p(M_x L_{ay} + M_y L_{ax}), \quad n = a, b, c.$$
 (38)

For an easy-axis antiferromagnet (Fe<sub>2</sub>TeO<sub>6</sub>) with the structure  $\mathbf{L}_{b}^{0}$  ( $|\mathbf{L}_{b}^{0}| = 4M_{0}$ ), the terms

$$-\frac{1}{4M_0}L_{bz}^0[s(M_xE_x+M_yE_y)+f(L_{ax}E_y+L_{ay}E_x)]$$
 (39)

for the ME and AFE interactions, respectively, must be added to the right-hand side of equation (38) [the common factor  $L_{bz}^0/4M_0=1$  is introduced once again for the sole purpose of verifying the invariance of expression (39)]. For each of the other structures, we must write out the interactions inherent in it (interactions that incorporate the variables of the respective modes).

According to expression (38), the state with  $\mathbf{L}_b^0 \parallel Z$  is stable for  $A_b < 0$ ,  $A_M > 0$ ,  $A_a > 0$ ,  $A_c > 0$ ,  $K_M > 0$ , and  $K_n > 0$ . Here, we adopt the equal-modulus model  $\mathbf{M}_v^2 = M_0^2$ , which corresponds to the conditions

$$\mathbf{M}^{2} + \sum_{n} \mathbf{L}_{n}^{2} = (4M_{0})^{2},$$

$$\mathbf{M}\mathbf{L}_{a} + \mathbf{L}_{b}\mathbf{L}_{c} = 0,$$

$$\mathbf{M}\mathbf{L}_{b} + \mathbf{L}_{a}\mathbf{L}_{c} = 0,$$

$$\mathbf{M}\mathbf{L}_{c} + \mathbf{L}_{a}\mathbf{L}_{b} = 0.$$
(40)

and the LL equations of motion [18, 25, 27]:

$$\dot{\mathbf{M}} = \gamma \left( \mathbf{M} \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{M}} + \mathbf{L}_a \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_a} + \mathbf{L}_b \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_b} + \mathbf{L}_c \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_c} \right),$$

$$\dot{\mathbf{L}}_a = \gamma \left( \mathbf{M} \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_a} + \mathbf{L}_a \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{M}} + \mathbf{L}_b \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_c} + \mathbf{L}_c \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_b} \right),$$

$$\dot{\mathbf{L}}_b = \gamma \left( \mathbf{M} \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_b} + \mathbf{L}_a \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_c} + \mathbf{L}_b \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{M}} + \mathbf{L}_c \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_a} \right),$$

$$\dot{\mathbf{L}}_c = \gamma \left( \mathbf{M} \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_c} + \mathbf{L}_a \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_b} + \mathbf{L}_b \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{L}_a} + \mathbf{L}_c \times \frac{\partial \boldsymbol{\Phi}}{\partial \mathbf{M}} \right).$$

This system will be examined in Section 4.2, and conditions (40) actually follow from it.

## 4.2 Oscillation modes, solution of equations, and natural frequencies

Using the rules of transformation of the basis vectors (36), we immediately see that for the AFM phase with the CAS basis vector  $\mathbf{L}_b^0 \parallel Z$  (this vector corresponds to Fe<sub>2</sub>TeO<sub>6</sub>) there must be spin-wave representations with the oscillation variables

(a) 
$$M_x$$
,  $L_{ay}$ ,  $L_{by}$ ,  $L_{cx}$ , (42)

(b) 
$$M_{v}$$
,  $L_{ax}$ ,  $L_{bx}$ ,  $L_{cv}$ , (43)

which are related to each other via the symmetry axis  $\mathbf{4}_{2z}$ . Therefore, it is enough to solve the problem in question (to determine the spectrum, susceptibility, etc.) for one of these representations. We chose (a).

Let us isolate in expression (38) a quadratic form in the variables (42):

$$\Phi_{2} = \frac{1}{2} \widetilde{A}_{M} M_{x}^{2} + \frac{1}{2} \widetilde{A}_{a} L_{ay}^{2} + \frac{1}{2} \widetilde{A}_{c} L_{cx}^{2} + \frac{1}{2} K_{b} L_{by}^{2} 
+ p M_{x} L_{ay} + r L_{by} L_{cx} - s M_{x} E_{x}(t) - f L_{ay} E_{x}(t) - M_{x} H_{x}(t),$$
(44)

where we have also allowed for the respective parts of the ME and AFE interactions from formula (39) as well as the Zeeman energy in the alternating magnetic field  $H_x(t)$  and introduced the notation

$$\widetilde{A}_a = A_a - A_b + K_a$$
,  $\widetilde{A}_M = A_M - A_b + K_M$ ,

$$\widetilde{A}_c = A_c - A_b + K_c.$$

Of course, we would like to have taken into account a constant magnetic field  $\mathbf{H}_0 \parallel Z$ , but this usually leads to substantial complications in the solution of the problem. The thing is that the phase  $L_{bz}^0$  in question does not contain the vector  $\mathbf{M}^0$  generated by the field  $\mathbf{H}_0$ , i.e., this will be a different phase, which means that, generally speaking, in this case one should find the modes anew, instead of Eqns (42) and (43).

The relevant equations from set (41) combined with formula (44) form the following system

$$\begin{split} \dot{M}_{x} &= -\omega_{0} \frac{\partial \Phi_{2}}{\partial L_{by}} = -\omega_{0} (K_{b}L_{by} + rL_{cx}) \,, \\ \dot{L}_{by} &= \omega_{0} \frac{\partial \Phi_{2}}{\partial M_{x}} = \omega_{0} \big[ \widetilde{A}_{M}M_{x} + pL_{ay} - \big( sE_{x}(t) + H_{x}(t) \big) \big] \,, \\ \dot{L}_{cx} &= -\omega_{0} \frac{\partial \Phi_{2}}{\partial L_{ay}} = -\omega_{0} (\widetilde{A}_{a}L_{ay} + pM_{x} - fE_{x}) \,, \end{split} \tag{45}$$

$$\dot{L}_{ay} &= \omega_{0} \frac{\partial \Phi_{2}}{\partial L} = \omega_{0} (\widetilde{A}_{c}L_{cx} + rL_{by}) \,, \end{split}$$

where  $\omega_0 = \gamma 4M_0$ . Clearly, in accordance with the number of variables (four), the system of equations (45) must produce two oscillation modes.

In the exchange approximation (r = p = 0), system of equations (45) falls apart into two independent systems involving the first two and the last two equations, with the second system yielding an exchange mode with a frequency

$$\omega_E \equiv \omega_{ca} = \omega_0 \sqrt{\widetilde{A}_a \widetilde{A}_c} \tag{46}$$

and the following components of the oscillating AFM vectors:

$$L_{ay} = \frac{\omega_0^2}{\omega_F^2 - \omega^2} f \widetilde{A}_c E_x , \qquad L_{cx} = -\mathrm{i} \frac{\omega}{\widetilde{A}_c} L_{ay} .$$

The observable quantity in this mode is the *x*-component of the polarization vector:

$$P_{\scriptscriptstyle X} = -rac{\partial \Phi_{
m AFE}}{\partial E_{\scriptscriptstyle X}} = arkappa_{\perp} E_{\scriptscriptstyle X} \,,$$

where

$$\varkappa_{\perp} = \frac{\omega_0^2}{\omega_E^2 - \omega^2} f^2 \widetilde{A}_c. \tag{47}$$

However, here we are more interested in the other mode, the one generated by the field  $E_x$ , or the exchange-relativistic mode which corresponds to the system of the first two equations in set (45) with the variables  $M_x$  and  $L_{by}$ . In this mode, the variables  $L_{cx}$  and  $L_{ay}$  follow  $M_x$  and  $L_{by}$  in a quasiequilibrium manner. Minimizing  $\Phi$  (38) in  $L_{cx}$  and  $L_{ay}$ , we find that the thermodynamic potential depends only on  $M_x$  and  $L_{by}$ , and then the equations of motion for these variables (with allowance for the field terms) yield the following expression for the corresponding quasiantiferromode:

$$M_x = (\alpha_0 E_x + \chi_0 H_x) \frac{\omega_{AK}^2}{\omega_{AK}^2 - \omega^2}, \qquad (48)$$

where  $\alpha_0 = s/\widetilde{A}_M$  and  $\chi_0 = 1/\widetilde{A}_M$  are the respective static magnetoelectric and magnetic susceptibilities, and

$$\omega_{AK} = \omega_0 \sqrt{\widetilde{A}_M K_b} \tag{49}$$

[cf. expression (35)].

Formula (48) determines the quasiantiferro-resonances of the acoustic mode with frequency (49) both in an electric field  $E_x(t)$  and in a magnetic field  $H_x(t)$ : the first is to be detected in the antinode of  $\mathbf{E}(t)$ , and the second in the antinode of  $\mathbf{H}(t)$ . In this case,  $|E_x| = |H_x|$ , and the ratio of the absolute values of the respective magnetizations is determined by the ratio of the static magnetoelectric and magnetic susceptibilities:

$$\frac{M_{\alpha}^{E}}{M_{x}^{h}} = \frac{\alpha_{0}}{\chi_{0}} \,. \tag{50}$$

As for the exchange mode, which emerges because of the last two equations in set (45), the terms with p and r have very little effect on its frequency, since at this frequency ( $\omega_E \geqslant \omega_{AK}$ ) the variables  $M_x$  and  $L_{by}$  remain practically at their equilibrium values, i.e., close to zero. Actually, of course, the relativistic constants  $p \neq 0$  and  $r \neq 0$  ensure the weak coupling of the exchange and quasiantiferromagnetic

<sup>&</sup>lt;sup>3</sup> This is not true for the equal-modulus model, where  $\chi_{\parallel}=0$ , and for this reason  $M_0=0$  even after the field  $\mathbf{H}_0 \parallel \mathbf{L}_b^0 \parallel Z$  has been applied. As a result, the problem with the field  $H=H_z$  can easily be solved. Here, in the term  $-H_zM_z$  one must take into account the third equal-modulus condition in Eqn (40). Note that Krivoruchko and Yablonskii [11] took into consideration the role that a nonzero field  $\mathbf{H}_0$  plays in the respective problem for hematite.

modes [see Eqns (45) and (46)], somewhat renormalizing their frequencies and generating the respective poles, e.g., the exchange pole on the quasiantiferro-mode, with a small intensity, however.

Notice that similar results have been obtained for other tetragonal four-sublattice antiferromagnets, in particular, for rare-earth phosphates of the TbPO<sub>4</sub> and HoPO<sub>4</sub> type, and vanadates of the GdVO<sub>4</sub> and TbVO<sub>4</sub> type, which have the  $D_{4h}^{19}$  group (the zirconium structure) [3].

Since the characteristics of various magnetic substances vary within a broad range ( $\alpha_0 \approx 10^{-2}-10^{-5}$  and  $\chi_0 \approx 10^{-3}-10^{-5}$  in the centimeter–gram–second system of units), a situation in which ME resonance will be comparable in intensity, according to relationship (50), with AFM resonance is quite possible [8].

The respective results for the spin-wave representation in formula (43) can be obtained from Eqns (44)–(50) via the substitutions  $x \leftrightarrow y$  and  $\omega \rightarrow -\omega$ .

Unfortunately, we were unable to find in the scientific literature any data concerning a specific antiferromagnet for which both susceptibilities (with close enough values) are known from experiments. The problem is quite intriguing, especially if we note that here we are speaking of an AFM resonance in an electric field at a fairly low frequency. From the viewpoint of the search for such antiferromagnets, most promising among tetragonal antiferromagnets are, in addition to trirutiles (not necessarily Fe<sub>2</sub>TeO<sub>6</sub>), the aforementioned phosphates and vanadates in which the magnetic rareearth ions occupy the four-fold position  $4a\{2_x\}$  (the expression inside the braces indicates the local symmetry of the position). As for the other crystal systems, we would like to point out  $Cr_2O_3$  and  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> (rhombohedral crystals) which have been thoroughly studied theoretically in Refs [10-14]. Unfortunately, experimental data on the magnetic susceptibilities and corresponding ME coupling constants (especially, the AFE constants) are not usually given together. Despite the fact that the effects associated with AFE interaction and discussed in the present review were discovered rather long ago (in 1988), so far no relevant experiments have been conducted in this area of research.

Many theoretical predictions of such effects are known today [8, 10-14, 25]. We discussed the main physical phenomena in Section 1 using a simple two-sublattice model, so we would like to finalize this section by only indicating the phenomena related to ME and AFE interactions solely in multisublattice antiferromagnets (v = 4). Due to its popularity and simplicity, of greatest interest here from the viewpoint of principal questions is the case of hematite  $(\alpha-\text{Fe}_2\text{O}_3)$  [11, 14]. The thing is that in the ground state hematite possesses a structure described by such a centrally symmetric basis vector as  $L_a$  in formulas (36), so that  $1 \mathbf{L}_a = \mathbf{L}_a$ . However, when we allow for all four basis vectors, which is required by dynamics, the two remaining vectors  $\mathbf{L}_{b,c}$  are CAS vectors (although the numbering of atoms may be different [8] from that in trirutiles, this is not really important in the sense that this does not introduce new phenomena — the numbering of atoms can be changed [8]). Fortunately, Krivoruchko and Yablonskii in their paper [11] provided almost everything that we want to know about hematite, and there is no need to go into further details about that paper. However, in Section 4.3 we present some results for rhombohedral antiferromagnets, so that we can compare them to the results for tetragonal antiferromagnets (the easyaxis case).

## 4.3 Rhombohedral antiferromagnets: hematite and chromite

Hematite (α-Fe<sub>2</sub>O<sub>3</sub>) and chromite (Cr<sub>2</sub>O<sub>3</sub>) are the most thoroughly studied antiferromagnets belonging to the rhombohedral crystal system: the space group is  $R\overline{3}c \equiv D_{3d}^6$ , and the magnetic Fe<sup>3+</sup> and Cr<sup>3+</sup> ions occupy the four-fold position  $4c\{3_z\}$ . If we number these ions in a certain manner (the same for both cases), instead of the vectors  $\mathbf{M}_{v}$ (v = 1, 2, 3, 4) we can introduce their linear combinations (36) which coincide closely with those for trirutiles. What is important is that here, in contrast to the commonly adopted system of atomic numbering [4], the atomic numbers 2 and 4 are permuted here. As a result, the centrally symmetric vector  $L_a$  corresponds to the ground state of hematite, while the CAS vector  $\mathbf{L}_c$  corresponds to the ground state of chromite. Therefore, in hematite there is static weak ferromagnetism (WFM) and no static ME effect. On the other hand, chromite exhibits an ME effect but no WFM. However, in the dynamical case, where there may be oscillations of vectors [see formula (42)] that do not belong to the ground state, these oscillations can be excited by an electric field due to either an ME interaction of type (3) or an AFE interaction (4). If the ground state is centrally symmetric (being determined by vector  $L_a$ , as it is in hematite), the electric field  $\mathbf{E}(t) \propto \exp(-\mathrm{i}\omega t)$  in the linear approximation excites oscillations of a CAS vector  $\mathbf{L}_b$  or  $\mathbf{L}_c$ .

Here, the AFE interaction (4) plays an active dynamical role. On the contrary, in a CAS state (e.g., defined by the vector  $\mathbf{L}_c$ , as in chromite) the field  $\mathbf{E}(t)$  excites oscillations of the centrally symmetric vector  $\mathbf{M}$  or  $\mathbf{L}_a$  thanks to the ME or AFE interaction, respectively. Moreover, the vector  $\mathbf{M}$  may, of course, also be excited by the alternating magnetic field  $\mathbf{H}(t) \propto \exp{(-\mathrm{i}\omega t)}$  as a result of the Zeeman interaction  $\mathbf{MH}(t)$ .

Next, one should write down the thermodynamic potential  $\Phi$  in terms of the equal-modulus phenomenology adopted here ( $\mathbf{M}_{v}^{2} = M_{0}^{2} = \text{const}$ ) or the Hamiltonian, as in the Krivoruchko-Yablonskii approach [11] in which the dynamical properties of the system are described on the basis of quantum equations of motion. If we ignore purely quantum phenomena, such as, say, covering zero-oscillation energy, both approaches for the spectrum and the amplitude of oscillations yield the same results corresponding to the LL equations (41) (e.g., see Refs [28-30]). A number of misprints have been discovered in Ref. [11], and so to verify the results we used an approach described in detail in Ref. [3] and employed in Sections 4.1 and 4.2. Here, of course, we base our reasoning on the requirement that  $\Phi$  is invariant under the transformations of the space group  $R\overline{3}c \equiv D_{3d}^6$ , and express  $\Phi$  and the ME and AFE interactions (3) and (4) in the form of a series in dynamic variables (36).

Krivoruchko and Yablonskii [11] showed that, as in the case of antiferromagnetic trirutiles, the dynamical properties of rhombohedral antiferromagnets strongly depend on whether the fundamental vector in formulas (36), which determines the ground state, is centrally symmetric or centrally antisymmetric. As noted earlier, in hematite and chromite these vectors are  $\mathbf{L}_a$  and  $\mathbf{L}_c$ , respectively. It is advisable to compare these properties with those of trirutiles (we again have in mind easy-axis states).

**4.3.1 Hematite.** In hematite below the Morin point in the  $\mathbf{L}_a^0 \parallel Z$  state, there are two independent spin-wave representa-

tions

$$\Gamma_1(M_x, L_{ay}, L_{bx}, L_{cy}), \qquad (51)$$

$$\Gamma_2(M_v, L_{ax}, L_{bv}, L_{cx}). \tag{52}$$

These representations correspond to the following thermodynamic potential density (the expression allows for the ME and AFE interactions):

$$\Phi = \frac{1}{2} A_1 \mathbf{L}_a^2 + \frac{1}{2} A_2 \mathbf{L}_b^2 + \frac{1}{2} A_3 \mathbf{L}_c^2 + \frac{1}{2} A_0 \mathbf{M}^2 
- \frac{1}{2} K_1 L_{az}^2 + \frac{1}{2} K_2 L_{bz}^2 + \frac{1}{2} K_3 L_{cz}^2 + \frac{1}{2} K_0 M_z^2 
+ d(M_y L_{ax} - M_x L_{ay}) + d'(L_{bx} L_{cy} - L_{by} L_{cx}) 
+ E_x (R_2 L_{bx} + R_3 L_{cy}) + E_y (R_2 L_{by} - R_3 L_{cx}) - \mathbf{MH}(t),$$
(52)

where  $A_1 < 0$  and  $A_{2,3,0} > 0$ , while  $\mathbf{E}(t) \perp Z$  and  $\mathbf{H}(t) \perp Z$  are the fields that excite transverse oscillations. In contrast to Ref. [11], here we introduced constants similar to those used in Sections 4.1 and 4.2 and related to the constants from Ref. [11] by the following formulas:  $H_{\rm ci} = 4M_0A_i$  and  $H_{\rm c0} = 4M_0A_0$  for exchange,  $H_{\rm Ai} = 4M_0K_i$  and  $H_{\rm A0} = 4M_0K_0$  for magnetic (relativistic) anisotropy (i=1,2,3), and, finally,  $H_{\rm D} = 4M_0d$  and  $H_{\rm D'} = 4M_0d'$  for the exchange-relativistic Dzyaloshinskii constants. Clearly, the thermodynamic potential density  $\Phi$  (53) falls apart into two independent parts corresponding to spin-wave representations (51) and (52). Similarly, there are two independent systems of LL equations (or, as in Ref. [11], two systems of quantum-mechanical equations of motion).

Below, we present the approximate results for the natural frequencies and the susceptibilities (the case of homogeneous oscillations). Since the centrally symmetric vectors ( $\mathbf{L}_a$  and  $\mathbf{M}$ ) and the CAS vectors ( $\mathbf{L}_b$  and  $\mathbf{L}_c$ ) participate, respectively, in the acoustic oscillation modes and the exchange oscillation modes, the form of the field terms in  $\Phi$  (53) suggests that the exchange modes and the acoustic modes can be excited by alternating electric and magnetic fields, respectively.

For the exchange modes, we first find  $L_{cy}$ ,  $L_{bx}$  and  $L_{cx}$ ,  $L_{by}$  as well as the exchange frequencies  $\omega_{01}$  and  $\omega_{02}$ . Then we evaluate the corresponding components of the polarization vector  $\mathbf{P} = -\partial \Phi/\partial \mathbf{E} = \hat{\mathbf{z}}_{\perp} \mathbf{E}$ :

$$P_{x} = -(R_{2}L_{bx} + R_{3}L_{cy}), \quad P_{y} = R_{2}L_{by} + R_{3}L_{cx};$$
  
 $P_{x} = \varkappa_{\perp}E_{x}, \quad P_{y} = \varkappa_{\perp}E_{y},$  (54)

where  $\varkappa_{\perp}$  is the electric susceptibility equal to

$$\varkappa_{\perp} = \omega_0^2 \, \frac{R_3^2 (A_2 - A_1) + R_2^2 (A_3 - A_1)}{\omega_F^2 - \omega^2} \,, \tag{55}$$

and  $\omega_E$  is a frequency given by the formulas

$$\omega_E \equiv \omega_{E1} = \omega_{E2}$$

$$= \omega_0 \left[ (A_3 - A_1 + K_1)(A_2 - A_1 + K_1) - d'^2 \right]^{1/2}, \quad (56)$$

$$\omega_0 = \gamma 4 M_0.$$

Notice that here  $R_2$  and  $R_3$  are antiferromagnetoelectric exchange-relativistic constants. Moreover, from expression (55) it follows that the absorption intensity related to  $\mathbf{P}$  is exchange-enhanced by the presence of the appropriate exchange factors.

Reasoning in a similar manner, we can find the two acoustic modes with the variables  $M_x$ ,  $L_{ay}$  and  $M_y$ ,  $L_{ax}$ , which are frequency degenerate:

$$\omega_{A1} = \omega_{A2} \equiv \omega_{AK} = \omega_0 \left\{ K_1 [-A_1 + A_M + K_1] \right\}^{1/2}.$$
 (57)

The degeneracy is removed by applying a constant magnetic field  $\mathbf{H} \parallel Z$  [11]. The amplitudes equal  $M_x = \chi_{\perp}(\omega)H_x$  and  $M_y = \chi_{\perp}(\omega)H_y$ , where  $\chi_{\perp}(\omega)$  is the magnetic susceptibility defined as

$$\chi_{\perp}(\omega) = \frac{\omega_0^2 K_1}{\omega_{AK}^2 - \omega^2} \,. \tag{58}$$

Unfortunately, the structure of trirutile with  $\mathbf{L}_a^0 \parallel Z$ , with which it would be advisable to compare the above results for hematite, so far has not been realized in experiments, which means one is left with theoretical formulas. This case for trirutile was not considered above, but the result can be taken from Ref. [27]. The expression for exchange modes, given in that paper, coincides with Eqn (54), but instead of formula (55) we have

$$\varkappa_{\perp} = \omega_0^2 \, \frac{f_1^2 \widetilde{A}_c + f_2^2 \widetilde{A}_b}{\omega_F^2 - \omega^2} \,,$$

where  $\widetilde{A}_b \approx A_b - A_a$  and  $\widetilde{A}_c \approx A_c - A_a$ , while  $f_1$  and  $f_2$  are AFE coupling constants [from an expression of type (4)]. What is important is that these constants are purely relativistic, in contrast to the exchange-relativistic parameters  $R_2$  and  $R_3$  which are present in formula (55) for hematite. Actually, the difference amounts to this.

But if we turn to acoustic modes, in the adopted approximation which ignores anisotropy in the basis plane, no difference from formulas (57) and (64) emerges (to within the distinction in notation).

**4.3.2 Chromite.** In contrast to hematite,  $\operatorname{Cr}_2\operatorname{O}_3$  has a ground state with the CAS vector  $\mathbf{L}_c^0 \parallel Z$ . Here, the potential  $\Phi$  can be derived from expression (53) via the substitutions  $A_1 \to -A_1$ ,  $A_3 \to -A_3$ , and  $K_3 \to -K_3 < 0$ . Moreover, the AFE interaction in Eqn (53) is replaced with a combination of AFE and ME interactions, with the following respective terms

$$R_3(E_x L_{av} - E_v L_{ax}) - r_3(E_v M_v + E_x M_x),$$
 (59)

where the term with  $R_3$  represents the AFE interaction, and the term with  $r_3$  the ME interaction.

**4.3.3 Acoustic modes.** If we allow for the conditions  $d \le A$  and  $K \le A$ , from the LL equations we find the acoustic-mode frequencies of the corresponding transverse oscillations of  $(M_x, L_{cy})$  and  $(M_y, L_{cx})$ :

$$\omega_{AK1} = \omega_{AK2} \equiv \omega_{AK} = \omega_0 [(A_0 + A_3)\tilde{K}_3]^{1/2},$$
 (60)

where  $\widetilde{K}_3$  is the effective anisotropy constant, which by order of magnitude is purely relativistic:

$$\widetilde{K}_3 = K_3 - d'^2(A_2 + A_3)^{-1}$$
.

<sup>&</sup>lt;sup>4</sup> These expressions (e.g., see Ref. [3]) known from many old papers were not given in Ref. [11].

Here, magnetization holds the contribution of two terms: the Zeeman term with the magnetic susceptibility

$$\chi_{\perp} \equiv \chi_{xx}(\omega) = \chi_{yy}(\omega) = \frac{\omega_0^2 \widetilde{K}_3}{\omega_{AK}^2 - \omega^2}$$

and the magnetoelectric term [caused by the ME interaction; the term with  $r_3$  in expression (59)]. Here, first one finds  $\mathbf{M}_{\perp}$  from the LL equations, and then expression (59) is used to determine the effective electric polarization  $\mathbf{P}_{\perp} = \varkappa_{\perp}(\omega)\mathbf{E}_{\perp}$ . In the latter case, the result is as follows

$$\varkappa_{\perp}(\omega) = \omega_0^2 r_3^2 \widetilde{K}_3 (\omega_{AK}^2 - \omega^2)^{-1}.$$

By comparing the susceptibilities  $\chi_{\perp}(\omega)$  and  $\varkappa_{\perp}(\omega)$ , we find that the ratio of the intensities of the ME and AFM resonances (i.e., when  $\mathbf{P}_{\perp}(\omega)$  is excited by the field  $\mathbf{E}_{\perp}$ , and  $\mathbf{M}_{\perp}$  by  $\mathbf{H}_{\perp}$ ) is determined by the AFE constant  $r_3$ .

**4.3.4 Exchange modes.** The exchange modes are determined by the transverse oscillations of  $(L_{ax}, L_{by})$  and  $(L_{ay}, L_{bx})$ ; and again, one vector must be centrally symmetric, and the other centrally antisymmetric. The natural frequency of an exchange mode (at  $H_0 = 0$ ) is given by the formula

$$\omega_{E1}^2 = \omega_{E2}^2 \equiv \omega_E^2 = \gamma^2 (4M_0)^2 \left[ (A_1 - A_3)(A_2 - A_3) + (A_1 + A_2 - 2A_3)K_3 + d'(d' + 2d)(A_0 - A_3)(A_2 - A_3)^{-1} \right].$$

However, the variables (60) which enter into the acoustic modes are mixed into these oscillations by interactions of the Dzyaloshinskii type (the term with coefficients d and d'), but at the same exchange frequency  $\omega_E$ . As a result, in this (exchange) frequency range, the following susceptibilities are nonzero:

$$\chi_{xx}(\omega) = \chi_{yy}(\omega) \equiv \chi_{\perp} = \frac{\omega_0^2 d'^2 (A_2 - A_3)^{-1}}{\omega_E^2 - \omega^2},$$
(61)

$$\varkappa_{xx}(\omega) = \varkappa_{yy}(\omega) \equiv \varkappa_{\perp} \approx \frac{R_3^2 \omega_0^2 (A_2 - A_3)}{\omega_F^2 - \omega^2} . \tag{62}$$

Actually, the magnetic susceptibility  $\chi_{\perp}$  (61) is related to the Zeeman energy. It contributes to the exchange mode due to the Dzyaloshinskii interaction [in this case the term with d' in formula (53)] which generates an exchange pole for the acoustic-mode variables, too. Here, the magnetic contribution will, probably, be small compared to the antiferroelectric contribution, since

$$\frac{\chi_{\perp}(\omega)}{\varkappa_{\perp}(\omega)} = \frac{d'^2}{(A_2 - A_3)^2 R_3^2}.$$

Indeed, according to the estimates made by Krivoruchko and Yablonskii [11], one has

$$\frac{d'^2}{(A_2 - A_3)^2} \approx \frac{H_A}{H_E} \approx 10^{-4}, \quad R_3^2 \approx 10^{-3}.$$

Thus, the conditions needed for detecting exchange modes in  $Cr_2O_3$  by electric-field absorption are more favorable than those by magnetic-field absorption.

Finally, comparing formulas (47) (Fe<sub>2</sub>TeO<sub>6</sub>) and (62) (Cr<sub>2</sub>O<sub>3</sub>), we find that

$$\frac{\varkappa_{\perp}(\text{Fe}_2\text{TeO}_6)}{\varkappa_{\perp}(\text{Cr}_2\text{O}_3)} \sim \frac{f^2}{R_3^2} \,. \tag{63}$$

Since f and  $R_3$  are, respectively, the relativistic and exchangerelativistic constants with  $f \ll R_3$ , formula (63) implies that the intensity of an exchange AFE resonance for  $Cr_2O_3$  is, probably, higher than that for  $Fe_2TeO_6$ , so that we can expect that the observation of this effect in the first compound will be easier than in the second compound (incidentally, their Néel points do not differ very much: they are 310 and 290 K, respectively).

The ever increasing volume of the present review makes it impossible to discuss and compare the results of the work of the Ukrainian researchers and the researchers from the Ural region with other magnetic substances, such as orthoferrites (cf. Refs [12] and [24]).

### 5. Antimagnons in ferrimagnets

The theoretical aspects of a behavior of antimagnons in the tetragonal Mn<sub>2</sub>Sb ferrimagnet have been thoroughly studied in a recent paper by Mirsaev and Turov [25]. Therefore, in this section we discuss two-position ferrimagnets only briefly, with the focus on the features that make these ferrimagnets different from those examined in Section 4 (single-position ferromagnets and antiferromagnets).

## 5.1 The crystallochemical and magnetic structures of the Mn<sub>2</sub>Sb ferrimagnet

The unit cell of the intermetallic  $Mn_2Sb$  compound (Curie point  $T_C = 550$  K) contains two pairs of magnetic Mn ions which occupy the positions a (MnI) and c (MnII) (Fig. 3). A characteristic feature is that in each pair the ions are coupled through the symmetry center [26, 31], i.e., the criterion of existence of electroactive antimagnons is true for both pairs.

The tetragonal  $\mathrm{Mn_2Sb}$  ferrimagnet has a lattice whose symmetry is described by the  $P4/nmm(D_{4h}^7)$  group. The neutron diffraction research done by Wilkinson et al. [31] has shown that the exchange magnetic structure of this compound consists of (001) planes that incorporate MnI and MnII atoms belonging to different positions and, therefore, having different magnetic moments.

Figure 3 shows that the atoms from the positions a 1(0,0,1/2) and 2(1/2,1/2,1/2) and the positions c 3(0,1/2,1/2+z) and 4(1/2,0,1/2-z) are indeed related to each other (within their limits) through the symmetry center  $\overline{\bf 1}$ . At temperatures ranging from 240 K to  $T_{\rm C}=550$  K, the atomic magnetic moments of Mn<sub>2</sub>Sb are parallel to the crystallographic axis  $\bf 4$  (easy axis), while in the temperature interval from 0 to 240 K they are at right angles to the c-axis (easy plane) [26].

Depicted in Fig. 3 is a unit cell incorporating a group of atoms consisting of three layers, MnII-MnI-MnII (four atoms per unit cell), which repeat themselves along the Z-axis. Within an each group of this kind, the total magnetic moment is nonzero, since the magnetic moments of MnI (position a) and MnII (position c) are different and amount to  $2.13 \pm 0.20\mu_{\rm B}$  and  $3.87 \pm 0.40\mu_{\rm B}$ , respectively [31]. The magnetic moments of the MnI and MnII layers are antiparallel in the easy-axis and easy-plane states, which is a characteristic feature of ferrimagnets. In the paper [26], both

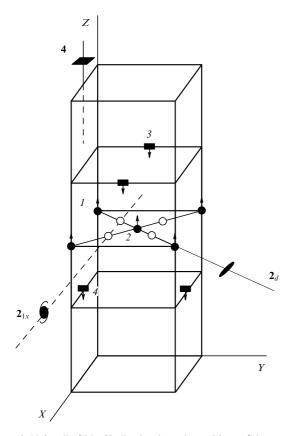


Figure 3. Unit cell of Mn<sub>2</sub>Sb. Depicted are the positions of the magnetic ions MnI (black circles) and MnII (black rectangles) and the symmetry elements, the generators of group P4/nmm ( $D_{4h}^7$ ): the symmetry center  $\overline{\mathbf{I}}$  (open circles) and the rotational axis  $\mathbf{4} \parallel Z$  and  $\mathbf{2}_d \parallel$  (110); also shown is the rotational axis  $\mathbf{2}_{1x}$ . Represented is the magnetic structure of the easy-axis type:  $\mathbf{M}_1 \uparrow \uparrow \mathbf{M}_2 \downarrow \uparrow \mathbf{M}_3 \uparrow \uparrow \mathbf{M}_4 \parallel Z \parallel \mathbf{4}$ .

orientation states were consistently examined, but here we give only a few results pertaining to the easy-axis state.

It has proven convenient to consider the four-sublattice exchange magnetic structure of  $\mathbf{M}_{n_2}\mathbf{S}\mathbf{b}$  with the magnetizations  $\mathbf{M}_{v}$  (v=1,2,3,4) a collection of two-sublattice subsystems a and c with basis vectors  $\mathbf{M}_{\xi}$  of ferromagnetism and  $\mathbf{L}_{\xi}$  of antiferromagnetism ( $\xi=a,c$ ):

$$\mathbf{M}_a = \mathbf{M}_1 + \mathbf{M}_2$$
,  $\mathbf{L}_a = \mathbf{M}_1 - \mathbf{M}_2$ ,  
 $\mathbf{M}_c = \mathbf{M}_3 + \mathbf{M}_4$ ,  $\mathbf{L}_c = \mathbf{M}_3 - \mathbf{M}_4$ .

Figure 3 also depicts the independent elements of the crystallographic space symmetry — the symmetry center  $\overline{\bf 1}$ , the four-fold symmetry axis  ${\bf 4}\parallel Z$ , and the diagonal binary symmetry axis  ${\bf 2}_d\parallel (110)$  — which can be considered the generators of group  $P{\bf 4}/nmm$ . In terms of these symmetry elements, the type of each position can be coded by indicating how they interchange the ions: into themselves  $(1\leftrightarrow 1, 2\leftrightarrow 2$  and  $3\leftrightarrow 3, 4\leftrightarrow 4)$  or into each other  $(1\leftrightarrow 2)$  and  $(1\leftrightarrow 2)$  and  $(1\leftrightarrow 2)$  and  $(1\leftrightarrow 2)$  are 'minus' sign in parentheses. Figure 3 clearly shows that the following permutation codes correspond to the positions in question:  $(1\leftrightarrow 2)$  and  $(1\leftrightarrow 2)$ 

Note that if instead of the  $\mathbf{2}_d$  axis we use the binary axis  $\mathbf{2}_{1x} = \mathbf{2}_d \mathbf{4}_z$  (which is also displayed in the figure), for both positions it turns out that  $\mathbf{2}_{1x} \equiv \mathbf{2}_x(-)$ , and the codes a and c differ only in the sign in front of the  $\mathbf{4}_z$  axis ('minus' or 'plus'). We note once again that here we are dealing only with the

**Table 2.** Rules of transformations of the M, L, H, and E vectors, positions a and c, group  $D_{4h}^7$ .

_		<b>=</b> / \		• ( )	4 ( )	4 ( )
$\Gamma_i$	H, E, M, L	<u>1</u> (-)	$2_{x}(-)$	$2_{y}(-)$	$4_{z}(-)$ $(a)$	$4_{z}(+)$ (c)
$\Gamma_1$	$M_{x}, H_{x}$	+1	+1	-1	$M_y, H_y$	
$\Gamma_2$	$M_y, H_y$	+1	-1	+1	$-M_{\scriptscriptstyle X},-H_{\scriptscriptstyle X}$	
$\Gamma_3$	$M_z, H_z$	+1	-1	-1	$M_z, H_z$	
$\Gamma_4$	$L_x, E_y$	-1	-1	+1	$-L_y, -E_x$	$L_y, -E_x$
$\Gamma_5$	$L_y, E_x$	-1	+1	-1	$L_x, E_y$	$-L_x, E_y$
	$L_z$	-1	+1	+1	$-L_z$	$L_z$
	$E_z$	-1	-1	-1	$L_y, -E_x$	

permutation properties of the symmetry elements in relation to a chosen position, irrespective of the type of the exchange magnetic structure (ferromagnetic, ferrimagnetic, or antiferromagnetic). Thus, for antimagnons (excited by an electric field) to exist, the condition  $\overline{\bf 1} \equiv \overline{\bf 1}(-)$  must be met, which in the present case, as noted earlier, holds for both positions.

### 5.2 Oscillation modes and thermodynamic potential

The calculation of the natural oscillation frequencies gets much simpler if for the ground state (phase) in question we are able to establish how the dynamical (oscillation) variables are divided into what is known as spin-wave representations for this phase [8, 23, 25].

For this we must once again build, as we did earlier, a table of transformations (see Table 2) for the rhombic subgroup Pmmn of the true tetragonal group P4/nmm (the four columns to the left of the double vertical line), to which we must add the rules of the transformations of the same variables caused by the symmetry axes  $\mathbf{4}_z(+)$  and  $\mathbf{4}_z(-)$ . Let us now examine only the spin-wave representations (oscillaeasy-axis for the modes) phase,  $\mathbf{M}_a \downarrow \uparrow \mathbf{M}_c \parallel \mathbf{H} \parallel Z$ . As before, using Table 2 and rule (8), we establish how the oscillation variables  $\Delta M_{\xi i}$ ,  $\Delta L_{\xi j}$  ( $\xi = a, c$ ; i, j = x, y) are divided into modes. For both positions, we get the same spin-wave representations:  $\Gamma_{12}(\Delta M_{\xi x}, \Delta M_{\xi y})$  for the two ferromagnetic modes, and  $\Gamma_{45}(\Delta L_{\xi x}, \Delta L_{\xi y})$  for the two antimagnon modes. Further symmetry considerations that allow for the symmetry axes  $\mathbf{4}_z(+)$  ( $\xi = c$ ) and  $\mathbf{4}_z(-)$  ( $\xi = a$ ) show that the modes  $\Gamma_{12}$  and  $\Gamma_{45}$  remain independent in the tetragonal case, too. What remains is to use Table 2 and write out the invariant expression for the thermodynamic potential  $\Phi$ . Then, isolating in this expression the quadratic form in the respective oscillation variables for each mode, we must solve equation (41). Here, we allow for the equal-modulus conditions  $|\mathbf{M}_1| = |\mathbf{M}_2| = M_a$  and  $|\mathbf{M}_3| = |\mathbf{M}_4| = M_c$  or, putting it differently, the conditions

$$\mathbf{M}_{\xi}^2 + \mathbf{L}_{\xi}^2 = (2M_{\xi})^2 \,, \quad \mathbf{M}_{\xi} \mathbf{L}_{\xi} = 0 \quad (\xi = a, c) \,.$$

As a result, for the part of  $\Phi$  that is quadratic in the homogeneous oscillations  $L_{\xi x}$  and  $L_{\xi y}$  and corresponds to antimagnon modes, we have

$$\Phi_{2} = \frac{1}{2} \widetilde{A}_{a} (L_{ax}^{2} + L_{ay}^{2}) + \frac{1}{2} \widetilde{A}_{c} (L_{cx}^{2} + L_{cy}^{2}) 
+ \lambda (L_{ax} L_{cx} - L_{ay} L_{cy}) 
- D_{a} (E_{x} L_{ax} - E_{y} L_{ay}) - D_{c} (E_{x} L_{cx} + E_{y} L_{cy}).$$
(64)

Here,  $\widetilde{A}_a$  and  $\widetilde{A}_c$  are the exchange constants that have been renormalized once more, but now by the interposition exchange interaction B, by the single-position  $K_{a,c}$  and the interposition  $K_{ac}$  anisotropies, and by the external field  $\mathbf{H} \parallel \mathbf{M}_0 \parallel Z (\mathbf{M}_0 = \mathbf{M}_a^0 + \mathbf{M}_c^0)$ :

$$\begin{split} \widetilde{A}_{a} &= A_{a} + K_{a} + \frac{(B - K_{ac})2M_{c} - H_{z}}{2M_{a}} \;, \\ \widetilde{A}_{c} &= A_{c} + K_{c} + \frac{(B - K_{ac})2M_{a} + H_{z}}{2M_{c}} \;. \end{split}$$

The quadratic form in  $M_{\xi x}$  and  $M_{\xi y}$ , which determines the *ferromagnon* modes, separates from expression (64) (there are no common terms). For these components, we arrive at well-known results (e.g., similar to those given in Ref. [3]) which we will not consider here, since ferromagnon modes are not excited by an electric field.

#### 5.3 Natural antimagnon frequencies

To find the oscillation frequencies and the amplitude of antimagnons excited by an alternating electric field  $E_{x,y}(t) \propto \exp(-i\omega t)$ , we employ LL equations of the types (13) and (14) for each position  $\xi = a, c$ .

From the LL equation (14) it follows that the purely antiferromagnetic mode  $\Gamma_{45}(L_{\xi x}, L_{\xi y})$  of the phase  $\Gamma_{3}(M_{\xi z})$  with  $\mathbf{M}_{a}^{0} \downarrow \uparrow \mathbf{M}_{c}^{0} \parallel Z$  is described by the equations

$$\dot{L}_{\xi x} + \Gamma L_{\xi x} = -\gamma M_{\xi z}^{0} \frac{\partial \Phi_{2}}{\partial L_{\xi y}}, 
\dot{L}_{\xi y} + \Gamma L_{\xi y} = \gamma M_{\xi z}^{0} \frac{\partial \Phi_{2}}{\partial L_{\xi y}} \qquad (\xi = a, c),$$
(65)

into which we have introduced the damping parameter  $\Gamma$  in the simplest way (according to Bloch), and we have assumed that  $\Gamma_a = \Gamma_c = \Gamma$ . The full solution of these equations can be found in Ref. [26]. In the same paper, the phase transitions in temperature and field **H** are examined, and so is their effect on the spectrum. Here, however, we are interested in the phase transitions between easy-axis and easy-plane states or, to put it differently, in the region of stability of the easy-axis state. This state is temperature-stable in the interval from 240 K to  $T_{\rm C} = 550$  K, which corresponds to the condition

$$4K_aM_a^2 + 4K_cM_c^2 + 8K_{ac}M_aM_c > 0$$

in a field  $\mathbf{H} \parallel Z$  with the field strength  $H_z$  in the interval from 0 to  $H_{B1} = 2B(M_c - M_a)$ .

For such field strengths  $H_z$  and temperatures T, equations (65) yield antimagnons with the frequencies  $\omega_1$  and  $\omega_2$  given by the following relations

$$\omega_{1,2}^2 = \frac{1}{2} \left( \omega_c^2 + \omega_a^2 + 2\omega_\lambda^2 \pm \left( \omega_c + \omega_a \right) \sqrt{\left( \omega_c - \omega_a \right)^2 + 4\omega_\lambda^2} \right),$$

where

$$\begin{split} \omega_c &= \gamma \big[ (A_c + K_c) 2 M_c + (B + K_{ac}) 2 M_a + H_z \big] , \\ \omega_a &= \gamma \big[ (A_a + K_a) 2 M_a + (B + K_{ac}) 2 M_c - H_z \big] , \\ \omega_\lambda^2 &= 4 \gamma^2 \lambda^2 M_a M_c . \end{split}$$

The appropriate formulas for the frequencies in the easy-plane state can be found in Ref. [26].

The interposition coupling of the a and c antimagnons is characterized by the relativistic coupling parameter  $\lambda \ll A_{\xi}$ .

At  $\lambda=0$  in expression (64),  $\omega_1=\omega_a$  and  $\omega_2=\omega_c$  are the frequencies of two independent antimagnons, separately for each position a and c (incidentally, both frequencies depend through B on the exchange interaction between the positions). The parameter  $\lambda$  governs the weak coupling between these antimagnons. We believe it would be interesting to do an experimental investigation of the dependence of the  $\omega_1$  and  $\omega_2$  frequencies of the Mn<sub>2</sub>Sb ferrimagnet on the strength  $H_z$  of the external magnetic field.

## 6. NMR excitation by an electric field in two-sublattice ferromagnets

In all the magnetic structures (phases) and orientation states mentioned earlier, the buildup of  $\mathbf{M}_{v}$  oscillations brought about by an alternating electric field  $\mathbf{E}(t)$  at low frequencies  $\omega \approx \omega_{n}$  (where  $\omega_{n}$  is the NMR frequency) leads, as a result of ME and AFE interactions, to one more (low-frequency) phenomenon: nuclear magnetoelectric resonance. Here, if (as is often the case) the frequency indicated is low compared to natural frequencies of oscillations of the electron subsystem (the magnon frequency  $\omega_{\rm mag}$ ),  $|\omega_{n}| \ll \omega_{\rm mag}$ , the oscillations follow in a quasiequilibrium manner the oscillations of the field  $\mathbf{E}(t)$  (see Ref. [8, p. 67]).

The excitation of nuclear spins by an electric field occurs because of hyperfine (HF) interaction. In the simplest model of such an interaction, which we also apply here, it is assumed that the nuclear spin interacts only with its parent atom (the nuclear magnetization  $\mathbf{m}_{\nu}$  interacts only with the inherent electron magnetization  $\mathbf{M}_{\nu}$ ).

Let us consider a magnetic substance with two single-position magnetic sublattices. We introduce the vectors of nuclear magnetization  $\mathbf{m}$  and antiferromagnetism  $\mathbf{l}$  in the same way as we earlier introduced the vectors  $\mathbf{M}$  (1) and  $\mathbf{L}$  (2) for the electron subsystem (we note the obvious fact that the transformation properties of the vectors  $\mathbf{m}$  and  $\mathbf{M}$ , as well as  $\mathbf{l}$  and  $\mathbf{L}$  are the same). In this case, the HF interaction can be written as follows:

$$F(\mathbf{m}_1 \mathbf{M}_1 + \mathbf{m}_2 \mathbf{M}_2) \equiv \frac{1}{2} F(\mathbf{m} \mathbf{M} + \mathbf{l} \mathbf{L}).$$
 (66)

Here, we have denoted the HF interaction parameter by F (in contrast to the notation commonly used in the literature [29]) to distinguish it from the exchange parameters introduced earlier. It is also assumed that F is a scalar (HF interaction is purely isotropic).

We study only the FM phase with  $\mathbf{M}_1 \uparrow \uparrow \mathbf{M}_2 || \mathbf{M} || \mathbf{H} || Z$  (representation  $\Gamma_3(M_z)$  in Table 1). In contrast to the FM structure  $\mathbf{M} || \mathbf{H} || X (\Gamma_1(M_x))$  discussed in Section 2.1, here the magnetization  $\mathbf{M}^0$  is directed along the odd symmetry axis  $\mathbf{2}_z(-)$ , which simplifies the oscillation part of the ME interaction. For the thermodynamic potential density we must take  $\Phi$  (9), where, in accordance with Table 1 and rule (8), the following substitutions in the energy of second-order magnetic anisotropy must be made:  $M_y \to M_x$ , and  $M_z \to M_y$ . However, to simplify matters we ignore this energy, and from the exchange energy (the terms with  $A_L$  and  $A_M$ ), bearing in mind the equal-modulus property (10), we exclude the term with  $\mathbf{M}^2$ , which renormalizes the constant  $A_L$ , i.e., leads to the substitution  $A_L \to \widetilde{A_L} = A_L - A_M$  ( $A_M < 0$ ).

In accordance with the results of Section 2 and bearing in mind the above remarks, we can write down the thermodynamic potential of the electron-nuclear system as follows:

$$\Phi = \frac{1}{2} \widetilde{A_L} \mathbf{L}^2 - (m_z + M_z) H_z + \frac{1}{2} F(\mathbf{Mm} + \mathbf{Ll})$$
$$- \frac{M_z^0 (s_L L_x + s_l l_x) E_z}{2M_0} \qquad (M_z^0 = 2M_0).$$
 (67)

In addition to taking into account the ME interaction for the electron subsystem (the term with  $s_L$ ), we have allowed for a similar interaction between the nuclear spins and the field  $\mathbf{E}(t)$  through the same electron subsystem (the term with  $s_l$ ). Moreover,  $\Phi$  also incorporates the HF interaction (66) between these subsystems.

The NMER problem is solved on the basis of a system of two LL equations for  $\mathbf{m}_1$  and  $\mathbf{m}_2$ . After we have gone over to the variables  $\mathbf{m}$  and  $\mathbf{l}$ , these equations acquire the forms (13), (14) with the substitution  $\mathbf{M}, \mathbf{L}, \gamma \to \mathbf{m}, \mathbf{l}, -\gamma_n$  (the nuclear gyromagnetic ratio, in contrast to the similar ratio for the electron system, can be either positive or negative for different nuclei). In the linear approximation in the oscillation variables with the field  $E_z \propto \exp(-i\omega t)$  and without an alternating magnetic field  $\mathbf{H}(t)$ , one of these equations yields the free precession of vector  $\mathbf{m}$  about the constant field  $H_{nz}^0 = H_z - FM_0$ , which allows for the hyperfine field  $H_n = -FM_0$  (being usually negative).

The second LL equation considered here determines the  $l_x$  and  $l_y$  oscillations caused by the field  $\mathbf{E}(t)$ . Here, we must allow for two channels of action of  $\mathbf{E}(t)$  on the variables. First, through the term with  $s_l$  in expression (67), the field  $E_z$  directly causes the component  $l_x$  to oscillate (which means that the component  $l_y$  will oscillate, too). Second, due to the term with  $s_L$  in expression (67), the electric field, as noted earlier, will 'sway'  $L_x$ , which, in turn, again causes  $l_x$  to oscillate [through the HF interaction (66) that enters into  $\Phi$  (67)].

In conditions of quasiequilibrium coupling between **L** and **E**(t) (mentioned earlier), the quantities  $L_j$  can be found from the conditions  $\partial \Phi / \partial L_x = 0$  and  $\partial \Phi / \partial L_y = 0$ , which yield

$$L_x = \frac{s_L E_z - F l_x / 2}{\widetilde{A}} , \qquad L_y = -\frac{F}{2\widetilde{A}} l_x . \tag{68}$$

If we allow for Eqn (68), the system of LL equations for  $l_x$  and  $l_y$  becomes closed. Solving it, we find the sought quantities with allowance for both channels of action of the field  $\mathbf{E}(t)$  on the nuclear subsystem:

$$l_x \approx \chi_n(\omega) \left( s_l - \frac{F}{2\widetilde{A}} s_L \right) E_z \,, \qquad l_y \approx \mathrm{i} \, \frac{\omega}{\omega_n} \, l_x \,,$$

where

$$\chi_n(\omega) \approx \chi_{n0} \frac{\omega_n^2}{\omega_n^2 - \omega^2}$$

is the dynamic, and  $\chi_{n0}$  the static NMR susceptibilities, and  $\omega_n \approx \gamma_n (H_z - FM_0)$  is the NMER frequency which differs from the ordinary NMR frequency by the presence of a small correction of order

$$\frac{\chi_{n0}F^2}{4\widetilde{A}} \approx \frac{F}{4\widetilde{A}} \frac{m_n}{M_0} \ll 1$$

 $(m_n$  is the absolute value of the nuclear magnetization), which is not taken into account here.

According to expression (67), the excitation of L oscillations by an electric field is equivalent to the excitation of the polarization vector  $\mathbf{P} \parallel Z$ :

$$P_z = -\frac{\partial \Phi}{\partial E_z} = s_L L_x + s_l l_x \,,$$

where  $L_x$  also incorporates the second term in Eqn (68), related to the HF interaction (66). As a result, we find that the total effective polarization that is caused only by NMER and allows for the contribution from

$$\Delta L_{x} = -\frac{F}{2\widetilde{A}} l_{x}$$

is given by the following formula

$$P_n = \left(s_l - \frac{F}{2\widetilde{A}} s_L\right) l_x = \chi_n(\omega) \left(s_l - \frac{F}{2\widetilde{A}} s_L\right)^2 E_z.$$

This formula describes the linear dynamic response of the ferromagnet to an electric field whose frequency is close to the NMR frequency. Actually, the sample must be located in the antinodes of the field  $\mathbf{E}(t)$ , in contrast to ordinary NMR, where the sample is in the antinodes of the field  $\mathbf{H}(t) \propto \exp(-\mathrm{i}\omega t)$ .

The experimental method merits a special discussion. Unfortunately, so far nothing can be said about the magnitude of the effect, since the constants  $s_l$  and  $s_L$  remain unknown in the model under discussion. However, some quantitative estimates (within extremely broad limits) that use experimental data on the static ME effect in other magnetic substances can be made, so that one may infer that we are dealing here with a new experimentally measurable phenomenon, although in the static case (reconstruction of L by a static electric field) there is no such an effect.

In concluding this section a remark is in order. Above we described only the principles underlying the NMER phenomenon caused by the ME and AFE interactions and used only a simple model of a two-sublattice ferromagnet, with no direct reference to specific magnetic substances. However, we consistently allowed for both NMER channels, in contrast to previous papers [8, 9, 25] where only one channel was taken into consideration. A fuller analysis that agrees with the present section and concerns the NMER effect in the Mn<sub>2</sub>Sb ferrimagnet was done in a recent paper written by Kurkin, Mirsaev, and Turov [32]. Mn<sub>2</sub>Sb is probably the most promising magnetic substance for experimental examination of both electroactive antimagnons (see Section 5) and NMER.

### 7. Concluding remarks

Despite the long history of spin-wave dynamics, a large section of it, devoted to the effect of an alternating electric field on the properties of magnetic substances, has never really been developed, although it would seem to be of practical interest and have certain prospects for discovering new physical phenomena.

The present review focuses not only on filling this gap in the physics of magnetic phenomena, but also on acquiring an understanding of the reasons for its emergence, both objective and subjective. The reasons are conditioned by the existence of special features of multiple-points positions occupied by the magnetic atoms of the substance, with the features occurring very often. It is enough to flip through the International Tables for X-ray Crystallography [15] to see that the situation where the symmetry center  $\overline{\bf 1}$  is not one of the elements of local (island) symmetry of the position occurs very often.

The element  $\overline{1}$  must perform a nonidentity permutation of the atoms in this position (see, in particular, the code (37) of position 4e for trirutiles, and also Fig. 1 for a two-fold position). Even in the most thorough works (reviews and monographs) on the physics of magnetic phenomena, usually no attention is paid to the existence of such positions of magnetic atoms or, precisely, to the properties associated with these positions. Yablonskii and Krivoruchko [10-14] were the first not to follow this tradition. However, as noted earlier, their ideas and results did not promote the development of a new section in the physics of magnetic phenomena. In the monograph on magnetism [3], where spin dynamics occupies a sizable portion of the material, this area was not covered either. Meanwhile, the of dynamics development of these ideas could have led to a new, fairly broad area of spin-wave dynamics. It is our hope that the present review will serve as a 'trigger' and will attract the attention of researchers to the new section of magnetodynamics — there is certainly a need for this (paper [8] and the present review should be considered an additional chapter to monograph [3]).

Here, we touched only on a few phenomena related to that area of spin-wave dynamics. (The reader should know that paper [8] covers more than a dozen additional problems waiting for their theoretical development and experimental verification.) Note, for instance, that the existence of antimagnons as a new type of spin waves may manifest itself in the physics of magnetic phenomena not only as a result of the combined action of ME and AFE interactions but also in the absence of the field E (thermodynamics and magnetoacoustics), in nonlinear phenomena, etc. We believe that some of the ideas considered in this review and the related phenomena will soon be discussed in textbooks on magnetism. In particular, the textbooks will contain figures like Fig. 2 depicting antimagnons (in addition to ferromagnons) and including the inhomogeneous case. We also believe that experimenters should do everything possible to detect these new interesting phenomena predicted theoretically.

In conclusion, we list some additional promising areas of research (both theoretical and experimental) in the present section of magnetodynamics (they have been partially listed in Ref. [8]).

- Surface electroactive magnons (including antimagnons).
- Interaction between electroactive magnons and hypersound.
- Nonlinear phenomena in electromagnetoelastic dynamics.
- Longitudinal (in relation to the fundamental basis vector) electroactive oscillations (waves).
- Electroactive oscillations in multilayer ferromagnetic and antiferromagnetic macrostructures (see Ref. [8]).
- Cubic ferrimagnets in the presence of ME and/or AFE interactions. The study of such ferrimagnets will make it possible to develop an approach to the important problem of spin-wave dynamics of the famous yttrium-iron garnet (YIG) Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>. We are speaking of the existence of electroactive magnons excited by the above-mentioned interactions. There

are experimental data (true, rather contradictory) pointing to the presence of an ME effect in YIG (e.g., see Refs [33–35]). The magnetic structure of this ferrimagnet contains 40 magnetic sublattices, of which 16 occupy the a positions (these are known as a-ions) with the local symmetry  $\{\overline{3}\}\$ , while 24 sublattices occupy the d position (these are known as dions) with the local symmetry  $\{\overline{4}\}$ . The space symmetry of YIG is  $O_h^{10} \equiv Ia3d$ . The magnon spectrum of this compound has been thoroughly studied by Kolokolov et al. [36] but, first, without allowing for ME and AFE interactions (i.e., at E = 0) and, second, with no mention of antimagnons. To avoid the difficulties in calculations involving such a large number of sublattices (actually, thanks to translations related to body centering (I) we can limit ourselves to 20 sublattices), the authors suggest first considering a simplified model of a collinear cubic ferromagnet with a smaller number of sublattices with noncentrally symmetric local symmetry, e.g., for magnetic ions in the six-fold position  $6f\{4\}$  of group  $O_h^1 \equiv Pm3m$  with six magnetic sublattices. It is hoped that this model will help answer some questions concerning YIG dynamics in the field  $\mathbf{E}(t)$ , and especially the question of whether antimagnons actually exist.

• Electroactive and magnetoactive magnons in exchangenoncollinear antiferromagnets [up to now we have spoken of collinear and weakly (relativistically) noncollinear magnetic substances]. This problem was not treated in the current review, although it is of substantial interest in connection with important features and has been discussed in several papers. Here, we will not examine the discussion of this topic. The point is that there is still no unified position even in relation to the space group of the magnetic substances in question. All this requires a special study of the dynamics of such exchange-noncollinear magnetic substances, which has been done in a paper by Mirsaev and Turov [37].

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