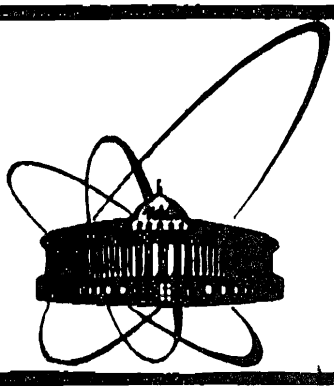

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TOROID CURRENT STRUCTURES IN FERRO AND ANTIFERROMAGNETS

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1. INTRODUCTION

It is known that the magnetic structure of matter in the thermodynamic equilibrium state is complitely determined by the microscopic current density $\vec{j}(\vec{r})$ and microscopic spin density $\vec{S}(\vec{r})$ at every point. In crystalline magnets the belonging to a space group of magnetic symmetry is characterised by the behaviour of functions $\vec{j}(\vec{r})$ and $\vec{S}(\vec{r})$ relative to space transformations and time inversion. However, in the macroscopic electrodynamics dealing with quantities averaged over a "physically infinitesimal" volume there arises the problem of adequate description of the magnetic state of a system in terms of respective macroscopic characteristics. A convenient mathematical approach, for constructing such quantities is the scheme of multipole parametrization. Choosing an appropriate complete set of basis functions one may obtain an infinite series of multipole characteristics: moments and their power radii. As a rule, for the problems of macroscopic physics it is important to know only some of them.

To study the orbital magnetic ordering, it is necessary to apply the procedure of multipole parametrization to the function $\vec{j}(\vec{r})$. Though this problem has a very long history, it is only quite recently that a clear understanding has been achieved of the multipole expansion of $\vec{j}(\vec{r})$ /1,2/ Here we shall discuss only some important points related with its formulation rather than with the solution, until now these points were not clearly presented in the literature. The main body of the paper is devoted to the problem of existence of the so-called toroid structures in crystals with ferro- and antiferromagnetic properties.

2. THE SIMPLEST DEFINITIONS OF CURRENT TOROID MOMENTS IN ELECTRODYNAMICS OF CONDENSED MATTER

Since in a steady state, the only object under consideration, the current is conserved, $div\,\vec{j}(\vec{r}) = 0$. The vector field $\vec{j}(\vec{r})$

contains as much as two functional-independent components (linearly independent in a conjugate to coordinate space of wave vectors: $\vec{k}\vec{j}(\vec{k}) = 0$. With this fact in mind, one may express the transverse field $\vec{j}(\vec{r}) \equiv \vec{j}_\perp(\vec{r})$ through two scalar fields $\psi(\vec{r})$ and $\chi(\vec{r})$ as follows:

$$\vec{j}_\perp(\vec{r}) = c\,rot\left(\vec{r}\psi(\vec{r})\right) + c\,rot\,rot\left(\vec{r}\chi(\vec{r})\right) \equiv \tag{1}$$

$$\equiv \vec{L}\psi + \vec{K}\chi\ ; \qquad \vec{L} = \frac{1}{c}\vec{\nabla}\times\vec{r},\ \vec{K} = \frac{1}{c}\vec{\nabla}\times\vec{L}.$$

We shall call this representation by the Neumann-Debye representation /3/.

The differential operators $\vec{L}$ and $\vec{K}$ obey the following operations of multiplication:

$$\vec{\nabla}\cdot\vec{L} = \vec{r}\cdot\vec{L} = \vec{\nabla}\cdot\vec{K} = \vec{L}\cdot\vec{K} = 0\ , \quad \vec{r}\cdot\vec{K} = -c\vec{L}^2. \tag{2}$$

Making use of these "rules" we perform the project (division) of components of the current density $\vec{j}_\perp(\vec{r})$:

$$\vec{L}\cdot\vec{j}_\perp = c^2L^2\psi(r,\theta,\varphi),\ \frac{1}{c^3}\vec{r}\cdot\vec{j} = -L^2\chi(r,\theta,\varphi). \tag{3}$$

One can easily see that the standard expansion ψ and χ over the solutions of the scalar Helmholtz equation $f_\ell(kr)Y_{\ell m}(\theta,\varphi)$:

$$\vec{L}\cdot\vec{j}_\perp = c^2L^2\sum_{\ell,m}\int k^2dk\,\alpha_\ell M_{\ell m}(k^2)f_\ell Y_{\ell m} = \sum_{\ell,m,k}\alpha_\ell M_{\ell m} f_\ell\,\ell(\ell+1)Y_{\ell m}, \tag{4}$$

$$\vec{r}\cdot\vec{j}_\perp = -c^2L^2\sum_{\ell,m}\int k^2dk\,\beta_\ell T_{\ell m}(k^2)f_\ell Y_{\ell m} = c\sum_{\ell,m,k}\beta_\ell T_{\ell m} f_\ell\,\ell(\ell+1)Y_{\ell m} \tag{5}$$

provides magnetic $M_{\ell m}$ and toroid $T_{\ell m}$ multipole moments easily normalised as follows:

$$M_{\ell m} = \frac{-1}{\ell+1}\sqrt{\frac{4\pi}{2\ell+1}}\int d^3r\,r^\ell Y^*_{\ell m}\,\frac{1}{c}\left[(\vec{r}\times\vec{\nabla})\cdot\vec{j}_\perp\right], \tag{6}$$

$$T_{\ell m} = \frac{1}{\ell+1}\sqrt{\frac{4\pi}{2\ell+1}}\int d^3r\,r^\ell Y^*_{\ell m}\,\frac{1}{c}\left(\vec{r}\cdot\vec{j}_\perp\right). \tag{7}$$

The set of moments (6) and (7) does not form a full parametrization of $\vec{j}_\perp$, as the system of radial harmonics if not closed in the three-dimensional space. The full parametrization includes together with each 1th moment, an infinite ($n = 0,1,\ldots$) series of parameters - $2n$ - power radii specifying radial distribution of the moment density: integrals (6,7) with changing $r^\ell \to r^{\ell+2n}$ In a systematic way

one can obtain such a representation by using the expansion $M_{\ell m}(k^2)$, $T_{\ell m}(k^2)$ into the power series over k^2 /1/. A particular role in (6) and (7) is played by the lowest (dipole) moments: magnetic dipole moment $\vec{M}$ and toroid dipole moment $\vec{T}$

$$\vec{M} = \frac{1}{2c}\int [\vec{r}\times\vec{j}_\perp]\, d^3r, \qquad (8)$$

$$\vec{T} = \frac{1}{2c}\int \vec{r}\,(\vec{r}\,\vec{j})\, d^3r. \qquad (9)$$

The current transversality condition in terms of the moments and their radii for weight $\ell = 1$ may be written in the form of a zero derivative with respect to time of the charge dipole mean-square radius

$$\dot{\vec{Q}}^{(n=2)} = \frac{1}{c}\int [2\vec{r}\,(\vec{r}\,\vec{j}) + r^2\vec{j}\,]\, d^3r = 0. \qquad (10)$$

Considering (10) as the relation between its two right members we get one more definition of the toroid moment $\vec{T}$:

$$\vec{T} = -\frac{1}{4c}\int r^2 \vec{j}_\perp\, d^3r. \qquad (11)$$

It is seen that the toroid dipole may be called a mean square radius of the current transverse part. From this a geometric shape of that dipole can easily deduced. In the simplest case of axial symmetry of the distribution $\vec{j}(\vec{r})$ integral (11) is a characteristic of a double cylindrical layer. Provided that the current lines are closed in a finite volume, this layer can be continuously transformed into a torus with poloidal currents on it. These geometrical properties of of the toroid-dipole model were predetermined by the type of symmetry of the operator $\vec{K}$. When there is no transversality condition ($\frac{1}{c}\vec{\nabla}\cdot\vec{j} = -\dot{\rho}$, where ρ is the charge density), it is necessary to make use of a more general definition of the toroid moment /1,2/

$$\vec{T} = \frac{1}{10c}\int [\vec{r}\,(\vec{r}\,\vec{j}) - 2r^2\vec{j}\,]\, d^3r \qquad (12)$$

that reduces to (9) or (11) if (10) is taken into account.

3. PROPER AND PSEUDOPROPER TOROID MAGNETISM IN CRYSTALS

From the multipole parametrization (6,7) it follows that the simplest ordered states of orbital magnets are the ferromagnetic (FM) and toroid states characterised by vectors $\vec{M}$ and $\vec{T}$, respectively. With definite symmetry properties, these vectors may serve as order parameters defining the genesis of a low-symmetry phase at magnetic second order phase transitions in crystals. The toroid ordering will further be called "the toroid current state" (TCS) /4/; and crystals with TCS are called the "toroics".

To establish what places is occupied by the toroics in the general classification of magnets, some definitions are to be recalled. From a group-theoretical point of view the antiferromagnets (AFM) are crystals of the Shubnikov white and black-white spatial symmetry with zero mean magnetization (hereafter we imply long-periodic structures). In this sense the toroics are antiferromagnets described by a vector order parameter $\vec{T}$ changing sign at the time inversion and characterized by the limit group of magnetic symmetry $\frac{\infty}{m}mm$. Out of 122 classes of magnetic symmetry, according to ref. /5/, 31 admit the existence of a vector with such transformation properties (the same number as the vector $\vec{M}$ with the limit group $\frac{\infty}{m}m'm'$). These numbers indicate that toroics should not be treated as exotic objects.

It should be emphasized that the very introduction of the polar-vector order parameter, that is odd with respect to the time inversion, for describing the AFM structure in crystals is not something essentially new. Indeed, consider how are things going in the spin AFM. Since the order parameter in AFM cannot be chosen universally (in contrast with FM where the choice of the magnetization vector $\vec{M}$ with the relevant symmetry is natural), it is worth while to consider two limiting cases.

In the case of AFM with localized spins the separation of the subsystem into independent magnetic lattices is the most effective approach. In this case as an order parameter the vector $\vec{L}$ is introduced, that is a linear combination of the magnetizations of sublatices and as a result, has an additional symmetry associated with interchange of atoms of various sublattices. The vector $\vec{L}$ may in fact have the symmetry properties that coincide in a concrete space group with the properties of the vector $\vec{T}$ (however, the vector $\vec{L}$ is never characterised by any limit group).

In the case of AFM with delocalised spins an approach is needed that is not based on the concept of magnetic sublattice. It has been proposed in [6,7] and further developed in [8]. The generic idea of these papers consists in the classification of magnets into groups of exchange symmetry (space crystalline group supplemented by three-dimensional rotations and reflections in the spin space). The order parameters in [6-8] are introduced as expansion coefficients of the spin density function $\vec{S}(\vec{r})$ over irreducible representations of the crystal symmetry space group. Among the quantities thus constructed there may appear those transforming over the vector irreducible representation isomorphic $\vec{T}$ under space transformations though not coinciding with $\vec{T}$ under spin rotations.

Thus, the definition of toroics as a separate class of magnets is due not to the new properties of space symmetry of the vector but to the orbital nature of its formation and specific properties of TCS (optical [9], magneto-optical [10], magneto-electric [11] and other anomalies). In this connection it is indispensable to study in detail concrete magnetic structures permitting TCS and to point out possible objects of experimental investigation. Such a statement of the problem determined the aim of the present paper.

In accordance with the general theory of second order phase transitions the toroid moment $\vec{T}$ may arise independently as a result of a spontaneous symmetry breaking (proper TCS). The microscopic model of such a transition has been proposed in Ref. [12]. However, the conditions for an proper TCS to arise in orbital AFM are rigorous enough and may probably be realizes rarely, which complicates their experimental detection. According to [13] a proper TCS may be realized in the nickel-iodine boracite. At the same time, it is reasonable to consider situations in which TCS appears as a result of another type of ordering. From this point of view it is most tempting to study a pseudoproper TCS in the spin FM and AFM due to invariants of the type TL and TM (or of a higher order in $\vec{L}$ and $\vec{M}$) in the thermodynamic potential. Note that for pseudoproper transitions (in the given case for TCS) an additional lowering of symmetry does not occur (in the given case of the FM or AFM structure). It is clear that a pseudoproper TCS should inevitably arise from relativistic corrections to interactions in crystals with the usual spin mechanism of FM or AFM if only the system symmetry admits the coexistence of $\vec{T}$ with $\vec{L}$ or $\vec{M}$, respectively.

Classification of possible types of coexistence of the vectors $\vec{L}$, $\vec{M}$ and $\vec{T}$ can be made on the basis of the results of [5] shown in table 1. It is to be noted that 18 magnetic classes admit coexistence of $\vec{L}$ and $\vec{T}$ but do not admit a weak FM; 13 classes admit coexistence of $\vec{M}$ and $\vec{T}$, and 7 classes admit under certain orientations of $\vec{L}$ TCS and (or) a weak FM. In this case a simultaneous appearance of both weak FM and TCS does not change the magnetic system symmetry.

Table 1. Toroid magnetic classes.

Type of ordering		Magnetic classes
magnetic	electric	
L	P	$mm2, 4mm, 3m, 6mm$
L	$\bar{P}$	$\bar{4}', \bar{4}'2'm, \bar{6}', \bar{6}'m2', \bar{1}', 2/m', 2'/m, mmm', 4/m', 4/m'mm, \bar{3}', \bar{3}'m, 6/m', 6/m'mm$
M	P	$3, 4, 6$
M	$\bar{P}$	$4\,2'2', 32', 62'2'$
L, M weak FM	P	$1, 2, m, 2', m', mm'2'$
L, M weak FM	$\bar{P}$	$2'2'2$

4. CONDITIONS FOR TCS IN THE SPIN FM AND AFM

In this paragraph we shall apply the method allowing us to formulate conditions for arising a pseudoproper TCS in the spin magnets. We mean the conditions imposed by the presence of a symmetry elements in the crystalline space group of a magnetic, by the position of magnetic atoms in a lattice and finally, by the nature of the magnetic spin ordering (orientation of the vectors $\vec{L}$ and $\vec{M}$ with respect to crystallographic directions). For all space crystalline groups we shall compile a table allowing classification of pseudoproper toroid magnets according to the type of invariants responsible for the appearance of TCS. Following the theory of weak ferromagnetism formulated by Dzyaloshinsky[14], one should analyse a possible existence of the relevant invariant combinations of the vectors ($\vec{T}$ and $\vec{L}$) and ($\vec{T}$ and $\vec{M}$). In its main features the approach used is analogous to that used by Turov[15] for classification of weak ferromagnets.

It should be noted that the AFM structure is called even with respect to a given symmetry elements if the relevant transformation interchanges magnetic moments within one and the same magnetic sublattice; in the case when magnetic moments of various magnetic sublattices are interchanged, the structure is called odd. Taking into account the symmetry properties of the vector $\vec{T}$, one may conclude that the toroid ordering is possible only in the AFM structures even with respect to all the translations and odd with respect to the symmetry centre if the one exists in the system (further, in describing concrete structures, the parity will be denoted by signs $(\pm)$, namely $6_z^{(\pm)}$, $1^{(\pm)}$, $2_z^{(\pm)}$ etc.).

Thus, for the AFM structures admitting TCS

a) the magnetic and chemical elementary cells should coincide;

b) the directions of magnetic moments in all the sites corresponding to the same Bravais lattice should coincide as well;

c) the directions of magnetic moments in the sites transforming into each other under space inversion should be strictly opposite.

For the FM structures admitting TCS, the points a) and b) are fulfilled automatically. However, the vector $\vec{M}$ is even with respect to space inversion and the vector $\vec{T}$ is odd, the point c) should be formulated differently;

d) the space group of the crystal symmetry should not have inversion. Though the very division of the FM structures into even or

Table 2. Invariants due to crystal symmetry elements.

N	Symmetry element	even (+) odd (−) structure	Invariants
1	$\bar{1}$	−	$L_xT_x, L_yT_y, L_zT_z, L_xT_y, L_xT_z, L_yT_x, L_yT_z, L_zT_x, L_zT_y$
2		+	—
3	2_z	+	$L_xT_x, L_xT_y, L_yT_x, L_yT_y, L_zT_z$
4		−	$L_xT_z, L_yT_z, L_zT_x, L_zT_z$
5	2_x	+	$L_xT_x, L_yT_y, L_yT_z, L_zT_y, L_zT_z$
6		−	$L_xT_y, L_xT_z, L_yT_x, L_zT_x$
7	2_y	+	$L_xT_x, L_xT_z, L_yT_y, L_zT_x, L_zT_z$
8		−	$L_xT_y, L_zT_y, L_yT_x, L_yT_z$
9	2_{xy}	+	$L_xT_x + L_yT_y, L_zT_z, L_xT_y + L_yT_x$
10		−	$L_xT_x - L_yT_y, L_xT_y - L_yT_x$
11	4_z	+	$L_xT_x + L_yT_y, L_zT_z, L_xT_y - L_yT_x,$ $(L_xT_y - L_yT_x)(L_x^2 + L_y^2), (L_xT_y - L_yT_x)L_z^2,$ $(L_xT_x + L_yT_y)L_z^2, T_zL_z^3, (L_xT_x + L_yT_y)(L_x^2 + L_y^2),$ $L_zT_z(L_x^2 + L_y^2), (L_xT_x - L_yT_y)L_xL_y, (L_xT_y + L_yT_x)L_xL_y,$ $(L_xT_y + L_yT_x)(L_x^2 - L_y^2), (L_xT_x - L_yT_y)(L_x^2 - L_y^2)$
12		−	$L_xT_x - L_yT_y, L_xT_y + L_yT_x,$ $(L_xT_x - L_yT_y)(L_x^2 + L_y^2), (L_xT_x - L_yT_y)L_z^2,$ $(L_xT_y + L_yT_x)(L_x^2 + L_y^2), (L_xT_y + L_yT_x)L_z^2,$ $(L_xT_x + L_yT_y)(L_x^2 - L_y^2), (L_xT_x + L_yT_y)L_xL_y,$ $L_zT_z(L_x^2 - L_y^2), L_zT_zL_xL_y$ $(L_xT_y - L_yT_x)(L_x^2 - L_y^2), (L_xT_y - L_yT_x)L_xL_y$
13	3_z	+	$L_zT_z, L_xT_x + L_yT_y, L_xT_y - L_yT_x,$ $T_z(L_x \pm iL_y)^3, (T_x \pm iT_y)(L_y \pm iL_y)^2L_z$
14		−	—
15	6_z	+	$L_zT_z, L_xT_x + L_yT_y, L_xT_y - L_yT_x$
16		−	$T_z(L_x \pm iL_y)^3, (T_x \pm iT_y)(L_x \pm iL_y)^2L_z$

odd ones is senseless, it is convenient for our purposes to use the fact that the symmetry properties of the AFM structure even with respect to some elements formally coincide with the relevant properties of the FM structure. Therefore, the table constructed below for invariant combinations of the vectors $\vec{T}$ and $\vec{L}$ by changing $\vec{L} \to \vec{M}$ for even AFM structures provides all invariant combinations of the vectors $\vec{T}$ and $\vec{M}$ in FM.

Let us make the table of invariant combinations of the vectors $\vec{L}$ and $\vec{T}$ under proper and improper rotations specific of the crystalline lattices (table 2). With the knowledge of the concrete crystallographic structure of a magnet, one can find the AFM structures admitting TCS as well as the direction of the vectors $\vec{T}$ at which $\vec{T} \neq 0$. In the general case one should use table 2 for the relevant group generators thus defining an invariant in the thermodynamic potential responsible for the appearance of TCS. Note that if magnetic atoms lie on a simple (not spiral) rotational symmetry axis or on a simple (not sliding) symmetry plane, the AFM structure is always even with respect to these elements.

Now we pass to the discussion of sufficient conditions for TCS in each system (see table 3). It is assumed further that the structure is even with respect to pure translations. Let us consider in more detail the AFM structures.

TRICLINIC SYSTEM (1)

If the positions of magnetic sites are single, there cannot be AFM ordering without increasing dimensions of the cell and TCS is also impossible. If the positions of magnetic sites are double and related by the symmetry centre, the TCS is possible (N2 by the Fedorov classification).

MONOCLINIC SYSTEM (2, m ; 2/m)

a) There is no symmetry centre (2, m) ; there is only one independent symmetry element: plane or second order axis with respect to which the AFM structure should be odd (NN 3-9). The magnetic sites in this case are necessarily in the general position;

b) There is symmetry centre (2/m), and since the AFM structure should necessarily be odd with respect to it, with respect to axis 2 the structure may be even and odd (NN 10-15). In this case the magnetic sites may be both in general and special crystallographic po-

Table 3. Classification of toroid antiferromagnets.

N	System (singony)	Space group	Type of structure	Invariants
1	Triclinic	2	$\bar{1}^{(-)}$	$L_xT_x, L_yT_y, L_zT_z, L_xT_y, L_xT_z, L_yT_x, L_yT_z, L_zT_x, L_zT_y$
2	Monoclinic	3-5	$2_z^{(-)}$	$L_xT_z, L_yT_z, L_zT_x, L_zT_y$
3		6-9	$\bar{2}_z^{(-)}$	$L_xT_x, L_yT_y, L_zT_z, L_xT_y, L_yT_x$
4		10-15	$\bar{1}^{(-)}, 2_z^{(+)}$	$L_xT_x, L_yT_y, L_zT_z, L_xT_y, L_yT_x$
5			$\bar{1}^{(-)}, 2_z^{(-)}$	$L_xT_z, L_yT_z, L_zT_x, L_zT_y$
6	Rombic	16-24	$2_z^{(+)}, 2_x^{(-)}$	L_xT_y, L_yT_x
7			$2_z^{(-)}, 2_x^{(+)}$	L_yT_z, L_zT_y
8			$2_z^{(-)}, 2_x^{(-)}$	L_zT_x, L_xT_z
9		25-46	$2_z^{(+)}, \bar{2}_x^{(-)}$	L_xT_x, L_yT_y, L_zT_z
10			$2_z^{(-)}, \bar{2}_x^{(+)}$	L_xT_z, L_zT_x
11			$2_z^{(-)}\ \bar{2}_x^{(-)}$	L_zT_y, L_yT_z
12		47-74	$\bar{1}^{(-)}, 2_z^{(+)}, 2_x^{(+)}$	L_xT_x, L_yT_y, L_zT_z
13			$\bar{1}^{(-)}, 2_z^{(+)}, 2_x^{(-)}$	L_xT_y, L_yT_x
14			$\bar{1}^{(-)}, 2_z^{(-)}, 2_x^{(+)}$	L_yT_z, L_zT_y
15			$\bar{1}^{(-)}, 2_z^{(-)}, 2_x^{(-)}$	T_xL_z, L_xT_z
16	Tetragonal	75-80	$4_z^{(-)}$	$L_xT_x - L_yT_y, L_xT_y + L_yT_x$
17		81-82	$\bar{4}_z^{(-)}$	$L_xT_x + L_yT_y, L_zT_z, L_xT_y - L_yT_x$
18		83-88	$\bar{1}^{(-)}, 4_z^{(+)}$	$L_xT_x + L_yT_y, L_zT_z, L_xT_y - L_yT_x$
19			$\bar{1}^{(-)}, 4_z^{(-)}$	$L_xT_x - L_yT_y, L_xT_y + L_yT_x$
20		89-98	$4_z^{(+)}, 2_x^{(-)}$	$L_xT_y - L_yT_x$
21			$4_z^{(-)}, 2_x^{(+)}$	$L_xT_x - L_yT_y$
22			$4_z^{(-)}, 2_x^{(-)}$	$L_xT_y + L_yT_x$
23		99-110	$4_z^{(+)}\ \bar{2}_x^{(-)}$	$L_xT_x + L_yT_y, L_zT_z$
24			$4_z^{(-)}, \bar{2}_x^{(+)}$	$L_xT_y + L_yT_x$
25			$4_z^{(-)}, \bar{2}_x^{(-)}$	$L_xT_x - L_yT_y$

Table 3. (Continue) - 1

26	Tetragonal	111-114	$\bar{4}_z^{(+)}, 2_x^{(-)}$	$L_x T_y + L_y T_x$
27		121-122	$\bar{4}_z^{(-)}, 2_x^{(+)}$	$L_x T_x + L_y T_y,\ L_z T_z$
28			$\bar{4}_z^{(-)}, 2_x^{(-)}$	$L_x T_y - L_y T_x$
29		115-120	$\bar{4}_z^{(+)} \bar{2}_x^{(-)}$	$L_x T_x - L_y T_y$
30			$\bar{4}_z^{(-)}, \bar{2}_x^{(+)}$	$L_x T_y - L_y T_x$
31			$\bar{4}_z^{(-)}, \bar{2}_x^{(-)}$	$L_x T_x + L_y T_y,\ L_z T_z$
32		123-142	$\bar{1}^{(-)}, 4_z^{(+)}, 2_x^{(+)}$	$L_x T_x + L_y T_y,\ L_z T_z$
33			$\bar{1}^{(+)}, 4_z^{(+)}, 2_x^{(-)}$	$L_x T_y - L_y T_x$
34			$\bar{1}^{(+)}, 4_z^{(-)}, 2_x^{(+)}$	$L_x T_x - L_y T_y$
35			$\bar{1}^{(-)}, 4_z^{(-)}, 2_x^{(-)}$	$L_x T_y + L_y T_x$
36	Trigonal	147-148	$\bar{3}_z^{(-)}$	$L_x T_x + L_y T_y,\ L_x T_y - L_y T_x,\ L_z T_z$
37		149-155	$3_z^{(+)}, 2_x^{(-)}$	$L_x T_y - L_y T_x$
38			$3_z^{(+)}, 2_y^{(-)}$	$L_x T_y - L_y T_x$
39		156-161	$3_z^{(+)}, \bar{2}_x^{(-)}$	$L_x T_x + L_y T_y,\ L_z T_z$
40			$3_z^{(+)}, \bar{2}_y^{(-)}$	$L_x T_x + L_y T_y,\ L_z T_z$
41		162-167	$\bar{1}^{(-)}, 3_z^{(+)}, 2_x^{(+)}$	$L_x T_x + L_y T_y,\ L_z T_z$
42			$\bar{1}^{(-)}, 3_z^{(+)}, 2_x^{(-)}$	$L_x T_y - L_y T_x$
43			$\bar{1}^{(-)}, 3_z^{(+)}, 2_y^{(+)}$	$L_x T_x + L_y T_y,\ L_z T_z$
44			$\bar{1}^{(-)}, 3_z^{(+)}, 2_y^{(-)}$	$L_x T_y - L_y T_x$
45	Hexagonal	168-173	$6_z^{(-)}$	$T_z (L_x \pm i L_y)^3,\ L_z (T_x \pm i T_y)(L_x \pm i L_y)^2$
46		174	$\bar{6}_z^{(-)}$	$T_z L_z,\ L_x T_x + L_y T_y,\ L_y T_x - L_x T_y$
47		175-176	$\bar{1}^{(-)}\ 6_z^{(+)}$	$L_z T_z,\ L_x T_x + L_y T_y,\ L_x T_y - L_y T_x$
48			$\bar{1}^{(-)}\ 6_z^{(-)}$	$T_z (L_x \pm i L_y)^3,\ L_z (T_x \pm i T_y)(L_x \pm i L_y)^2$
49		177-182	$6_z^{(+)}, 2_x^{(-)} (2_y^{(-)})$	$L_x T_y - L_y T_x$
50			$6_z^{(-)}, 2_x^{(-)} (2_y^{(+)})$	$T_z [(L_x + i L_y)^3 + (L_x - i L_y)^3]$ $L_z [(T_x + i T_y)(L_x + i L_y)^2 +$ $+ (T_x - i T_y)(L_x - i L_y)^2]$

Table 3. (Continue) - 2.

51	Hexagonal		$6_z^{(-)}, 2_x^{(+)} (2_y^{(-)})$	$i T_z [(L_x + i L_y)^3 - (L_x - i L_y)^3]$ $i L_z [(T_x + i T_y)(L_x + i L_y)^2 -$ $- (T_x - i T_y)(L_x - i L_y)^2]$
52		183-186	$6_z^{(+)}, 6_x^{(-)} (6_y^{(-)})$	$L_z T_z,\ L_x T_x + L_y T_y$
53			$6_z^{(-)}, 6_x^{(-)} (6_y^{(+)})$	$i T_z [(L_x + i L_y)^3 - (L_x - i L_y)^3]$ $i L_z [(T_x + i T_y)(L_x + i L_y)^2 -$ $- (T_x - i T_y)(L_x - i L_y)^2]$
54			$6_z^{(-)}, 6_x^{(+)} (6_y^{(-)})$	$T_z [(L_x + i L_y)^3 + (L_x - i L_y)^3]$ $L_z [(T_x + i T_y)(L_x + i L_y)^2 +$ $+ (T_x - i T_y)(L_x - i L_y)^2]$
55		187-188	$\bar{6}_z^{(-)}, 6_x^{(+)} (6_y^{(+)})$	$L_x T_y - L_y T_x$
56			$\bar{6}_z^{(-)}, 6_x^{(-)} (6_y^{(-)})$	$L_z T_z,\ L_x \bar{T}_x + L_y T_y$
57			$\bar{6}_z^{(+)} 6_x^{(-)}$	$i T_z [(L_x + i L_y)^3 - (L_x - i L_y)^3]$ $i L_z [(T_x + i T_y)(L_x + i L_y)^2 -$ $- (T_x - i T_y)(L_x - i L_y)^2]$
58			$\bar{6}_z^{(+)} 6_y^{(-)}$	$T_z [(L_x + i L_y)^3 + (L_x - i L_y)^3]$ $L_z [(T_x + i T_y)(L_x + i L_y)^2 +$ $+ (T_x - i T_y)(L_x - i L_y)^2]$
59		189-190	$\bar{6}_z^{(-)} 2_x^{(+)} (2_y^{(+)})$	$L_z T_z,\ L_x T_x + L_y T_y$
60			$\bar{6}_z^{(-)} 2_x^{(-)} (2_y^{(-)})$	$L_x T_y - L_y T_x$
61			$\bar{6}_z^{(+)} 2_x^{(-)}$	$T_z [(L_x + i L_y)^3 + (L_x - i L_y)^3]$ $L_z [(T_x + i T_y)(L_x + i L_y)^2 +$ $+ (T_x - i T_y)(L_x - i L_y)^2]$
62			$\bar{6}_z^{(+)} 2_y^{(-)}$	$i T_z [(L_x + i L_y)^3 - (L_x - i L_y)^3]$ $i L_z [(T_x + i T_y)(L_x + i L_y)^2 -$ $- (T_x - i T_y)(L_x - i L_y)^2]$

Table 3. (Continue - 3).

63	Hexagonal	191-194	$\bar{1}^{(-)}6_z^{(+)}2_x^{(+)}(2_y^{(+)})$	$L_z T_z,\ L_x T_x + L_y T_y$
64			$\bar{1}^{(-)}6_z^{(+)}2_x^{(-)}(2_y^{(-)})$	$L_x T_y - L_y T_x$
65			$\bar{1}^{(-)}6_z^{(-)}2_x^{(+)}(2_y^{(-)})$	$iT_z[(L_x+iL_y)^3-(L_x-iL_y)^3]$ $iL_z[(T_x+iT_y)(L_x+iL_y)^2-$ $-(T_x-iT_y)(L_x-iL_y)^2]$
66			$\bar{1}^{(-)}6_z^{(-)}2_x^{(-)}(2_y^{(+)})$	$T_z[(L_x+iL_y)^3+(L_x-iL_y)^3]$ $L_z[(T_x+iT_y)(L_x+iL_y)^2+$ $+(T_x-iT_y)(L_x-iL_y)^2]$
67	Cubic	200-206	$\bar{1}^{(-)}3_{xyz}^{(+)}2_z^{(+)}$	$L_x T_x + L_y T_y + L_z T_z$
68		207-214	$4_z^{(-)}3_{xyz}^{(+)}$	$T_x L_x(L_y^2-L_z^2)+T_y L_y(L_z^2-L_x^2)+$ $+T_z L_z(L_x^2-L_y^2)$
69		215-220	$\bar{4}_z^{(-)}3_{xyz}^{(+)}$	$T_x L_x(L_y^2+L_z^2)+T_y L_y(L_z^2+L_x^2)+$ $+T_z L_z(L_x^2+L_y^2)$
70		221-230	$\bar{1}^{(-)}3_{xyz}^{(+)}4_z^{(+)}$	$T_x L_x(L_y^2+L_z^2)+T_y L_y(L_z^2+L_x^2)+$ $+T_z L_z(L_x^2+L_y^2)$
71			$\bar{1}^{(-)}3_{xyz}^{(+)}4_z^{(-)}$	$T_x L_x(L_y^2-L_z^2)+T_y L_y(L_z^2-L_x^2)+$ $+T_z L_z(L_x^2-L_y^2)$

sitions not coinciding with the inversion centre (as is known, general positions cover not only those lattice points which neither symmetry element passes through but also those placed on spiral axes and sliding reflection planes.

ORTHORHOMBIC SYSTEM (222, $mm2$, mmm)

a) There is not symmetry centre (222, $mm2$): in these groups two independent symmetry elements are either two axes 2(perpendicular to each other) or two planes m perpendicular to each other. In the case of two axes 2 generating the third one perpendicular to them, the structure should be even with respect to one axis and odd with

respect to the others (NN 16-24). In the case of two planes the AFM structure should be odd with respect to axis 2 and even with respect to planes mm or even with respect to 2 and one of the planes m but odd with respect to the other (NN 25-26). Both in the first and second cases magnetic sites may occupy either general or special positions in the lattice. Thus, if magnetic sites are placed on the rotational axis 2 or on the mirror plane m, TCS is admitted by only one AFM structure for which two other symmetry elements transform the sites with antiparallel magnetic moments into each other. If the magnetic sites are at intersection of rotational axes or mirror planes, the AFM structure does not exist and TCS is impossible;

b) There is a symmetry centre (mmm): for general positions the structures are possible as in the case a) - odd with respect to one axis 2 and even with respect to the others; moreover, the AFM structure is possible that is even with respect to all the axes 2 but necessarily odd with respect to the inversion centre (NN 47-74). For special positions when the sites are placed on the rotational axis or mirror plane only two types of the AFM structure are admissible (even or odd with respect to the other two axes or planes). If the magnetic sites are placed at intersection points of two mirror axes or at the line of intersection of two mirror planes, the AFM structure should be even with respect to all rotational elements.

For crystals of higher symmetry (tetragonal, trigonal, hexagonal and cubic systems) the treatment is more tedious though can be performed by analogy with the previous one. The corresponding results are listed in table 3 in which the first column contains the numbers of space groups, the second one contains possible types of the AFM structure generating TCS and the third one contains concrete types of invariants. The absence in the table of the number of some group means that TCS is impossible in it (at least due to linear and cubic with respect to L invariants). Formally, it is admissible that T can be generated by any higher with respect to L invariants; however, such systems are not considered in this paper. Therefore, hereafter the possibility (or impossibility) of TCS should be treated as the presence (absence) of linear or cubic with respect to L invariants in Table 3.

For the FM structures the invariant combinations of the vectors $\vec{T}$ and $\vec{M}$ are constructed by analogy with AFM taking into account the remarks made above. The results are listed in Table 4.

Table 4. Classification of toroid ferromagnets.

N	System (singony)	Space group	Crystalline class	Invariants
1		1	C_1	$M_x T_x,\ M_y T_y,\ M_z T_z,\ M_x T_y,\ M_x T_z,$ $M_y T_x,\ M_y T_z,\ M_z T_x,\ M_z T_y$
2		3-5	C_2	$M_x T_y,\ M_y T_x,\ M_x T_x,\ M_y T_y,\ M_z T_z$
3		6-9	C_S	$M_z T_x,\ M_z T_y,\ M_x T_z,\ M_y T_z$
4		16-24	$\mathcal{D}_2$	$M_x T_x,\ M_y T_y,\ M_z T_z$
5		25-46	C_{2v}	$M_x T_y,\ M_y T_x$
6		75-80	C_4	$M_x T_y - M_y T_x,\ M_x T_x + M_y T_y,\ M_z T_z$
7		81-82	S_4	$M_x T_x - M_y T_y,\ M_x T_y + M_y T_x$
8		89-98	$\mathcal{D}_4$	$M_x T_x + M_y T_y,\ M_z T_z$
9		99-100	C_{4v}	$M_x T_y - M_y T_x$
10		111-114 121-122	$\mathcal{D}_{2d}$	$M_x T_x - M_y T_y$
11		115-120		$M_x T_y + M_y T_x$
12		143-146	C_3	$M_x T_x + M_y T_y,\ M_x T_y - M_y T_x,\ M_z T_z$
13		149-155	$\mathcal{D}_3$	$M_x T_x + M_y T_y,\ M_z T_z$
14		156-161	C_{3v}	$M_x T_y - M_y T_x$
15		168-173	C_6	$M_x T_x + M_y T_y,\ M_x T_y - M_y T_x,\ M_z T_z$
16		174	C_{3h}	$T_z[(M_x+iM_y)^2+(M_x-iM_y)^2]$ $iT_z[(M_x+iM_y)^3-(M_x-iM_y)^3]$ $M_z[(T_x+iT_y)(M_x+iM_y)^2+$ $+(T_x-iT_y)(M_x-iM_y)^2]$ $iM_z[(T_x+iT_y)(M_x+iM_y)^2-$ $-(T_x-iT_y)(M_x-iM_y)^2]$
17		177-182	$\mathcal{D}_6$	$M_x T_x + M_y T_y,\ M_z T_z$
18		183-186	C_{6v}	$M_x T_y - M_y T_x$

Table 4. (Continue - 4).

19		187-188	$\mathcal{D}_{3h}$	$T_z[(M_x+iM_y)^3+(M_x-iM_y)^3]$ $M_z[(T_x+iT_y)(M_x+iM_y)^2+$ $+(T_x-iT_y)(M_x-iM_y)^2]$
20		189-190		$iT_z[(M_x+iM_y)^3-(M_x-iM_y)^3]$ $iM_z[(T_x+iT_y)(M_x+iM_y)^2-$ $-(T_x-iT_y)(M_x-iM_y)^2]$
21		195-199	T	$M_x T_x + M_y T_y + M_z T_z$
22		207-214	O	$T_x M_x (M_y^2+M_z^2)+T_y M_y (M_z^2+M_x^2)$ $+T_z M_z (M_x^2+M_y^2)$
23		215-220	T_d	$M_x T_x (M_y^2-M_z^2)+M_y T_y (M_z^2-M_x^2)+$ $+M_z T_z (M_x^2-M_y^2)$

5. CONCRETE EXAMPLES OF MAGNETS WITH TCS

A. ANTIFERROMAGNETS

Before applying the approach developed in the previous paragraph for the analysis of concrete AFM combinations, we should like to make the following remark. The consideration of a pseudoproper TCS regardless of a concrete AFM structure generating it may turn out to be aimless, and that is the reason why.

In listing in the corresponding column of Table 3 the types of the AFM structure generating TCS for each crystalline system, we implicitly assume that the magnetic atoms occupy general places in the lattice. The consideration of real magnets[16] indicates that in most cases the magnetic atoms are on some symmetry elements (rotational axes or symmetry planes), i.e. occupy special positions. Transition from the general type to the special one may lead to violation of sufficient conditions for the existence of TCS.

We start consideration with the space group $D_h^{16}(P_{nma})$, that for methodical purposes will be made thoroughly.

Placing the magnetic atom into the general position 8d(x, y, z)[17] and using the available 8 symmetry elements (in notation of

point groups: the elements E, $2x$, $2y$, $2z$, $\bar{1}$, σ_x, σ_y, σ_z), we get 8 atoms inside the elementary cell with coordinates

$$1(x,y,z),\ 2\left(\tfrac{1}{2}+x,\tfrac{1}{2}-y,\tfrac{1}{2}-z\right),\ 3\left(\bar{x},\tfrac{1}{2}+y,\bar{z}\right),\ 4\left(\tfrac{1}{2}-x,\bar{y},\tfrac{1}{2}+z\right),\quad (13)$$
$$5(\bar{x},\bar{y},\bar{z}),\ 6\left(\tfrac{1}{2}-x,\tfrac{1}{2}+y,\tfrac{1}{2}+z\right),\ 7\left(x,\tfrac{1}{2}+\bar{y},z\right),\ 8\left(\tfrac{1}{2}+x,y,\tfrac{1}{2}-z\right).$$

Let us denote the relevant spin local moments by S_i ($i = 1,\dots, 8$). Due to the presence of the inversion transformation connecting the pairs of sites $1 \leftrightarrow 5, 2 \leftrightarrow 6$, $3 \leftrightarrow 7$ and $4 \leftrightarrow 8$ and to the condition of odd AFM structure with respect to the inversion centre, it follows that $S_1 = -S_5, S_2 = S_6, S_3 = -S_7$ and $S_4 = S_8$. Taking into account atom interchanging, realized by the rest symmetry elements, we get 4 types of collinear antiferromagnetic structures admitting TCS:

$$\begin{aligned} &S_1 \| S_2 \| S_3 \| S_4 && \left(\bar{1}^{(-)}\ 2_z^{(+)}\ 2_x^{(+)}\right),\\ &S_1 \| S_4 \| S_6 \| S_7 && \left(\bar{1}^{(-)}\ 2_z^{(+)}\ 2_x^{(-)}\right),\\ &S_1 \| S_2 \| S_7 \| S_8 && \left(\bar{1}^{(-)}\ 2_z^{(-)}\ 2_x^{(+)}\right),\\ &S_1 \| S_3 \| S_6 \| S_8 && \left(\bar{1}^{(-)}\ 2_z^{(-)}\ 2_x^{(-)}\right). \end{aligned} \quad (14)$$

There are also three types of special positions of magnetic atoms each having multiplicity 4. The first two types (4a, 4b)[17] coincide with the position of the symmetry centre: respectively, with the coordinates (0,0,0) for position 4a and (0,0,1/2) for 4b. The third position 4c with coordinates (x, 1/4, z) is in the plane σ_y. When passing from the general 8d position to special 4a and 4b, the AFM structure generating TCS disappears. Such a situation occurs, for instance, in yttrium orthoferrites with the structure $YMeO_3$ (Me is the 3d metal) where the magnetic atoms Me occupy 4b positions.

When passing from 8d to 4c the points connected by the plane σ_y coincide, i.e. $1 \leftrightarrow 7$, $2 \leftrightarrow 8$, $3 \leftrightarrow 5$ and $4 \leftrightarrow 6$ so that instead of eight sites in the elementary lattice we have four: I (instead of 1,7), II (instead of (3.5), III (instead of 2,8) and IV (instead of 4,6). In particular, this is valid for the compound α-FeOOH in which ions of Fe occupy 4c positions. The AFM structure generated is of the type

$$\left(S_{\mathrm{I}} \| S_{\mathrm{II}}\right) = -\left(S_{\mathrm{III}} \| S_{\mathrm{IV}}\right) \quad \left(\bar{1}^{(-)}\ 2_z^{(-)}\ 2_x^{(+)}\right) \quad (15)$$

and admits TCS with the invariant $L_y T_z$ (see Table 3). It is interesting to note that three compounds in this group $LiCoPO_4$, $LiMnPO_4$ and $LiNiPO_4$, in which the corresponding magnetic atoms Co, Mn and Ni also occupy 4c positions, are characterised by the structure $\bar{1}^{(-)} 2_z^{(-)} 2_x^{(+)}$. In this case the vectors of antiferromagnetism in them are directed along the axis of y (Co), x (Mn) and y (Ni) and the relevant invariants are different. A similar situation occurs in Co_2SiO_4 and $KFeCl_3$ where the magnetic ions of Co or Fe also occupy 4c positions; however, the AFM structure is even with respect to the inversion center and TCS is impossible.

Now we undertake a systematic consideration of magnets belonging to all the systems starting from the monoclinic one (we are not aware of any real antiferromagnetic belonging to the triclinic system though for the space group No. 2 the existence of TCS is not forbidden by the symmetry conditions).

The group C_{2h}^2 ($P2_1/m$): such a type is represented by the compound ErOOH where ions of Er, the magnetic moments of which are directed along the axis y, occupy special two-dimensional 2 positions. According to the general classification of AFM, the structure is of the type

$$S_1(S_4) = S_2(S_3) \quad \left(\bar{1}^{(-)}\ 2_y^{(-)}\right) \quad (16)$$

and admit, according to Table 3, TCS corresponding to invariants $L_y T_x$ and $L_y T_z$. In the isomorphic compound DyOOH the magnetic ions of Dy occupy 2e positions as well but are placed in the plane x, the invariants having the form $L_x T_y$ and $L_z T_y$.

In the orthorhombic system apart from the group D_{2h}^{16} considered above we are interested in D_{2h}^{14} (P_{bna})-compound $CrVO_4$ and $D_{2h}^{14}(C_{mcm})$ -compound γ-FeOOH. For $CrVO_4$ magneto-active ions of Cr and V occupy special positions 4c. The spin AFM structure is of the type $\bar{1}^{(-)} 2_z^{(-)} 2_x^{(+)}$ the moments having only a component along the axis y (L_y) with

$$\left(S_1 \| S_4\right) = -\left(S_2 \| S_3\right) \quad (17)$$

and TCS is characterised by the invariant $L_y T_z$.

For γ-FeOOH ions of Fe occupy 4c positions. The AFM structure is of the type $\bar{1}^{(-)} 2_z^{(-)} 2_x^{(-)}$ $\left((S_1 \| S_3) = -(S_2 \| S_4)\right)$ and TCS is specified by the invariant $L_x T_z$ (since spins are directed along the axis x). Note that in the compound from the same group D_{2h}^{17} – Cr_2VO_4, ions of Cr occupy 4a positions, the AFM structure being even with respect to the inversion centre and TCS is impossible.

Consider now the tetragonal system.

The group $D_{4h}^{14}(P4_2/mnm)$ is represented by Cr_2WO_6. Ions of

Cr occupy 4e positions, the magnetic moments lie in the basis plane xy, the structure is

$$(S_1 \| S_2) = -(S_3 \| S_4) \qquad (4_z^{(-)} \sigma_x^{(-)}) \tag{18}$$

and TCS is characterised by the invariant $L_x T_x - L_y T_y$.

The group $C_{4v}^{10}(I4_1cm)$ is represented by $KCrF_3$. Ions of Cr occupy 4e positions, the magnetic moments lie in the plane xy the structure is of the type

$$(S_1 \| S_2) = -(S_3 \| S_4) \quad (4_z^{(-)} \sigma_x^{(-)}) \tag{19}$$

and TCS is specified by an invariant of the type $L_x T_x - L_y T_y$. In the compound Fe_2TeO_3 from the same group of symmetry, the AFM structure is of the type $\bar{1}^{(-)} 4_z^{(+)} 2_x^{(-)}$ TCS is impossible in this case since the magnetic moments of Fe ions are directed along the axis z and necessary invariants are absent. For the same reason TCS is absent in the compound $CrCl_2$ (group $D_{4h}^{17}(I4_1/mmm)$ though the AFM structure is of the type $\bar{1}^{(-)} 4_z^{(+)} 2_x^{(-)}$.

In the hexagonal system we shall consider the group $D_{3d}^6(R\bar{3}c)$ represented by V_2O_3. Ions of V occupy 4c positions (on the third order axis). The magnetic moments in this compound cannot be thought well localized; however, it follows from the neutron-diffraction measurements[16] that maximum of the spin density is placed at the sites V and the spin vectors at the sites are oriented along the third order axis. The vector of antiferromagnetism for the structure V_2O_3 corresponds to the type[18]

$$L = S_1 - S_2 - S_3 + S_4 \quad (\bar{1}^{(-)} 3_z^{(+)} 2_x^{(+)}). \tag{20}$$

The only invariant admitting TCS has the form ($L_z T_z$), i.e. for V_2O_3 the symmetry of the vectors L and T coincides (this is solely due to the character of the spin density distribution).

Of particular interest are crystals of the cubic system. In a variety of cubic AFM we could not find any for which sufficient conditions for the existence of TCS could be fulfilled. At the same time, as it follows from table 3, the appearance of TCS in the cubic AFM are not in principle prohibited.

B. FERROMAGNETS

Since there are no real FM belonging to triclinic and monoclinic systems[16], we immediately pass to the orthorhombic system.

The group $C_{2v}^{20}(Imm2)$ is represented by the ferromagnet $NaNiFeF_7$. The vector M is derected along the axis x; therefore, the corresponding invariant has the form $M_x T_y$.

The tetragonal system: the group $C_{4v}^{10}(I4cm)$, the ferromagnet Cr_2FeMoO_6. The direction of the vector $\vec{M}$ is not indicated in ref.[16]. If $\vec{M}$ lies on the principal axis, the linear with respect to $\vec{T}$ invariants are absent.

The hexagonal system: the group $D_{2d}^{12}(I\bar{4}2d)$, the ferfimagnet $CuCr_2O_4$. The vector $\vec{M}$ lies in the basis plane and an invariant generating TCS has the form $M_x T_x - M_y T_y$. One more representative in this system is the ferromagnetic $MnNb_2S_6$ group $D_6^6(P6_322)$ and the invariant $M_x T_x + M_y T_y$.

The cubic system: group $T^4(P2_13)$, is represented by $MnSi$ the magnetic structure of which admits only the invariant $\vec{T}\vec{M}$ ($\vec{M}$ is directed along one of the coordinate axes). The data of neutron difraction indicate a weak helicity of FM structure (the helicity period is $\sim 120^\circ$A), that is due to the relativistic interactions. Probably, this is an indirect indication of TCS since the presence of the invariants $\vec{T}\cdot\vec{M}$ and $\vec{T}\cdot \mathrm{rot}\,\vec{M}$ indicates a weak inhomogeneity of the FM structure.

The second representative of this system is the ferromagnet $Li_{0,5}Fe_{2,5-x}Al_xO_4$ group $O^7(P4_132)$, invariant $T_x M_x(M_y^2 + M_z^2) + T_y M_y(M_x^2 + M_z^2)$ The exact data on the direction of $\vec{M}$ are not available. The third representative is the ferromagnetic $NiMnSb$, group $T_d^2(F\bar{4}3m)$, invariant $T_x M_x(M_y^2 - M_z^2) + T_y M_y(M_z^2 - M_x^2) + T_z M_z(M_x^2 - M_y^2)$ and the vector $\vec{M}$ has the general direction.

6. SOME MICROSCOPIC MODELS OF PSEUDOPROPER TCS

The microscopic model of proper TCS in the systems with electron hole pairing has thoroughly been investigated in ref.[12]. Under certain requirements on the symmetry of electron wave function, the orbital toroid ordering may appear already in the exchange approximation, i.e. the relativistic smallness is absent. For a pseudoproper TCS, one should take into consideration the spin-orbital interaction.

Let us consider two simplest models of a pseudoproper TCS in ferro and antiferromagnets, respectively, allowing one to trace the origin of this unusual orbital ordering.

A. FERROMAGNETIC SEMICONDUCTOR

Assume that the crystalline potential $V(\vec{r})$ of the system can be expressed through the sum $V(\vec{r}) = V_s(\vec{r}) + V_a(\vec{r})$ where V_s is the even and V_a is the odd potential parts with respect to spatial inversion; for simplicity we shall assume $V_a \ll V_s$. In what follows we shall consider V_a as perturbation. Let at $V_a \equiv 0$ the wave functions of extrem of the conduction and valence bands transform according to even and odd one-dimensional representations of the potential V_s at the point k_0 of the Brillouin zone. Then, in the two-band κP model, that is the aim of our consideration, the model Hamiltonian of a ferromagnetic semiconductor is

$$\hat{H} = \hat{H}_0 + \hat{H}_{ex} + \hat{H}_{so}, \tag{21}$$

$$\hat{H}_0 = \begin{pmatrix} \varepsilon_1(\hat{\vec{k}}) & i\vec{v}\hat{\vec{k}} + \eta \\ -i\vec{v}\hat{\vec{k}} + \eta & \varepsilon_2(\hat{\vec{k}}) \end{pmatrix}, \tag{22}$$

$$\hat{H}_{ex} = -\begin{pmatrix} \gamma_1 & 0 \\ 0 & \gamma_2 \end{pmatrix}(\hat{\vec{S}}\hat{\vec{\sigma}}), \tag{23}$$

$$\hat{H}_{so} = \begin{pmatrix} \vec{\varphi}_1 \times \hat{\vec{\sigma}} & \vec{\lambda} \times \hat{\vec{\sigma}} \\ \vec{\lambda} \times \hat{\vec{\sigma}} & \vec{\varphi}_2 \times \hat{\vec{\sigma}} \end{pmatrix} \cdot \hat{\vec{k}}. \tag{24}$$

In the exchange part of the Hamiltonian (21) only the intraband terms H_{ex} of the interaction of itinerant electrons and local spins are retained for simplicity; $i\vec{v}$ is the interband velocity matrix element, $\eta = \langle 1|V_a|2\rangle$ is the matrix element of the potential V_a at the extremum point, $\vec{\lambda} = \frac{1}{4m^2c^2}\langle 1|\vec{\nabla}V_s|2\rangle$ and $\vec{\varphi}_{1,2} = \frac{1}{4m^2c^2}\langle 1,2|\vec{\nabla}V_a|1,2\rangle$ are the inter- and intraband matrix elements of the spin-orbital interaction. For simplicity we shall neglect in further calculations the contribution $\sim \vec{\varphi}_{1,2}$ in view of the assumed smallness of $V_a \ll V_s$ though at $V_a \sim V_s$ it should in fact be retained. Then, assume for simplicity $\varepsilon_1(\vec{k}) = -\varepsilon_2(\vec{k}) = E_g/2 + \frac{k^2}{2m^*} \equiv \varepsilon(\vec{k})$ where m^* is the effective mass of carriers, E_g is the width of the semiconductor band gap, and $|\gamma_{1,2}S| \ll E_g$ so that we can restrict our consideration to the mean field approximation with respect to the s-d exchange interaction.

The electron spectrum in the model (21) in the above assumptions is easily calculated and is of a "skew" nature

$$E_{\pm}^{\sigma_z}(\vec{k}) = -\frac{\gamma_1 + \gamma_2}{2}\langle\vec{S}\rangle\sigma_z \pm \left[\left(\varepsilon(\vec{k}) - \frac{\gamma_1 - \gamma_2}{2}\langle S\rangle\sigma_z\right)^2 + \right. \tag{25}$$

$$\left. + \left(\eta - (\vec{\lambda}\times\hat{\vec{k}})_z\sigma_z\right)^2 + |\vec{v}\vec{k}|^2 + (\vec{\lambda}\times\vec{k})_\perp^2\right]^{1/2}.$$

Here the axis z is directed along $\langle\vec{S}\rangle$.

Let us consider this expression in more detail. At $\eta = 0$, i.e. in the ferromagnet with the inversion center, we have $E^{\sigma}(\vec{k}) = E^{\sigma}(-\vec{k})$ but $E^{\sigma}(\vec{k}) \neq E^{-\sigma}(\vec{k})$. If $\eta \neq 0$ then $E^{\sigma}(\vec{k}) \neq E^{\sigma}(-\vec{k})$ and the spectrum is "skew" (it is natural that $E^{\sigma}(\vec{k}) \neq E^{-\sigma}(-\vec{k})$). The symmetry loss $\vec{k} \to -\vec{k}$ of the electronic spectrum of magnet without the inversion centre is not unexpected by itself; the thoroics posses just this asymmetry [13].

We now calculate the orbital toroid moment of a ferromagnet with the Hamiltonian (21). For this purpose we proceed as follows. By introducing the orbital current operator

$$\hat{\vec{j}}_L = e\frac{\delta\hat{H}}{\delta\hat{\vec{k}}}, \tag{26}$$

where $\hat{\vec{k}}$ is the quasimomentum operator, we calculate the Fourier component $\langle\hat{j}_q\rangle = j_q$ for small short-wave fluctuations of magnitization $\langle\vec{j}\rangle$ $(\vec{q}\to 0)$. Then, using the known relation of the orbital current and toroid moment $\vec{j}_L = c\,\mathrm{rot}\,\mathrm{rot}\,\vec{T}$, as a rezult of cumbersome calculations we get

$$\vec{T}_L = \frac{m}{m^*}\mu_B\langle\vec{S}\rangle\times\vec{\lambda}(\gamma_1 - \gamma_2)\eta\bar{N}\frac{3\pi}{40E_g^2}, \tag{27}$$

$$\bar{N} = \frac{(m^*)^{3/2}E_g^{1/2}}{2\pi^2}.$$

All the calculations have been performed at the temperature $\theta \ll E_g$ and under the assumption $\{|\vec{v}\vec{k}|, |\gamma S|, \eta\} \ll E_g$ in the first order in the spin-orbital interaction λ.

Thus, in our model at $\gamma_1 \neq \gamma_2$ the toroid moment of a ferromagnetic semiconductor without the inversion centre dirrers from zero. However, the asymmetry property of spectrum (25) with restpect to the time inversion ($\vec{k} \to -\vec{k}$) is conserved also at $\gamma_1 = \gamma_2$.

Note that in the model considered another physical quantity analogous in symmetry to T_L is different from zero: the spin (or "inducted" by the terminology of ref.[2]) toroid moment T_S characteristing the redistribution of the spin density of itinerant electrons under ferromagnetic ordering of local spins. The calculation of

$\vec{T}_S$ performed on the basis of the expression for magnetization $\vec{m} = rot\,\vec{T}_S$ of itinerant electrons gives:

$$\vec{T}_S = \frac{\pi}{4E_g^2}\,\vec{v}\times\langle\vec{S}\rangle\mu_B N \eta\,(\gamma_1-\gamma_2). \qquad (28)$$

However, we should like to note that all specific features of TCS making it into a special type of an orbital magnetic ordering are associated just with $\vec{T}_L$. In particular, as is seen from (25), at $\vec{\lambda} = 0$ when $\vec{T}_L = 0$ but $\vec{T}_S \neq 0$ the electron spectrum becomes "nonskew".

B. ANTIFERROMAGNETIC SEMICONDUCTOR

Now we consider the model Hamiltonian analogous to (21) but with another term of $s-d$ exchange interaction and at $\eta \equiv 0$, $\gamma = \gamma_2 = 0$, $\varphi_{1,2} = 0$:

$$H'_{ex} = -\begin{pmatrix} 0 & \gamma_{12} \\ \gamma_{12} & 0 \end{pmatrix}\left(\hat{\vec{S}}_a - \hat{\vec{S}}_b\right)\vec{G}, \qquad (29)$$

where $\hat{\vec{S}}_a$ and $\hat{\vec{S}}_b$ are the local spins at the sites of the sublattices a and b that are symmetrically placed with respect to the inversion centre, so that $\langle 1|\gamma^a_{12}(\vec{r})|2\rangle = -\langle 1|\gamma^b_{12}(\vec{r})|2\rangle = \gamma_{12}$ due to a different parity of representations $\langle 1|$, $\langle 2|$ at extremum. Assume that a ferromagnetic order is established in the system and $\langle\vec{S}_a\rangle = -\langle\vec{S}_b\rangle$. The electronic spectrum of an antiferromagnetic semiconductor is of the form

$$E^{\sigma_z}_{\pm}(\vec{k}) = \pm\left[\varepsilon^2(\vec{k}) + \left((\vec{\lambda}\times\vec{k})_z + \gamma_{12}L\right)^2 + |\vec{v}\vec{k}|^2 + (\vec{\lambda}\times\vec{k})^2_{\perp}\right]^{1/2}, \qquad (30)$$

$\vec{L} = \vec{S}_a - \vec{S}_b$ (the axis z is directed along $\vec{L}$) and is degenerated with respect to spin projections σ_z; it has the property $E(\vec{k}) \neq E(-\vec{k})$. Calculating the toroid moments $\vec{T}_L$ and $\vec{T}_S$ in the same manner as in the pervious section A, we get

$$\vec{T}_L = \left(\frac{m}{m^*}\right)\mu_B N\,(\vec{\lambda}\times\vec{L})\,\gamma_{12}\left(-\frac{\pi}{6E_g}\right),$$
$$\vec{T}_S = \mu_B N\,(\vec{v}\times\vec{L})\,\gamma_{12}\left(-\frac{3\pi}{2E_g}\right). \qquad (31)$$

It is seen that at $\vec{\lambda} = 0$ or $\vec{L} = 0$ simultaneously $\vec{T}_L = 0$ and the spectrum as asymmetry $E^{\sigma_z}_{\pm}(\vec{k})$ disappears. It is obvious that for $\vec{\lambda} = 0$ antiferromagnets with an odd magnetic structure with respect

to inversion, the parity proportion of the electronic spectrum $E(\vec{k})$ are unambiguously associated with the presence or absence of the toroid moment $\vec{T}$.

7. CONCLUSION

The above treatement of pseudoproper TCS in ferro and antiferromagnet allows us to make the following conclusions:

1. There exists a large class of concrete compounds of different types of symmetries (mainly low and middle) in which the toroid ordering is induced by the spin FM or AFM ordering depending on a type of the magnetic structure.

2. Even under favourable crystalline symmetry essential limitations of the appearance of TCS are imposed by the type of magnetic atoms, orientation of their magnetic moments and location in an elementary cell.

3. In this paper we have thoroughly studied only the simplest types of invariants mixing the spin FM or AFM and orbital TCS structures. The inclusion of higher order invariants in L and M as well as of the contributions associated with striction interactions may lead to a larger number of crystals admitting TCS. In this case the system, in which TCS appeared, will obviously belong to one of 31 classes magnetic according to Table 1.

4. We have considered the collinear AFM structures and homogeneous TCS generated by them. Transition to more complex (noncollinear, long-period, etc.) structures may cause more complex types of TCS. For instance, in rare-earth orthoferrites, where cells contain two kinds of magnetic ions (4 f and 3 d metals), there may appear a noncollinear AFM structure in the rare-earth-sublattice as a result of competition of intra- and inter-sublattice exchange and relativistic interactions. Since the 4f ions occupy favourable for TCS 4c positions (it should be noted that ions of 3d metals are in unfavourable 4-positions), there appears an inhomogeneous structure described by the highest toroid multipoles.

5. In the case of long-periodic FM and AFM structures the requirements on the their parity change: there appears a possibility for generating incommensurable TCS due to an invariant type $\vec{T}\,rot\,\vec{L}$ or $\vec{T}\,rot\,\vec{M}$. (here $\vec{L}$ may be even with respect to the inversion centre). Thus, the class of systems, where the search for TCS is expedient, enlarges.

6. In this paper we did not touch upon the AFM structures with multiplication of periods of an elementary cell. However, one can easily realize that the relevant TCS structures, if any, should belong to the type of "antitoroids" and can be described only by using higher toroid multipoles.

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Тороидные токовые структуры в ферро и антиферромагнетиках

Проведен симметрийный анализ ферро- и антиферромагнетиков с целью выявления в них структур, допускающих тороидное упорядочение орбитальных токов. Построена теория псевдособственного тороидного токового состояния в кристаллических магнетиках, проанализированы некоторые его микроскопические модели. Предсказано существование ТТС в целом ряде конкретных веществ.

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Toroid Current Structures in Ferro and Antiferromagnets

A symmetry analysis of ferro and antiferromagnets is made in order to establish in them structures admitting a toroid ordering of orbital currents. A theory of pseudoproper toroid current state (TCS) in crystalline magnets is presented and some microscopic models are analysed. The existence of TCS in a number of concrete materials is predicted.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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