Pseudoproper toroidal ordering in antiferromagnets

S. S. Krotov and V. V. Tugushev

P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow

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A classification is given for spin antiferromagnetic structures which induce a pseudoproper toroidal current state (TCS) in crystals in proportion to the relativistic interactions. Specific antiferromagnets in which a pseudoproper TCS should arise are listed, and a number of unusual mechanisms are considered which cause rearrangement of the magnetic structure and alter the antiferromagnetic resonance frequencies in the system.

1. INTRODUCTION

From the standpoint of a group-theoretical classification, antiferromagnets (AFM) are systems having Shubnikov white and black-white spatial symmetry with zero average moment (we will not be considering long-period structures).[1] The phase transition to the AFM state is described by a suitable order parameter—the antiferromagnetism vector. However, unlike ferromagnets (both spin and orbital), for which the choice of the magnetization vector **M** (with its characteristic space-time symmetry) as the order parameter is quite natural, for AFM there is no universal way of choosing the order parameters.

In the extreme case of AFM with localized spins a constructive approach is to break the system up into magnetic sublattices, whereupon the order parameter is taken to be the vector **L**, which is a linear combination of the sublattice magnetizations and consequently has an additional symmetry associated with the permutation of the atoms of the various sublattices. If there are many sublattices, however, this approach is difficult, and in the limiting case of systems with delocalized spins it is altogether impossible.

A convenient mathematical apparatus for describing spin magnets without introducing the concept of magnetic sublattices was proposed by Dzyaloshinskiĭ[2] and Andreev and Marchenko[3] and was later developed by Bar'yakhtar and Yablonskiĭ.[4] The basic idea of these papers is to classify magnets according to exchange symmetry groups (the crystal space group augmented by three-dimensional rotations and reflections in spin space). The AFM order parameters in this approach[2–4] are the coefficients of the expansion of the spin density function $\mathbf{S}(\mathbf{r})$ in irreducible representations of the space group of the crystal symmetry.

In orbital AFM the initial microscopic quantity for describing magnetic ordering is the electron current density $\mathbf{j}(\mathbf{r})$. In particular, the symmetry properties of the function $\mathbf{f}(\mathbf{r})$ determine the magnetic space group to which the magnet belongs.[1] Since $\mathbf{j}(\mathbf{r})$ is transverse, it is very convenient to represent it in the Neumann-Debye form[5]

$$\mathbf{j}(\mathbf{r}) = c \operatorname{rot} \mathbf{f}(\mathbf{r}), \tag{1}$$

$$\mathbf{f}(\mathbf{r}) = \mathbf{N}(\mathbf{r}) + \operatorname{rot} \mathbf{R}(\mathbf{r}), \tag{2}$$

$$\mathbf{N}(\mathbf{r}) = \mathbf{r}\psi(\mathbf{r}), \quad \mathbf{R}(\mathbf{r}) = \mathbf{r}\chi(\mathbf{r}), \tag{3}$$

where c is the speed of light, $\psi(\mathbf{r})$ is a pseudoscalar function, and $\chi(\mathbf{r})$ is a scalar function; "rot" denotes the curl. Neither of the functions $\mathbf{N}(\mathbf{r})$ and $\mathbf{R}(\mathbf{r})$ is reducible to the other, since they describe current configurations that are topologically different. The representation (1)–(3) permits a complete multipole parametrization of the current $\mathbf{j}(\mathbf{r})$ in classical electrodynamics, with $\mathbf{N}(\mathbf{r})$ generating the magnetic family of multipole moments and $\mathbf{R}(\mathbf{r})$ a toroidal family.[5,6] The generators in these families are the magnetic dipole moment **M** and the toroidal dipole moment **T**, respectively:

$$\begin{gathered} \mathbf{M} = \frac{1}{2c}\langle[\mathbf{r},\mathbf{j}]\rangle = \int \mathbf{N}(\mathbf{r})\,d^3r, \\ \mathbf{T} = \frac{1}{10c}\langle \mathbf{r}(\mathbf{r}\mathbf{j}) - 2r^2\mathbf{j}\rangle = \int \mathbf{R}(\mathbf{r})\,d^3r \end{gathered} \tag{4}$$

(in the notation used in these equations, $\mathbf{AB} = \mathbf{A}\cdot\mathbf{B}$ and $[\mathbf{A},\mathbf{B}] = \mathbf{A}\times\mathbf{B}$).

In this approach the preferred ordered states of orbital magnets are obviously a ferromagnetic and a toroidal state,1) which are described by the vectors **M** and **T**, respectively (in what follows the toroidal dipole moment **T** will simply be called the toroidal moment). The characteristic scale for the averaging of ρ_0 in (4) depends on the structure of the electron currents and can be of the order of the unit cell dimension or it can be much larger. For example, in the model of Ref. 7 the dimension of ρ_0 is of the order of the coherence length of an electron-hole pair.

From the standpoint of magnetic symmetry, toroidal systems are AFM (the antiferromagnetism vector **T** of which is polar, changes sign under time reversal, and is characterized by the limiting magnetic symmetry group $\infty/m'mm$, i.e., it transforms like the velocity vector **v**). Of the 122 magnetic symmetry classes, 31 admit the existence of a vector **M** and 31 admit the existence of **T** (Ref. 8). From these numbers alone it is clear that toroidal systems are not exotic objects.

It should be stressed that the introduction of a polar vector, which is odd under time reversal, for describing antiferromagnetic structures in crystals is not in itself anything new (in particular, the spin order parameters which arise in the schemes of Refs. 2–4 transform like **T**, and the antisymmetric component of the magnetoelectric tensor is the dual of **T**).[9,10] However, the specific dynamical properties of the toroidal current state (TCS) and various microscopic characteristics of the formation of the toroidal order parameter

TABLE I. Magnetic classes which admits a toroidal current state

Type of ordering		Magnetic class
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	$\bar{P}$	$42'2',\ 32',\ 62'2'$
L, M, weak FM	P	$1, 2, m, 2', m', mm'2'$
L, M, weak FM	$\bar{P}$	$22'2'$

put toroidal systems in a special class of magnets. It is thus necessary to make a detailed analysis of the particular AFM structures that admit a toroidal current state for the purpose of finding possible objects of experimental study. It is this problem which we address in the present paper.

According to the general theory of second-order phase transitions, a toroidal moment T can arise all by itself as a result of spontaneous symmetry breaking (proper toroidal ordering). A microscopic model of such a transition was proposed in Ref. 7. At the same time, it makes sense to consider situations in which a TCS arises as a result of some other kind of ordering.

It is possible to have pseudoproper TCS (e.g., due to the presence of bilinear invariants of the form $\mathbf{T}\cdot\mathbf{L}$ or $\mathbf{T}\cdot\mathbf{M}$ in the thermodynamic potential of an AFM or FM, respectively) and improper TCS [e.g., due to invariants of the form $\mathbf{T}\cdot(\mathbf{P}\times\mathbf{M})$ in ferroelectric magnets, where $\mathbf{P}$ is the polarization vector]. In this paper we consider the formation of a TCS against the background of the spin antiferromagnetic ordering as a consequence of invariants of the type $\mathbf{T}\cdot\mathbf{L}$. In this case the TCS is not accompanied by a change in the symmetry of the AFM structure, so it seems reasonable to use the term "pseudoproper" for this state.

The conditions under which a proper TCS can arise in orbital AFM are rather strict and apparently are seldom satisfied.[7] On the other hand, a pseudoproper TCS should inevitably arise in proportion to the relativistic interactions in crystals with the ordinary spin mechanism of ferromagnetism or antiferromagnetism if the symmetry of the system admits the coexistence of $\mathbf{T}$ with $\mathbf{M}$ or $\mathbf{L}$, respectively.

The possible types of coexistence of the vectors $\mathbf{T}$, $\mathbf{M}$, $\mathbf{P}$, and $\bar{\mathbf{P}}$ ($\bar{\mathbf{P}}$ is the antiferroelectricity vector) can be classified on the basis of the results of Ref. 8, which are tabulated in Table I. We see that 18 magnetic classes admit the coexistence of $\mathbf{L}$ and $\mathbf{T}$ but do not admit weak ferromagnetism, 13 classes admit the coexistence of $\mathbf{M}$ and $\mathbf{T}$, and 7 classes admit a TCS and/or weak ferromagnetism for certain orientations of $\mathbf{L}$. Neither the onset of weak ferromagnetism nor the appearance of a pseudoproper TCS changes the magnetic symmetry of the system.

Toroidal systems are interesting objects of study because the orbital character of the TCS gives rise to a number of features (anomalies of the magnetic susceptibility[11] and of the optical,[12] magnetooptical,[13] and magnetoelectric[9,10] properties). According to Ref. 14, a proper TCS possibly arises in Ni–I boracite. It is shown below that a pseudoproper TCS can occur in many real AFM of various structure types.

Some of the results of this study were reported briefly in Ref. 15.

2. COEXISTENCE CONDITIONS FOR VECTORS T and L

Let us consider an extremely simple model which illustrates clearly the mechanism which gives rise to at toroidal moment as a result of the establishment of AFM spin ordering. Suppose we have a two-sublattice spin magnet whose magnetic sublattices are related by the inversion transformation ($\bar{1}$), so that the AFM structure which arises is odd with respect to $\bar{1}$. We shall assume that in the absence of spin order the orbital microscopic "current loops" formed by itinerant electrons are randomly oriented and cancel one another, so that there are no orbital magnetic or toroidal moments on the scale of the unit cell. The establishment of spin AFM ordering ($\mathbf{M}_1 = -\mathbf{M}_2$, where $\mathbf{M}_{1,2}$ are the spin magnetizations of the sublattices) leads to imbalance (stratification) and deformation of the current loops in proportion to the spin-orbit or magnetic dipole interaction. Let us illustrate this with a pair of identical current loops oriented in the magnetic-ordering plane σ passing through magnetic atoms 1 and 2 (the axes of the current loops lie in the plane σ, and the currents are oppositely directed; see Fig. 1a). The magnetic field $\mathbf{h}$ arising from ordering pulls the loops in the directions of the magnetic atoms 1 and 2, depending on the directions of the currents (see Fig. 1b). The toroidal moment of the pair of displaced current loops is given in order of magnitude by [see (4)]

$$t \backsim xjl/c, \tag{5}$$

where x is the displacement of the loops, l is the characteristic dimension of a loop, and j is the current density in a loop. The value of x is determined from the condition for equilibrium of the system of current loops in the field of the $\mathbf{j}\times\mathbf{B}$ force; in order of magnitude

$$kx \backsim jl\bar{h}/c, \tag{6}$$

where $\bar{h}$ is the average field on the scale of the current loop, and k is the stiffness of the pair of loops that governs their mutual cancellation in the absence of field. Since $\bar{h} \propto L$, by

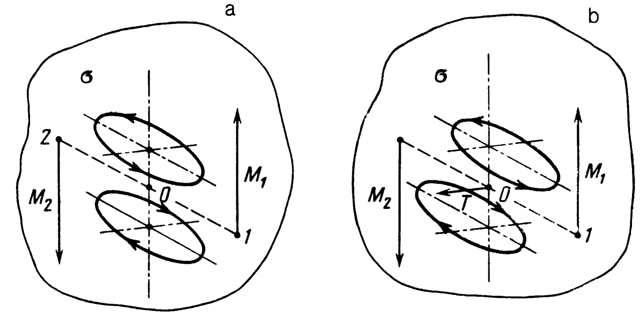

FIG. 1.

substituting x from (6) into (5) we get

$$t \infty (v/c)^2 \frac{e^2}{k} n^2 l^2 L, \qquad (7)$$

where e is the charge, v is the average velocity, and n is the volume density of the electrons. Thus the appearance of a pseudoproper TCS is a relativistic effect in a spin AFM.

Even in this extremely simplified model we see the fundamental difference between toroidal ordering and magnetic quadrupole ordering. The latter would correspond to a shift of the current loops along their common axis, and not perpendicular to it as in the toroidal case.

In what follows we use a method that permits formulation of the conditions under which a TCS can arise in a spin AFM. Specifically, we are referring to the conditions imposed by the presence of some symmetry element in the crystal space group of the magnet, the position of the magnetic atoms in the lattice, and, finally, the character of the magnetic spin ordering (the orientation of the vector **L** with respect to the crystallographic directions). All the crystal space groups are tabulated, permitting classifications of pseudoproper toroidal magnets according to the type of invariants that give rise to the TCS. Following the weak ferromagnetism theory of Dzyaloshinskiĭ,[16] or task is to analyze whether the corresponding invariant combinations of vectors **T** and **L** can exist. In its main features this approach is similar to that taken by Turov[17] in classifying weak ferromagnets.

We recall that an AFM structure is called even with respect to a given symmetry element if the transformation corresponding to this element permutes the magnetic moments within a single magnetic sublattice, and odd if it interchanges the magnetic moments of different magnetic sublattices. In view of the symmetry properties of the vector **T**, we can conclude that toroidal ordering is possible only in AFM structures which are even with respect to all translations and

TABLE II.

№	Symmetry element	Parity of structure	Invariant
1	$\bar{1}$*	+	—
2		—	$L_xT_x, L_xT_y, L_xT_z, L_yT_x, L_yT_y, L_yT_z, L_zT_x, L_zT_y, L_zT_z$
3	2_z**	+	$L_xT_x, L_xT_y, L_yT_x, L_yT_y, L_zT_z$
4		—	$L_xT_z, L_yT_z, L_zT_x, L_zT_y$
5	3_z***	+	$L_zT_z, L_xT_x+L_yT_y, L_xT_y-L_yT_x;$ $T_z(L_x\pm iL_y)^3, (L_x\pm iL_y)^2(T_x\pm iT_y)L_z$
6		—	—
7	4_z	+	$L_xT_x+L_yT_y, L_zT_z, L_xT_y-L_yT_x; (L_xT_y-L_yT_x)\times$ $\times(L_x^2+L_y^2), (L_xT_y-L_yT_x)L_z^2, (L_xT_x+L_yT_y)L_z^2,$ $T_zL_z^3, (L_xT_x+L_yT_y)(L_x^2+L_y^2), T_zL_z(L_x^2+L_y^2),$ $(L_xT_x-L_yT_y)L_xL_y, (L_xT_y+L_yT_x)L_xL_y, (L_xT_y+$ $+L_yT_x)(L_x^2-L_y^2), (L_xT_x-L_yT_y)(L_x^2-L_y^2)$
8		—	$L_xT_x-L_yT_y, L_xT_y+L_yT_x; (L_xT_x-L_yT_y)\times$ $\times(L_x^2+L_y^2), (L_xT_x-L_yT_y)L_z^2, (L_xT_y+L_yT_x)\times$ $\times(L_x^2+L_y^2), (L_x^2-L_y^2)(L_xT_x+L_yT_y), (L_xT_y+$ $+L_yT_x)L_z^2, (L_xT_x+L_yT_y)L_xL_y, (L_x^2-L_y^2)L_zT_z,$ $(L_xT_y-L_yT_x)L_xL_y, L_zT_zL_xL_y, (L_xT_y-L_yT_x)\times$ $\times(L_x^2-L_y^2)$
9	6_z	+	$L_zT_z, L_xT_x+L_yT_y, L_xT_y-L_yT_x$
10		—	$T_z(L_x\pm iL_y)^3, (T_x\pm iT_y)(L_x\pm iL_y)^2L_z$

*Of the improper symmetry elements we have included only the center of inversion. For the remaining elements (with allowance for the parity condition) the invariants are easily obtained by the following rule. The invariants corresponding to an even (odd) structure with respect to some improper n-fold axis are the same as the invariants for an odd (even) proper n-fold axis of the same order n.

**The invariants for symmetry elements coinciding with other coordinate axes are obtained by cyclic permutation.

***Beginning with the threefold axis, the terms cubic in **L** are included for the purpose of constructing Table III, where the invariants cubic in **L** are nonzero for groups of intermediate and higher symmetry.

TABLE III.

№	Crystal system	Group no.	Type of structure	Invariant
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		10—15	$\bar{1}^{(-)}$, $2_z^{(-)}$	L_xT_z, L_yT_z, L_zT_x, L_zT_y
6	Orthohombic	16—24	$2_z^{(+)}$, $2_x^{(-)}$	L_xT_y, L_yT_x
7		16—24	$2_z^{(-)}$, $2_x^{(+)}$	L_yT_z, L_zT_y
8		16—24	$2_z^{(-)}$, $2_x^{(-)}$	L_zT_x, L_xT_z
9		25—46	$2_z^{(+)}$, $\bar{2}_x^{(-)}$	L_xT_x, L_yT_y, L_zT_z
10		25—46	$2_z^{(-)}$, $\bar{2}_x^{(+)}$	L_xT_z, L_zT_x
11		25—46	$2_z^{(-)}$, $\bar{2}_x^{(-)}$	L_zT_y, L_yT_z
12		47—74	$\bar{1}^{(-)}$, $2_z^{(+)}$, $2_z^{(+)}$	L_xT_x, L_yT_y, L_zT_z
13		47—74	$\bar{1}^{(-)}$, $2_z^{(+)}2_x^{(-)}$	L_xT_y, L_yT_x
14		47—74	$\bar{1}^{(-)}$, $2_z^{(-)}2_x^{(+)}$	L_yT_z, L_zT_y
15		47—74	$\bar{1}^{(-)}$, $2_z^{(-)}$, $2_x^{(-)}$	L_zT_x, 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		83—88	$\bar{1}^{(-)}$, $4_z^{(-)}$	$L_xT_x - L_yT_y$, $L_xT_y + L_yT_x$
20		89—98	$4_z^{(+)}$, $2_x^{(-)}$	$L_xT_y - L_yT_x$
21		89—98	$4_z^{(-)}$, $2_x^{(+)}$	$L_xT_x - L_yT_y$
22		89—98	$4_z^{(-)}$, $2_x^{(-)}$	$L_xT_y + L_yT_x$
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			$4_z^{(-)}$, $\bar{2}_x^{(-)}$	$L_xT_x - L_yT_y$
26		111—114	$\bar{4}_z^{(+)}$, $2_x^{(-)}$	$L_xT_y + L_yT_x$
27		121—122	$\bar{4}_z^{(-)}$, $2_x^{(+)}$	$L_xT_x + L_yT_y$, L_zT_z
28			$\bar{4}_z^{(-)}$, $2_x^{(-)}$	$L_xT_y - L_yT_x$

TABLE III. (*continued*)

№	Crystal system	Group no.	Type of structure	Invariant
29	Tetragonal	115—120	$\bar{4}_z^{(+)}, \bar{2}_x^{(-)}$	$L_xT_x - L_yT_y$
30			$\bar{4}_z^{(-)}, \bar{2}_x^{(+)}$	$L_xT_y - L_yT_x$
31			$\bar{4}_z^{(-)}, \bar{2}_x^{(-)}$	$L_xT_x + L_yT_y,\ L_zT_z$
32		123—142	$\bar{1}^{(-)}, 4_z^{(+)}, 2_x^{(+)}$	$L_xT_x + L_yT_y,\ L_zT_z$
33			$\bar{1}^{(-)}, 4_z^{(+)}, 2_x^{(-)}$	$L_xT_y - L_yT_x$
34			$\bar{1}^{(-)}, 4_z^{(-)}, 2_x^{(+)}$	$L_xT_x - L_yT_y$
35			$\bar{1}^{(-)}, 4_z^{(-)}, 2_x^{(-)}$	$L_xT_y + L_yT_x$
36	Trigonal	147—148	$\bar{3}_z^{(-)}$	$L_xT_x + L_yT_y, L_xT_y - L_yT_x, L_zT_z$
37		149—155	$3_z^{(+)}, 2_x^{(-)}$	$L_xT_y - L_yT_x$
38			$3_z^{(+)}, 2_y^{(-)}$	$L_xT_y - L_yT_x$
39		156—161	$3_z^{(+)}, \bar{2}_x^{(-)}$	$L_xT_x + L_yT_y,\ L_zT_z$
40			$3_z^{(+)}, \bar{2}_y^{(-)}$	$L_xT_x + L_yT_y,\ L_zT_z$
41		162—167	$\bar{1}^{(-)}, 3_z^{(+)}2_x^{(+)}$	$L_xT_x + L_yT_y,\ L_zT_z$
42			$\bar{1}^{(-)}, 3_z^{(+)}2_x^{(-)}$	$L_xT_y - L_yT_x$
43			$\bar{1}^{(-)}, 3_z^{(+)}2_y^{(+)}$	$L_xT_x + L_yT_y,\ L_zT_z$
44			$\bar{1}^{(-)}, 3_z^{(+)}2_y^{(-)}$	$L_xT_y - L_yT_x$
45	Hexagonal	168—173	$6_z^{(-)}$	$T_z(L_x \pm iL_y)^3,\ L_z(T_x \pm iT_y) \times$ $\times (L_x \pm iL_y)^2$
46		174	$\bar{6}_z^{(-)}$	$L_xT_x + L_yT_y,\ L_zT_z,\ L_xT_y - L_yT_x$
47		175—176	$\bar{1}^{(-)}, 6_z^{(+)}$	$L_xT_x + L_yT_y,\ L_zT_z,\ L_xT_y - L_yT_x$
48			$\bar{1}^{(-)}, 6_z^{(-)}$	$T_z(L_x \pm iL_y)^3,\ L_z(T_x \pm iT_y) \times$ $\times (L_x + iL_y)^2$
49		177—182	$6_z^{(+)}2_x^{(-)}\ (2_y^{(-)})$	$L_xT_y - L_yT_x$
50			$6_z^{(-)}, 2_x^{(-)}\ (2_y^{(+)})$	$T_z[(L_x \pm iL_y)^3 + (L_x - iL_y)^3]$ $L_z[(T_x + iT_y)(L_x + iL_y)^2 +$ $+ (T_x - iT_y)(L_x - iL_y)^2]$
51			$6_z^{(-)}, 2_x^{(+)}\ (2_y^{(-)})$	$iT_z[(L_x + iL_y)^3 - (L_x - iL_y)^3]$ $iL_z[(T_x + iT_y)(L_x + iL_y)^2 -$ $- (T_x - iT_y)(L_x - iL_y)^2]$
52			$6_z^{(+)}\sigma_x^{(-)}\ (\sigma_y^{(-)})$	$L_xT_x + L_yT_y,\ L_zT_z$

TABLE III. (*continued*)

№	Crystal system	Group no.	Type of structure	Invariant
53		183—186	$6_z^{(-)}\sigma_x^{(-)}\ (\sigma_y^{(+)})$	$iT_z[(L_x+iL_y)^3-(L_x-iL_y)^3]$ $iL_z[(T_x+iT_y)(L_x+iL_y)^2-$ $-(T_x-iT_y)(L_x-iL_y)^2]$
54			$6_z^{(-)}\sigma_x^{(+)}$ $(\sigma_y^{(-)})$	$T_z[(L_x+iL_y)^2+(L_x-iL_y)^3]$ $L_z[(T_x+iT_y)(L_x+iL_y)^2+(T_x-$ $-iT_y)(L_x-iL_y)^2]$
55	Hexagonal	187—188	$\bar{6}_z^{(-)}\sigma_x^{(+)}\ (\sigma_y^{(+)})$	$L_xT_y-L_yT_x$
56			$\bar{6}_z^{(-)}\sigma_x^{(-)}\ (\sigma_y^{(-)})$	$L_xT_x+L_yT_y,\ L_zT_z$
57			$\bar{6}_z^{(+)}\sigma_x^{(-)}$	$iT_z[(L_x+iL_y)^3-(L_x-iL_y)^3]$ $iL_z[(T_x+iT_y)(L_x+iL_y)^2-$ $-(T_x-iT_y)(L_x-iL_y)^2]$
58			$\bar{6}_z^{(+)}\sigma_y^{(-)}$	$T_z[(L_x+iL_y)^3+(L_x-iL_y)^3]$ $L_z[(T_x+iT_y)(L_x+iL_y)^2+$ $+(T_x-iT_y)(L_x-iL_y)^2]$
59		189—190	$\bar{6}_z^{(-)}2_x^{(+)}\ (2_y^{(+)})$	$L_xT_x+L_yT_y,\ L_zT_z$
60			$\bar{6}_z^{(-)}2_x^{(-)}\ (2_y^{(-)})$	$L_xT_y-L_yT_x$
61		189—190	$\bar{6}_z^{(+)}2_x^{(-)}$	$T_z[(L_x+iL_y)^3+(L_x-iL_y)^3]$ $L_z[(T_x+iT_y)(L_x+iL_y)^2+$ $+(T_x-iT_y)(L_x-iL_y)^2]$
62			$\bar{6}_z^{(+)}2_y^{(-)}$	$iT_z[(L_x+iL_y)^3-(L_x-iL_y)^3]$ $iL_z[(T_x+iT_y)(L_x+iL_y)^2-$ $-(T_x-iT_y)(L_x-iL_y)^2]$
63		191—194	$\bar{1}^{(-)}6_z^{(+)}2_x^{(+)}\ (2_y^{(+)})$	$L_xT_x+L_yT_y,\ L_zT_z$
64			$\bar{1}^{(-)}6_z^{(+)}2_x^{(-)}$	$L_xT_y-L_yT_x$
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		191—194	$\bar{1}^{(-)}6_z^{(-)}$ $2_x^{(-)}\ (2_y^{(+)})$	$T_z[(L_x+iL_y)^3+(L_x-iL_y)^3]$ $L_z[(T_x+iT_y)(L_x+iL_y)^2+$ $+(T_x-iT_y)(L_x-iL_y)^2]$
67		200—206	$\bar{1}^{(-)}3_{xyz}^{(+)}2_z^{(+)}$	$L_xT_x+L_yT_y+L_zT_z$
68		207—214	$4_z^{(-)}3_{xyz}^{(+)}$	$L_xT_x(L_y^2-L_z^2)+L_yT_y(L_z^2-$ $-L_x^2)+L_zT_z(L_x^2-L_y^2)$
69	Cubic	215—220	$\bar{4}_z^{(-)}3_{xyz}^{(+)}$	$L_xT_x(L_y^2+L_z^2)+L_yT_y(L_z^2+L_x^2)+$ $+L_zT_z(L_x^2+L_y^2)$
70		221—230	$\bar{1}^{(-)}3_{xyz}^{(+)}4_z^{(+)}$	$L_xT_x(L_y^2+L_z^2)+L_yT_y(L_z^2+L_x^2)+$ $+L_zT_z(L_x^2+L_y^2)$
71			$\bar{1}^{(-)}3_{xyz}^{(+)}4_z^{(-)}$	$L_xT_x(L_y^2-L_z^2)+L_yT_y(L_z^2-L_x^2)+$ $+L_zT_z(L_x^2-L_y^2)$

odd with respect to the center of symmetry if the system has one (in describing specific structures below, we denote the parity by the sign ($\pm$), e.g., $\sigma_z^{(+)}, \bar{1}^{(\pm)}, 2_z^{(\pm)}$).

We thus have the following conditions for an AFM structure to admit a TCS: a) The magnetic and crystal-chemical unit cells must be the same, b) the directions of the magnetic moments at all sites belonging to the same Bravais lattice must be the same, c) the directions of the magnetic moments at sites which map into each other under spatial inversion must be exactly opposite.

The invariant combinations of **L** and **T** for the proper and improper rotations characteristic of crystal lattices are tabulated in Table II, by means of which, if one knows the specific crystallographic structure of a magnet, one can find the AFM structures which admits a TCS and also the directions of the vectors **L** for which $\mathbf{T} \neq 0$. In the general case one must use Table II for the corresponding generators of the groups and thereby determine the invariant in the thermodynamic potential that gives rise to the TCS. We note that if the magnetic atoms are located on a simple (nonscrew) rotation axis or on a simple (nonglide) symmetry plane, the AFM structure is always even with respect to these elements.

Triclinic system($\bar{1}$)

If the positions of the magnetic sites are of multiplicity one, AFM ordering cannot occur without an increase in the dimensions of the unit cell, and, accordingly, no TCS can arise. If the positions of the magnetic sites are of multiplicity two, connected by a center of symmetry, then a TCS is possible (No. 2 in the Fedorov classification).

Monoclinic system (2,*m*,2/*m*)

a) No center of symmetry (2,*m*). There is only one independent symmetry element (a plane of twofold axis), with respect to which the AFM structure must be odd. (Nos. 3–9). The magnetic sites must occur in general positions.

b) There is a center of symmetry (2/*m*), and since the AFM structure must be odd with respect to it, the structure can be even or odd with respect to the axis 2 (Nos. 10–15). The magnetic sites can be in general crystallographic positions or in particular positions that do not coincide with the center of inversion (as we know, general positions include not only those lattice points through which no symmetry element passes but also positions lying on screw axes and glide planes).

Orthohombic system (222,*mm*2,*mmm*)

a) No center of symmetry (222,*mm*2). In such groups the two independent symmetry elements are two mutually perpendicular axes 2 or two mutually perpendicular planes *m*. In the case of the two axes 2, which generate a third axis 2 perpendicular to them, the structure must be even with respect to one axis and odd with respect to the other two (Nos. 16–24). In the case of the two planes, the AFM structure must be odd with respect to the axis 2 and even with respect to the planes *m* or else even with respect to the axis 2 and to one of the planes *m* but odd with respect to the other (Nos. 25–46). In both the first and second cases the magnetic sites can occupy general or particular positions in the lattice. For example, if the magnetic sites lie on a rotation axis 2 or mirror plane *m*, only one AFM structure admits a TCS, namely, that for which the two other symmetry elements take sites having antiparallel magnetic moments into one another. On the other hand, if there are magnetic sites lying at intersections of rotation axes or mirror planes, there can be no AFM structure and no TCS.

b) There is a center of symmetry (*mmm*). For general positions the possible structures, as in case a, are odd with respect to one axis 2 and even with respect to the other two; in addition, an AFM structure that is even with respect to all the axes 2 can exist, but it must be odd with respect to the inversion center (Nos. 47–74). For particular positions, when the sites lie on a rotation axis or mirror plane, only two types of AFM structures are possible (even or odd with respect to the other two axes or planes). If magnetic sites lie at points of intersection of two reflection axes or on the line of intersection of two mirror planes, the AFM structure must be even with respect to all the rotation elements.

For crystals of higher symmetry (the tetragonal, trigonal, hexagonal, and cubic systems) the treatment becomes more awkward but in principle can be carried out in an analogous way. The results are collected in Table III, which gives the number of the space group in the first column, the possible types of AFM structures which generate a TCS in the second column, and the explicit forms of the invariants in the third column. If the number of a group is missing from the table, it means that no TCS can exist in that group (at least on account of linear or cubic invariants). Formally, situations are possible in which **T** is generated by some invariants of higher order in **L**, but we are not considering such systems in this paper. Therefore, conclusions as to the possibility or impossibility of a TCS should henceforth be taken to mean the presence or absence in Table III of invariants linear (or cubic) in **L**.

3. SPECIFIC SAMPLES OF ANTIFERROMAGNETS WITH A TCS

Before applying the approach developed in the previous section to the analysis of specific AFM compounds, let us make the observation that it can be pointless to discuss a pseudoproper TCS without reference to the specific AFM structure that generates it. The reason is that in listing in the corresponding column of Table III the types of AFM structures which generate a TCS for each crystal system, we have implicitly understood that the magnetic atoms occupy general sites in the lattice. In turning to real compounds,[19] however, we see that in the majority of cases the magnetic atoms lie on some symmetry elements (rotation axes or symmetry planes); in other words, they occupy particular positions. The transition from a general to a particular type of site can lead to a violation of the sufficient conditions for the existence of a TCS.

Let us begin with the space group D_{2h}^{16} (*Pnma*), which, for methodological reaons, we shall elaborate in greater detail. If we place the magnetic atom in the general position $8d(x,y,z)$ (Ref. 18) and use the eight existing symmetry elements (in the point-group notation these elements are E, 2_x, 2_y, 2_z, $\bar{1}$, σ_x, σ_y, and σ_z), we obtain eight atoms in the unit cell, with coordinates

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

We denote the corresponding local spin moments as $\mathbf{S}_i$ ($i=1,\dots,8$). The presence of the inversion transformation, which connects the pairs of sites $1\leftrightarrow5, 2\leftrightarrow6, 3\leftrightarrow7, 4\leftrightarrow8$, and the condition that the AFM structure be odd with respect to the inversion center imply that $\mathbf{S}_1=-\mathbf{S}_5$, $\mathbf{S}_2=-\mathbf{S}_6$, $\mathbf{S}_3=-\mathbf{S}_7$, and $\mathbf{S}_4=-\mathbf{S}_8$. With allowance for the permutations of the atoms under the remaining symmetry elements, we obtain four types of collinear AFM structures which admit a TCS:

$$\begin{array}{ll} S_1 \parallel S_2 \parallel S_3 \parallel S_4 & (\bar{1}^{(-)}2_z^{(+)}2_x^{(+)}), \\ S_1 \parallel S_4 \parallel S_6 \parallel S_7 & (\bar{1}^{(-)}2_z^{(+)}2_x^{(-)}), \\ S_1 \parallel S_2 \parallel S_7 \parallel S_8 & (\bar{1}^{(-)}2_z^{(-)}2_x^{(+)}), \\ S_1 \parallel S_3 \parallel S_6 \parallel S_8 & (\bar{1}^{(-)}2_z^{(-)}2_x^{(-)}). \end{array} \qquad (9)$$

There are also three types of particular positions of the magnetic atoms, each of multiplicity 4. The positions of the first two types[19] ($4a$ and $4b$) coincide with a center of symmetry having coordinates (0,0,0) for position $4a$ and $(0,0,\frac{1}{2})$ for position $4b$. The third position, $4c$, with coordinates $(x,\frac{1}{4},z)$, is found in the plane σ_y. In going from the general position $8d$ to the particular positions $4a$ and $4b$, sites 1–4 become superposed on sites 5–8, and the spin AFM structure generating the TCS disappears. Such a situation arise, for example, in yttrium orthoferrites with the structure $\mathrm{YMeO_3}$ (Me is a $3d$ transition element), where the magnetic atoms Me occupy $4b$ positions.

In going from position $8d$ to $4c$, the points connected by the plane σ_y become superposed, ie., $1\leftrightarrow7, 2\leftrightarrow8, 3\leftrightarrow5, 4\leftrightarrow6$, so that instead of eight sites in the unit cell there are four: I (in place of 1 and 7), II (in place of 3 and 5), III (in place of 2 and 8), and IV (in place of 4 and 6). In particular, this happens in the compound α-FeOOH, in which the Fe ions occupy $4c$ positions. The AFM structure which arises is of the type

$$(S_{\mathrm{I}} \| S_{\mathrm{III}}) = -(S_{\mathrm{II}} \| S_{\mathrm{IV}}) \quad (\bar{1}^{(-)}2_z^{(-)}2_x^{(+)}) \qquad (10)$$

and admits a TCS with invariant L_yT_z (see Table III). Interestingly, three compounds in this group, $\mathrm{LiCoPO_4}$, $\mathrm{LiMnPO_4}$, and $\mathrm{LiNiPO_4}$, in which the magnetic atoms Co, Mn, and Ni also occupy $4c$ positions, have a structure $\bar{1}^{(-)}2_z^{(-)}2_x^{(+)}$. Here the antiferromagnetism vectors are directed along the axes y (Co), x (Mn), and z (Ni), and the corresponding invariants are different. A similar situation arises in $\mathrm{Co_2SiO_4}$ and $\mathrm{KFeCl_3}$, where the magnetic ions Co or Fe also occupy $4c$ positions, but the AFM structure here is even with respect to the center of inversion and no TCS can exist.

Let us now turn to a systematic study of the compounds belonging to all the crystal systems, beginning with monoclinic (we know of no real antiferromagnet belonging to the triclinic system, although for space group No. 2 the existence of a TCS is not forbidden by symmetry).

A compound belonging to the group $C_{2h}^2\left(P\frac{2_1}{m}\right)$ is ErOOH, where the Er ions, whose magnetic moments are directed along the y axis, occupy the particular positions $2e$, having multiplicity two. According to the general classification, the AFM structure is of the form

$$S_1(S_4) = -S_2(S_3) \quad (\bar{1}^{(-)}2_y^{(-)}) \qquad (11)$$

and, according to Table III, admits a TCS corresponding to the invariants L_yT_x and L_yT_z. In the isomorphic compound DyOOH the magnetic moments of the Dy ions also occupy $2e$ positions but lie in the xz plane. In this case the invariants are of the form L_xT_y, L_zT_y.

The groups of interest in the rhombic system, in addition to the group D_{2h}^{16} considered above, are D_{2h}^{14} ($Pbna$)—the compound $\mathrm{CrUO_4}$— and D_{2h}^{17} ($Cmcm$)—a representative of which is γ-FeOOH. For $\mathrm{CrUO_4}$ the magnetic ions Cr and U occupy the particular positions $4c$. The spin AFM structure is of the type $\bar{1}^{(-)}2_z^{(-)}2_x^{(+)}$, with the moments having only a component along the y axis (L_y),

$$(S_1 \| S_4) = -(S_2 \| S_3), \qquad (12)$$

and admits a TCS with the invariant L_yT_z.

For γ-FeOOH the Fe ions occupy positions $4c$, the AFM structure is of the type $\bar{1}^{(-)}2_z^{(-)}2_x^{(-)}$, $(S_1\|S_3) = -(S_2\|S_4)$, and the TCS is due to the invariant L_xT_z (since the spins are directed along the x axis). We note that in the compound $\mathrm{CrVO_4}$, which belongs to the same group D_{2h}^{17}, the Cr ions occupy positions $4a$, the AFM structure is even with respect to the center of inversion, and no TCS can exist.

Let us turn to the tetragonal system. The group $D_{4h}^{14}\left(P\frac{4_2}{m}nm\right)$ is represented by $\mathrm{Cr_2WO_6}$. The Cr ions occupy positions $4e$, the magnetic moments lie in the basal plane xy, the structure is of the type

$$(S_1 \| S_4) = -(S_2 \| S_3) \quad (\bar{1}^{(-)}4_z^{(-)}2_x^{(+)}), \qquad (13)$$

and the TCS is due to the invariant $L_xT_x - L_yT_y$.

The group C_{4v}^{10} ($I4cm$) is represented by $\mathrm{KCrF_3}$. The Cr ions occupy positions $4e$, the magnetic moments lie in the xy plane, the structure is of the type

$$(S_1 \| S_2) = -(S_3 \| S_4) \quad (4_z^{(-)}\sigma_x^{(-)}), \qquad (14)$$

and the TCS is due to an invariant of the form $L_xT_x - L_yT_y$. In the compound $\mathrm{Fe_2TeO_6}$, which belongs to the same symmetry group, the AFM structure is of the type $1^{(-)}4_z^{(+)}2_x^{(-)}$, and there is no TCS in this case, since the magnetic moments of the Fe ions are directed along the z axis and the necessary invariants are lacking. For this same reason there is no TCS in the compound CeC_2 (group $D_{4h}^{17}\left(I\frac{4}{m}mm\right)$), although the AFM structure is of the type $\bar{1}^{(-)}4_z^{(+)}2_x^{(-)}$.

In the hexagonal system, let us consider the group $D_{3d}^{6}(R\bar{3}c)$, represented by V_2O_3. The V ions occupy positions $4c$ (on the threefold axis). Strictly speaking, the magnetic moments in this compound cannot be considered well localized, but neutron-diffraction measurements[18] of these moments imply that the maximum spin density is distributed at the V sites, and the spin vectors at the sites are oriented along the threefold axis. The antiferromagnetism vector for the V_2O_3 structure is of type 1 (Ref. 20):

$$\mathbf{L}=\mathbf{S}_1-\mathbf{S}_2-\mathbf{S}_3+\mathbf{S}_4 \quad (\bar{1}^{(-)}\,3_z^{(+)}\,2_x^{(+)}).$$

The sole invariant that admits a TCS is of the form (L_zT_z), i.e., the symmetry of the vectors **L** and **T** is the same in V_2O_3 (this is due exclusively to the nature of the spin density distribution).

Crystals of the cubic system are in a special category. In all the wide variety of cubic AFM we could not find a single one in which the conditions for the existence of a TCS would be satisfied. At the same time, we see from Table III that there is no fundamental reason why TCS should not form in cubic AFM.

4. ANTIFERROMAGNETIC RESONANCE AND REARRANGEMENT OF THE SPIN STRUCTURE IN PSEUDOPROPER TOROIDAL SYSTEMS

By itself, the appearance of a pseudoproper TCS does not lead to any new effects beyond those associated with the proper TCS discussed previously[7] (in the latter the toroidal ordering arises in the exchange approximation and is not relativistically small). On the other hand, pseudoproper toroidal systems have the distinctive feature that the spin AFM structure of the ground state can be influenced through the toroidal subsystem, as can the collective spin excitations, i.e., antiferromagnons. The latter possibility is due to the intermixing of the antiferromagnons and the toroidal oscillations. A phenomenological theory of toroidal oscillations (oscillations of the orbital moment density) was constructed in Ref. 12, and certain microscopic aspects were investigated in Ref. 21.

Let us first discuss the rearrangement of the spin AFM structure in pseudoproper toroidal systems. In the model of a uniaxial AFM with two magnetic ions in the unit cell, in neglect of spatial dispersion and in the absence of external fields, we write the thermodynamic potential Φ of the system near the Néel point to lowest order in the vectors **L** and **T** as

$$\Phi=\frac{\alpha}{2}\mathbf{L}^2+\frac{K}{2}\mathbf{L}_z{}^2+\frac{\beta}{4}\mathbf{L}^4+\mathbf{f}[\mathbf{L}\mathbf{T}]+\frac{d}{2}\mathbf{T}^2, \qquad (15)$$

where the square brackets denote the vector (cross) product, K is the magnetic anisotropy constant, $\mathbf{f}=f\mathbf{z}$, **z** is a vector in the direction of the principal axis, the coefficients β and d are positive, and the coefficient α changes sign at the Néel point ($\alpha\propto\Theta-\Theta_N, \Theta_N$ is the Néel temperature); K and f are relativistically small [$|K|, f\sim(v/c)^2$], while the remaining quantities are nonzero in the exchange approximation.

Minimizing (15) with respect to **T** and then substituting the equilibrium value $\mathbf{T}_0=-(\mathbf{f}\times\mathbf{L})/d$, we obtain

$$\Phi=\frac{\tilde{\alpha}}{2}\mathbf{L}^2+\frac{\tilde{K}}{2}\mathbf{L}_z{}^2+\frac{\beta}{4}\mathbf{L}^4, \quad \tilde{\alpha}=\alpha-\frac{f^2}{d}, \quad \tilde{K}=K+\frac{f^2}{d}. \qquad (16)$$

It is clear from (16) that in the absence of external sources the onset of a pseudoproper TCS has practically no effect on either the position of the Néel point or on the value of the magnetic anisotropy; in what follows we therefore neglect the corrections $\sim(v/c)^4$ to α and K.

The situation is much more interesting if a uniform TCS is induced by external influence. The means by which a TCS can be induced have been discussed previously[7,10]:

1) the application of crossed electric and magnetic fields, giving rise to a vector **T** through the magnetoelectric effect:

$$\mathbf{T}_0^{(1)}\propto[\mathbf{E}\mathbf{H}]/d; \qquad (17)$$

2) the excitation of a TCS through the photovoltaic effect by an external Ohmic current $\mathbf{j}_0$ (not only in metallic magnets but also in semiconductors and insulators in the presence of nonequilibrium carriers) or by a photocurrent in crystals lacking a center of inversion:

$$\mathbf{T}_0^{(2)}\propto\mathbf{j}_0/dc; \qquad (18)$$

3) the induction of a TCS by an electric field **E** through the disturbance of the phase of the electronic states forming the toroidal moment in the presence of electron–impurity or electron–phonon collisions:

$$\mathbf{T}_0^{(3)}\propto\nu\mathbf{E}/d, \qquad (19)$$

where ν is the collision rate. Effects (17) and (19) occur even in the exchange approximation, whereas (18) is relativistically small.

It follows from (15) that for $\mathbf{f}\neq0$ an AFM ordering in the xy plane will be induced above the Néel point:

$$\mathbf{L}_0=-[\mathbf{T}_0\mathbf{f}]/d. \qquad (20)$$

For an easy-axis AFM ($K<0$), $|\mathbf{L}_0|$ reaches a value $\sim T_0$ at the Néel point ($\alpha-K=0$), i.e., it is not relativistically small in the case of mechanism (17) or (19). Below the Néel point the structure becomes canted in the xy plane. For an easy-plane AFM ($K>0$) the induction of a TCS leads to anisotropy in the xy plane; by changing the direction of the external fields in (17)–(19), one can reorient the spin sublattices ($\mathbf{L}_0\perp\mathbf{T}_0$). Because of the temperature dependence of the coefficients of proportionality[10] in (17)–(19), all the effects mentioned can have extremely specific behavior as functions of temperature [for example, in case (19) one has $L_0\propto\nu(\Theta)\sim\Theta^p$, where the exponent p depends on the particular electron damping mechanism]. For "soft" toroidal systems one has $d\propto\Theta-\Theta_T$ (Θ_T is the temperature of the proper toroidal transition) and even for $\Theta_T>\Theta_N$ the effects of induction of a spin AFM structure can increase anomalously, since $T_0 \propto d^{-1} \propto (\Theta - \Theta_T)^{-1}$.

It should be stressed that the "current" mechanism (18) is ineffective from the standpoint of inducing a spin AFM structure, since (18) occurs together with the stronger mechanism of direct induction of a current $\mathbf{j}_0$ through the magnetic field $\mathbf{H}$ ($\mathrm{curl}\mathbf{H} = 4\pi\mathbf{j}_0/c$). Actually, even in the exchange approximation the potential Φ should contain terms of the type $\mathbf{M}^2$, $\mathbf{L}\mathrm{curl}\mathbf{M}$ (Ref. 18), and $-\mathbf{M}\cdot\mathbf{H}$, which, after minimization with respect to $\mathbf{M}$, lead to terms in Φ which are linear in v/c [in case (18) a contribution of order $(v/c)^3$ appears]. The contribution of mechanism (17) is apparently noticeable only in soft toroidal systems, in which Θ_T is close to Θ_N.

Let us now turn to the influence of the induced TCS on the AFMR frequency for $\Theta \ll \Theta_N$. The thermodynamic potential of an easy-plane AFM can be written

$$\Phi=\Phi_0+\frac{K}{2}L_z^2+\mathbf{f}[\mathbf{L}\mathbf{T}_0]+\frac{a}{2}\mathbf{M}^2, \qquad (21)$$

where Φ_0 is the contribution containing exchange terms of the type $\mathbf{L}^2$, $\mathbf{L}^4$, etc., and $\mathbf{M}$ is the magnetization vector. Neglecting magnon damping, we write the Landau-Lifshitz equations for the oscillations of $\mathbf{L}$ and $\mathbf{M}$:

$$\begin{aligned}\frac{1}{\gamma}\dot{\mathbf{L}}&=\left[\mathbf{L}\frac{\delta\Phi}{\delta\mathbf{M}}\right]+\left[\mathbf{M}\frac{\delta\Phi}{\delta\mathbf{L}}\right],\\ \frac{1}{\gamma}\dot{\mathbf{M}}&=\left[\mathbf{M}\frac{\delta\Phi}{\delta\mathbf{M}}\right]+\left[\mathbf{L}\frac{\delta\Phi}{\delta\mathbf{L}}\right],\end{aligned} \qquad (22)$$

where γ is the gyromagnetic ratio.

The variation in (22) is done under the condition $|\mathbf{L}| = L_0 = \mathrm{const}$. Under steady-state conditions equations (22) are satisfied identically for $\mathbf{M}_0 = 0$,

$$\mathbf{L}_0=L_0[\mathbf{f}\mathbf{T}_0]/|[\mathbf{f}\mathbf{T}_0]|.$$

Linearizing (22) with respect to the corrections $\delta\mathbf{L} = \mathbf{L} - \mathbf{L}_0$ and $\mathbf{M}$ and then evaluating the determinant of the system, we obtain the spectrum of AFMR frequencies to lowest order in $(v/c)^2$ as

$$\omega_1^2/\gamma^2=aL_0^2(K+|[\mathbf{f}\mathbf{T}_0]|/L_0) \qquad (23)$$

for the oscillations of the component L_z, and

$$\omega_2^2/\gamma^2=aL_0|[\mathbf{f}\mathbf{T}_0]| \qquad (24)$$

for the oscillations of the component of $\mathbf{L}$ in the xy plane: $\delta L_\perp \perp \mathbf{L}_0$.

For an easy-axis AFM it is easy to show that to first order in $\mathbf{f}$ there is no renormalization of the AFMR frequencies. Thus, the induction of a TCS causes an increase in the stiffness of the antiferromagnons in an easy-plane AFM, and for branch (24) the spectrum develops a gap in proportion to $\mathbf{T}_0$.

Another interesting effect can occur in the excitation of low-frequency toroidal oscillations (by light, for example). In this case the bilinear invariant in (21) leads to the induction of a low-frequency component of the vector $\mathbf{L}$, i.e., to the generation of magnons. The detailed treatment of this effect requires an analysis in the spirit of the toroidal dynamics of Ref. 12 and will not be given here.

5. CONCLUSION

This investigation of the pseudoproper toroidal current state in antiferromagnets has yielded the following conclusions.

1. There is a large class of specific compounds of various symmetry types (predominantly of lower to intermediate symmetries) in which toroidal ordering is induced by a spin antiferromagnetic ordering in accordance with the type of magnetic structure. Thus the formation of a pseudoproper TCS is no more exotic than weak ferromagnetism.

2. Even if a compound has a favorable crystal symmetry the formation of a TCS is subject to important constraints imposed by the type of magnetic atoms, the orientation of their magnetic moments, and their positions in the unit cell.

3. In this paper we have considered only the simplest (bilinear) kinds of invariants which intermix the spin antiferromagnetic and orbital TCS structures. Allowance for invariants of higher order in $\mathbf{L}$ and for the contributions due to striction excitations might expand the number of crystals which admit a TCS. Here the system in which the TCS has arisen will, of course, belong to one of the 31 magnetic symmetry classes in Table I.

We have considered collinear AFM structures and the uniform TCS which they generate. Going to more complex (canted, long-period, etc.) structures may lead to more complicated types of TCS. For example, in rare earth orthoferrites, where the unit cell contains two kinds of magnetic ions (4*f* and 3*d* metals), a canted AFM structure can arise in the rare earth metal sublattice as a result of competition between the intrasublattice and intersublattice exchange and relativistic interactions. Since the rare earth metal ions occupy the 4*c* positions, which are favorable for the TCS (the 3*d* metal ions are found in the unfavorable 4*b* positions), a nonuniform structure arises which is described by higher toroidal multipoles.

Long-period AFM structures have different parity requirements: Incommensurate TCS can now be generated on account of invariants of the type $\mathbf{T}\mathrm{curl}\mathbf{L}$ (here $\mathbf{L}$ can be even with respect to the inversion center). The class of systems in which it is worthwhile to search for TCS is thus broadened.

We have not touched on AFM structures in which there is multiplication of the period of the unit cell. It is easy to see, however, that the TCS structures corresponding to them, if such structures exist, should be of the antitoroidal type and can be described only with the aid of higher toroidal multipoles.

It follows from Table I that a TCS can also arise in ferromagnets on account of invariants of the form $\mathbf{T}\cdot\mathbf{M}$. A classification of the ferromagnets (including weak ferromagnets) that admit a pseudoproper TCS can be done in the spirit of the present study for the case of antiferromagnets. Here, too, the invariant $\mathbf{T}\cdot\mathbf{M}$ will clearly be of a relativistic nature.

Since the symmetry properties of $\mathbf{M}$ are the same as those of the angular velocity vector $\mathbf{\Omega}$, one suspects that such systems will exhibit gyrotoroidal phenomena—interconnections between the microcurrents generating the toroidal moment and the macroscopic rotation of the crystal (the toroidal analogs of the Einstein, de Haas, and Barnett effects).

One could try to observe the gyrotoroidal effects by applying crossed electric **E** and magnetic **H** fields to the system [**T** and **M** are due to the presence of the invariants $\mathbf{T}\cdot(\mathbf{E}\times\mathbf{H})$ and $\mathbf{M}\cdot\mathbf{H}$] and then turning off **E**. By virtue of the conservation of angular momentum and the presence of the invariant $\mathbf{T}\cdot\boldsymbol{\Omega}$, the vanishing of **T** should import a mechanical rotation to the sample.

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1) To emphasize the orbital nature of the toroidal ordering, we use the term "toroidal current state" (TCS) to describe it.[11]

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