Ferroelectrics, **252**, 41-50, (2001)
(Proceedings of the International Symposium on Ferroic Domains and Mesoscopic Structures, ISFD-6, Nanjing, China, May 29-June 2, 2000)

On Ferrotoroidics and Electrotoroidic, Magnetotoroidic and Piezotoroidic Effects

Dedicated to Edgar Ascher at the occasion of his 80's birthday

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Some historical aspects of the discovery of the "axio-polar" (time-odd polar) vector, its appearance as an anapole moment in atomic nuclei and in the form of a spontaneous toroidal moment in "ferrotoroidic" crystals and domains are discussed. The related linear and bilinear "electrotoroidic" and "magnetotoroidic" effects, as well as the "piezotoroidic" and linear "toroido-optic effect" are postulated.

KEYWORDS : *toroidal moment; ferrotoroidic domains; magnetoelectric; electrotoroidic; magnetotoroidic; piezotoroidic*

> Il est évident que le qualitatif et le quantitatif ne constituent ni une hierarchie des valeurs dans la connaissance, ni un ordre chronologique dans le développement de cette dernière, mais bien deux de ses aspects complémentaires
>
> Edgar Ascher

1. INTRODUCTION

Although Pierre Curie described already geometrically in 1894 the axial vector symmetry of a magnetic field correctly by designing a circle with a small tangential arrow perpendicularly to the field [1], it became only clear with Wigner's introduction of the symmetry operation time reversal 1' in 1932 [2] that the axial vector magnetic field **H** (or magnetization) changes sign under application of 1' (1' **H** = **- H**) and that the polar vector electric field **E** (or polarization) remains unchanged by 1' (1' **E** = **E**), vice versa, the electric field (or polarization) changing sign under space inversion $\bar{1}$ ($\bar{1}$ **E** = **- E**) and the magnetic field **H** (or magnetization) remaining unchanged by $\bar{1}$ ($\bar{1}$ **H** = **H**). It took many more years that the existence of a special type of vector was disclosed, a kind of polar vector changing sign both under time reversal and space inversion. It was Ya. B. Zel'dovich in 1957 [3], who showed that a system which does not transform into itself under space inversion, i.e. has no parity, generates a special distribution of magnetic fields like in a toroidal winding and differing from common electromagnetic multipoles like dipole and electromagnetic quadrupole [4]. For this special source of electromagnetic fields, more precisely for the spin part of the toroidal dipole [5], the term "anapole" was suggested by A.S. Kompanets [3]. The anapole remained for many years a theoretical curiosity in atomic and nuclear physics and it is remarkable that in 1997, after long experimental and theoretical efforts, for the first time an anapole moment has been discovered by optical spectroscopy using an interference technique, the anapole of the cesium ^{133}Cs nucleus! [6,7]. This provided the first measurement of a nuclear spin dependence to atomic parity violation. The anapole violates parity and charge conjugation invariance. There are two sources of parity nonconservation in atoms, electron-nucleus weak interaction, predicted by Lee and Yang in 1956[7a] and the magnetic interaction of electrons with the nuclear anapole moment [8].

Independently of this research line of theoretical and experimental nuclear physics, Edgar Ascher classified in 1966 [9] the current density **j** as an "axio-polar" vector, i.e. *a polar vector, which changes sign both under space and time reversal* and determined the 31 Shubnikov point groups of crystals, allowing this kind of vector, 13 of which were found to admit also a spontaneous magnetization vector. He also showed that velocity, linear momentum and some other physical quantities are transforming in the same way as **j**[9],[10]. In an attempt at explaining the symmetry of superconductors, Ascher conjectured that under certain conditions domains of "spontaneous currents", with very small crystalline anisotropy and an axio-polar point group, might form and arrange in closed loops of spontaneous current and explain the symmetry and some properties of superconductors. This hope was not fulfilled, but it was realized later [11] that the axio-polar vector "spontaneous toroidal moment" **T**, which also changes sign both under space and time reversal ($\bar{1}\,\mathbf{T} = -\,\mathbf{T}$, $1'\,\mathbf{T} = -\,\mathbf{T}$), must be allowed in the same 31 Shubnikov point groups as determined by Ascher. Simple geometrical representations of a toroidal moment are a solenoid formed into a torus, with an *even number of windings* [12], four spin-bearing ions in the (001)-plane of a tetragonal unit cell with a head-to-tail arrangement of the spins, or the four possible triangular ferromagnetic domains of Aizu species 4/mmm1'/Fm'm'm(s), even though a m'm'm single domain does not allow a spontaneous toroidal moment!It was pointed out [5] that A.V. Shubnikov in 1975 [13] was probably the first to pay attention to the symmetry of a circular magnetic field, representing it by an arrow with a conical tip, but he did not go farther.

Ascher classified the quantities occurring in Maxwell's equations, i.e. charge density ρ, polarization **P**, magnetization **M** and current density **j** with respect to the four irreducible representations of the dihedral group $\bar{1}1'$ of order four (generated by space inversion $\bar{1}$ and time reversal 1' [9; Table 1 in:10]), corresponding to the identity, space inversion $\bar{1}$, time reversal 1' and the product of both $\bar{1}'$, respectively. In Table 1 a slightly modified version [14] of Ascher's Table is shown, including the magnetic limiting point groups and where **P**, **M**, and **T** (replacing **j**) stand for the vectors polarization (polar), magnetization (axial), toroidal moment (t(ime)-odd polar = axio-polar), respectively, and **G**, transforming like the scalar ρ, for the type of axial vector, the best known physical example of which is the order parameter director **n** in the theory of phase transitions of liquid crystals. Other examples of vector **G** can be found in reference [14]. All four types of vector can be taken as order parameter of phase transitions.

TABLE 1 Four types of vector and magnetic limiting point groups corresponding to the characters of the four irreducible representations of the dihedral group $\bar{1}1'$, generated by the space inversion $\bar{1}$ and the time reversal 1' [according to 9,10,14]

E	$\bar{1}$	1'	$\bar{1}'$	Vector basis	Magnetic limiting point group
1	1	1	1	**G**	∞/m1'
1	- 1	1	- 1	**P**	∞mm1'
1	1	- 1	- 1	**M**	∞/mm'm'
1	- 1	- 1	1	**T**	∞/m'mm

By intersecting the ensembles of 31 point groups permitting a spontaneous polarization ^{S}**P**, with those 31 groups allowing a spontaneous magnetization ^{S}**M** (both were first determined by Shuvalov and Belov in 1962 [15]), 13 ones were found to allow both ^{S}**P** and ^{S}**M**. By intersecting the respective ensembles of the "magic trinity" of the 31 groups of ^{S}**P** , ^{S}**M** and that of the 31 "axio-polar" groups, Ascher showed that 9 groups are overlapping and permit all three types of vector[10]. Several ferroelectric/ferromagnetic/ferrotoroidic/ferroelastic boracites have such types of magnetic point group (1, m, m', m'm2')[16].

2. FERROTOROIDICS

Keitsîro Aizu [17] coined the collective term "Ferroic" for ferroelectrics, ferromagnetics and ferroelastics, having in common "Ferro", "-ics" and domains, which can be switched and are giving rise to hysteresis loops. Really akin are only ferroelectrics and ferromagnetics (with the *vectors*

$^{S}\mathbf{P}$ and $^{S}\mathbf{M}$, resp.) which can be characterized by *a single Shubnikov point group*, whereas ferroelastics with the symmetric *2^nd^ rank tensor* spontaneous deformation $^{s}\boldsymbol{\varepsilon}$ have to be characterized by *a pair of point groups* , named "species" [17]. Based on the different terms of the density of stored free enthalpy [Table I of ref. 18], the "ferroic" nomenclature has been enlarged [19,20,21] with a subdivision into "Primary", "Secondary" and "Tertiary" ferroics (see Table 2). Keeping in line with the ferroic nomenclature, it is now tempting to extend the primary ferroics with the introduction of the notion "ferrotoroidics", bearing a spontaneous toroidal moment $^{s}\mathbf{T}$. This has been done by using $^{s}\mathbf{T}$ as an order parameter [22-25] with the nomenclature "Ferrotoroic"[26]. (N.B.: since the ending "oic" is reserved to the collective term "ferroic" only, we propose "ferrotoroidic" instead).
If one would now like to extend the domain switching driving force of ferroelectrics and ferromagnetics (Table 2) to an analogous ferrotoroidic one, the situation is the following:
A toroidal moment can be due both to orbital ordering and spin ordering. Here we are only interested in the spin part of the toroidal moment $^{S}\mathbf{T}$ which has been defined[26,27,28] by the sum over the spins of all spin-bearing particles in the unit cell, with the cell's center as origin:

$$^{S}\mathbf{T} = \tfrac{1}{2}\,\mu_B \sum_{a} \mathbf{r}_a \times \mathbf{S}_a \,,$$

where $\mathbf{S}_a$ stands for the spin moment and $\mathbf{r}_a$ for the radius vector of the magnetic cation-ion "a" in the unit cell. Such a moment $^{S}\mathbf{T}$ has been calculated using non-magnetic and magnetic structural data for the ferrimagnet $Ga_{2-x}Fe_xO_3$ [27,28]. It has been shown [26] that the toroidal moment density $\mathbf{T}$ can play the role of an order parameter with curl $\mathbf{H}$ or electric current $\mathbf{j}$ (including displacement current $(1/c)\ \partial D/\partial t$) serving as the thermodynamically conjugated field for the parameter $\mathbf{T}$, in the same way as the electric field $\mathbf{E}$ and the magnetic field $\mathbf{H}$ play that rôle for the spontaneous polarization and magnetization, respectively. Thus there is a contribution to the density of free enthalpy describing the interaction between the toroidal moment and the magnetic field [26]:

$$\delta F_T = -\ \mathbf{T} \times \mathrm{curl}\ \mathbf{B}$$

i.e. the "driving force" for reversing a toroidal moment will be proportional to that term. It means that we would have to apply a circular magnetic field in the plane perpendicular to the toroidal moment. It has been shown theoretically [29] that aggregates of microscopic particles bearing a magnetic moment (e.g. fixed in a plastic matrix), tend to arrange themselves in the absence of a magnetic field in the minimum energy state which has a toroid geometry. The reversal of their toroid vector is possible by a circular magnetic field (with "vortex" **G** = curl **H**), or better a vortex field plus a static magnetic bias field along the toroid vector direction, allowing to decrease the vortex coercive force G_c. Information storage by this kind of technique has been proposed [29].

However, reversing the spontaneous toroid dipole of a ferrotoroidic crystal by means of such a circular field does not appear possible, because this would require the action of coherent circular fields of the size of the unit cell. Fortunately there exists another more interesting contribution to the free enthalpy, $\sim T_i\ (\mathbf{E}\times\mathbf{H})_i$, in which *the physical meaning of the order parameter* "toroidal moment **T**" *has been identified (up to a constant* λ*) as the antisymmetric component of the magnetoelectric tensor* [30]. In the general case the three components of the vector **T** are proportional to the three components of the antisymmetric part of the magnetoelectric tensor, while the source for the vector **T** is the vector **S** with components $S_i \sim (\mathbf{E}\times\mathbf{H})_i$, in the same way as electric and magnetic field are source vectors for polarization and magnetization, respectively. This latter vector transforms like the spontaneous toroidal moment $^S\mathbf{T}$ and necessitates off-diagonal components of the magnetoelectric tensor. It is consistent that the 31 Shubnikov point groups permitting a spontaneous toroidal moment, do have off-diagonal coefficients, a few of them in addition diagonal ones[37, Fig.2]. Then it should also be permitted to add the driving force $\sim\Delta^S T_i\ S_i$ to the list of primary ferroics (Table 2), but this would be strictly adequate in case of the presence of an antisymmetric part of the magnetoelectric tensor only. Then one might speak of a pure "ferrotoroidic domain", but it would be an antiferromagnetic, necessarily magnetoelectric one and the term would not have a deeper signification than when O'Dell [31] called the antiferromagnetic domains of Cr_2O_3 "magnetoelectric domains". The switching and "poling" of such "ferrotoroidic" domains can be done by magnetoelectric poling and annealing as described for the first time for Cr_2O_3 [32,33], but with crossed electric and magnetic fields at right angle to $^S\mathbf{T}$.

More frequently, however, symmetric *and* antisymmetric components of the "secondary ferroic" magnetoelectric coefficient α_{ij} (Table 2) will occur in materials permitting $^S\mathbf{T}$ and will have to be separated for extracting the antisymmetric part. This has been done for the first time for the polar ferrimagnetic $Ga_{2-x}Fe_xO_3$ (point group m2'm') [27,28] and a high magnetic field-induced spin-flop phase of Cr_2O_3 [34] with monoclinic point group 2'/m [35].

The orthorhombic point group m'm2' of the boracites $Ni_3B_7O_{13}Br$, $Co_3B_7O_{13}Br$ and $Co_3B_7O_{13}I$ allows a spontaneous toroidal moment ${}^S\mathbf{T}$ and has the magnetoelectric coefficients α_{23} and α_{32} . Whereas the temperature dependence of α_{23} reflects that of the sublattice magnetization, α_{32} shows an unusual sharp diverging peak at T_c. By taking ${}^S\mathbf{T}$ as the order parameter, phenomenological theory showed that α_{23} is proportional to ${}^S\mathbf{T}$**,** whereas α_{32} is given by the sum of a term $\propto 1/{}^S\mathbf{T}$ and a second one $\propto {}^S\mathbf{T}$. The term inversely proportional to ${}^S\mathbf{T}$ explains the diverging peak and identifies it herewith as the signature of a spontaneous toroidal moment[25].

3. ELECTROTOROIDIC, MAGNETOTOROIDIC AND PIEZOTOROIDIC EFFECTS

With a view to describing moving crystals, Ascher also determined the 58 Shubnikov point groups for the kineto-electric effect and the 58 groups for the kinetomagnetic effect, both being ruled by a second rank tensor of the same type as that of the magnetoelectric effect [10]. In Table 2 of ref. [10] these groups are arranged on places with corresponding tensor form. The "ferrokinetic", "kineto-electric" and "kinetomagnetic" terms of the density of free enthalpy g were defined respectively as [10]

$$-g\,(\mathbf{E},\mathbf{B},\mathbf{v}) = \ldots + {}^o p_i\, v_i + \eta_{ik}\, v_i\, E_k + \xi_{ik}\, v_i\, c\, B_k \;,$$

where ${}^o\mathbf{p}$ = linear momentum without electric (**E**) and magnetic (**B**) fields, $\mathbf{v}$ = the "field" velocity and c = light velocity. Since ${}^o\mathbf{p}$ and $\mathbf{v}$ transform like the vectors **T** and **S** , we can anticipate on mere symmetry grounds under the heading "secondary ferroics" the existence of analogous "ferrotoroidic", "electrotoroidic" and "magnetotoroidic" terms $\sim T_i\, S_i$, $\sim S_i\, E_k$ and $\sim S_i\, H_k$, respectively. From the explicit forms $\sim(\mathbf{E}\times\mathbf{H})_i\, E_k$ and $\sim(\mathbf{E}\times\mathbf{H})_i\, H_k$ we can identify these effects simply as special cases of the bilinear magnetoelectric effects with the terms $\gamma_{ijk}\, H_i\, E_j\, E_k$ and $\alpha_{ijk}\, E_i\, H_j\, H_k$ [36, 37], respectively, in the same way as ${}^sT_i\, S_i$ was identified as a special case of the linear magnetoelectric effect. In the converse sense the $\sim S_i\, E_k$ and $\sim S_i\, H_k$ terms should also allow to produce electric and magnetic field-induced toroidal moments, respectively.

Since **S** transforms like the current density, the set of secondary ferroic effects may also be enlarged by a "piezotoroidic" effect term $\sim S_i\, \sigma_{jk}$, where σ_{ik} is the stress tensor, in full analogy with the "piezoconductive"

TABLE 2 Ferroic "driving forces" of domain switching and reorientation due to differences in domain states (adapted from references [18,19,20,21])

Type of Ferroic	**"Driving force"** $\Delta g \propto$	**States differ in:**	
Primary Ferroics			
Ferromagnetic	$\Delta^S M_i\, H_i$	spontaneous magnetization	^sM_i
Ferroelectric	$\Delta^S P_i\, E_i$	spontaneous polarization	^sP_i
Ferrotoroidics	$\Delta^S T_i\, S_i$	spontan. toroidal moment	^sT_i
Ferroelastic	$\Delta^S \varepsilon_{ij}\, \sigma_{ij}$	spontaneous deformation	$^s\varepsilon_{ij}$
Secondary ferroics§)			
Ferrobimagnetic	$\Delta\chi_{ij}\, H_i\, H_j$	magnetic susceptibility	χ_{ij}
Ferrobielectric	$\Delta\kappa_{ij}\, E_i\, E_j$	electric susceptibility	κ_{ij}
Ferrobielastic	$\Delta s_{ijkl}\, \sigma_{ij}\, \sigma_{kl}$	elastic compliance	s_{ijkl}
Ferroelastoelectric	$\Delta d_{ijk}\, E_i\, \sigma_{jk}$	piezoelectric coefficient	d_{ijk}
Ferromagnetoelastic	$\Delta q_{ijk}\, H_i\, \sigma_{jk}$	piezomagnetic coefficient	q_{ijk}
Ferromagnetoelectric***)	$\Delta\alpha_{ij}\, E_i\, H_j$	magnetoelectric*) coeff..	α_{ij}
Tertiary ferroics			
Ferro**tri**electricity	$\Delta\kappa_{ijk}\, E_i\, E_j\, E_k$	nonlinear electric suscept.	κ_{ijk}
Ferro**tri**magnetism	$\Delta\chi_{ijk}\, H_i\, H_j\, H_k$	nonlinear magnetic suscept.	χ_{ijk}
Ferroelasto**bi**electricity	$\Delta\gamma_{iljk}\, \sigma_{jk}\, E_i\, E_l$	electrostriction coefficient	γ_{iljk}
Ferroelasto**bi**magnetism	$\Delta\lambda_{iljk}\, \sigma_{jk}\, H_i\, H_l$	magnetostrictioncoefficient	λ_{iljk}
Ferromagneto**bi**electricity	$\Delta\gamma_{ijk}\, H_i\, E_j\, E_k$	magneto**bi**electric**)coeff.	γ_{ijk}
Ferroelectro**bi**magnetism	$\Delta\beta_{ijk}\, E_i\, H_j\, H_k$	electro**bi**magnetic**)coeff.	β_{ijk}
Ferromagnetoelectroelasticity	$\Delta\pi_{ijkl}\, H_i\, E_j\, \sigma_{kl}$	piezomagnetoelectric coeff.	π_{ijkl}
Ferromagneto**bi**elasticity	$\Delta\psi_{iklmn}\, H_i\, \sigma_{kl}\, \sigma_{mn}$	magneto**bi**elastic coeff.	ψ_{iklmn}

*) linear magnetoelectric effect,**) = bilinear magnetoelectric effects, ***) N.B.: in the nomenclature of Newnham the term "Ferromagnetoelectric", used in this table, is somewhat unhappy because it is also sometimes used in literature for a ferroelectric being simultaneously ferro(i)magnetic in the same phase.- If the toroidic domains would have been discovered before the ferromagnetic ones, we might probably have on equal right a long list of Primary, Secondary and Tertiary "Toroido-"s of "Toroics" today!!

§) Since the electrotoroidic and magnetotoroidic terms can be expressed by the magneto**bi**electric and electro**bi**magnetic ones, resp., due to $S_i = (\mathbf{E}\times\mathbf{H})_i$, they have not been specially mentioned in the list of Secondary ferroics.

effect, for which Ascher has determined the 66 Shubnikov point groups and their tensor form [9] (in transposed matrix form also in ref. [38], Fig.1, columns u). Since $S_i\, \sigma_{jk} = \sim(\mathbf{E}\times\mathbf{H})_i\, \sigma_{jk}$, the piezotoroidic effect can be identified as a special case, with condition ($\mathbf{E}\times\mathbf{H}$), of the "tertiary ferroic" piezomagnetoelectric effect [40] with term $\pi_{ijkl}\, H_i\, E_j\, \sigma_{kl}$ in the free enthalpy (TABLE 2). The latter effect has so far not yet been measured [41]. The piezotoroidic effect should allow to induce a toroidal moment by stress or a deformation by crossed electric and magnetic fields. Since we can replace the stress tensor σ_{jk} of the piezotoroidic term by the magnetic and electric susceptibility terms $H_j\, H_k$ and $E_j\, E_k$, resp., without changing the tensor form, we obtain the tertiary ferroic bilinear terms $S_i\, E_j\, E_k$ and $S_i\, H_j\, H_k$, respectively, allowed in the 66

TABLE 3 Tensor form and type of transposed matrix form (Fig.1 of ref. [38]) of some secondary and tertiary ferroic terms of stored free enthalpy

Piezoelectric tensor form (t-type. matrix) :	$E_i \sigma_{jk}$,	$E_i E_j E_k$,	$E_i H_j H_k$
Piezomagnetic tensor form (s-type matrix):	$H_i \sigma_{jk}$,	$H_i E_j E_k$,	$H_i H_j H_k$
Piezotoroidic tensor form (u-type matrix) :	$S_i \sigma_{jk}$,	$S_i E_j E_k$,	$S_i H_j H_k$

piezotoroidic (piezoconductive) point groups (Table 3). Due to $S_i \sim(\mathbf{E}\times\mathbf{H})_i$, the terms are also ruled by the piezomagnetoelectric tensor form. The $S_i E_j E_k$ term should for example give rise to the linear toroido-optic effect (linear birefringence induced by $\sim(\mathbf{E}\times\mathbf{H})$), in analogy with the linear magneto-optic effect term $H_i E_j E_k$ [42] and the linear electro-optic (Pockels-) effect term $E_i E_j E_k$ [18] of the stored free enthalpy.

4. CONCLUSIONS

In the general case the toroidal part of the magnetic spin structure of antiferromagnetic or (weakly) ferro(i)magnetic ferrotoroidics will be rigidly interwoven and coupled, so that "toroidic domains" will be identical with the ferro(i)magnetic or antiferromagnetic ones, in a similar way as fully ferroelectric/fully ferroelastic domains are identical and do not contain sub-domains[39]. The magnetic field-induced switching of ferromagnetic/ferrotoroidic domains will therefore reverse the sign both of the spontaneous toroidal moment and that of spontaneous magnetization. For reversing the sign of the toroidal moment of antiferromagnetic structures, the magnetoelectric driving force $\propto\Delta\alpha_{ij} E_i H_j$ can be used. The possibilities of "reorientation" (= switching of spin directions by angles other than 90°) of spontaneous toroidal moments will require ferroelasticity in the ferroic phase and obey the same rules as established earlier[39] for ferromagnetics, antiferromagnetics, ferroelectrics and ferroelastics.

Since the linear magnetoelectric effect allows to determine the magnetic point group of spin-ordered crystals, it has proved to be a precious complementary tool for magnetic structure determination by neutron diffraction. In the same way, the determination of the toroidal moment *via* the antisymmetric part of the magnetoelectric tensor and the cross-check by its calculation from magnetic and non-magnetic structural data will bring additional help for finalizing magnetic structures accurately.

The postulated secondary ferroic electrotoroidic, magnetotoroidic and piezotoroidic effects, as well as the tertiary ferroic bilinear electro- and magnetotoroidic effects may be difficult to disclose, but since Nature

usually uses all degrees of freedom which symmetry offers her, they may sooner or later become measured.
Thus several physical meanings of the axio-polar (t-odd polar) vector are now well established (anapole moment, current density, velocity, linear momentum, toroidal moment, etc). Nonetheless, this fact is still widely ignored in text-books. For example the current density **j** usually continues to be described as an ordinary polar vector. Although this is wrong, it may lead to correct results in para- and diamagnets, which are left invariant under time reversal. However, this holds no longer true in spin ordered phases, where the axio-polar character of **j** has to be respected for describing physical effects correctly, for example linear magneto-resistance [38],[43].
Summarizing, we can say that the recently evidenced anapole moment of ^{133}Cs and the spontaneous toroidal moment of the single crystals of $Ga_{2-x}Fe_xO_3$ and Cr_2O_3 open fascinating new horizons in nuclear and particle physics on the one hand and in solid state physics and crystallography on the other hand.

Acknowledgements
The author is grateful to Jean-Pierre Rivera for critical advice.

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