Multiferroics and beyond: electric properties of different magnetic textures

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Abstract

This article presents a survey of many nontrivial effects connected with the coupling of electric and magnetic degrees of freedom in solids — the field initiated by I. E. Dzyaloshinskii in 1959. I briefly consider the main physics of multiferroic materials, and concentrate on different effects “beyond multiferroics”, based on the same physical mechanisms which operate in multiferroics. In particular they lead to nontrivial electric properties of different magnetic textures — such as the appearance of dipoles on magnetic monopoles in spin ice, dipoles on some domain walls in the usual ferromagnets, on skyrmions etc. The inverse effect, the appearance of magnetic monopoles on electric charges in magnetoelectrics, is also discussed. This nontrivial electric activity of different magnetic textures has manifestations in many experimental properties of these materials, and it can potentially lead to novel applications.
Electricity and magnetism are two sides of the same physical phenomenon, which one sees for example from Maxwell equations. Their strong interplay is crucial for physics, but also for industry. A new twist in this story is the rapid development of a new field of spintronics [1], [2], which uses not only charge but also spin of electrons in electronic applications. Modern development of this field can be traced back to the seminal works of I. E. Dzyaloshinskii on weak ferromagnetism, introducing the concept of antisymmetric exchange — the Dzyaloshinskii, or Dzyaloshinskii–Moriya (DM) interaction [3], and almost at the same time suggesting the idea of linear magnetoelectric effect [4]. Both these papers strongly influenced the development of magnetism. In particular the second paper gave rise to an enormous development in such fields as magnetoelectrics and multiferroics (MF) [5–13]. And the know-how we got in studying these materials we can apply now to the investigation of many other phenomena dealing with the coupling of electric and magnetic properties of materials — and not only in special magnetoelectric and multiferroic compounds, but also in ordinary magnetic materials, in particular displaying a nontrivial electric activity of different magnetic textures: magnetic domain walls, defects, skyrmions etc. This is the topic of this paper which has partially a character of a review, but also contains some novel material. Thus in the title of this paper, “Multiferroics and beyond” the accent will be made on the second word, “beyond”. It is also noteworthy that in the most recent publications of Dzyaloshinskii he returned to this field [14–17]. The name of Dzyaloshinskii indeed appears in this field over and over again.

MULTIFERROICS; A BIT OF HISTORY

I will start by very briefly describing the developments of investigations in the field of multiferroics, as I see it. As mentioned above, the real activity in this field started after the publication of the seminal paper by Dzyaloshinskii [4], although people sometimes also cite a short sentence from a much older paper by Pierre Curie from 1894 [18], in which he mentioned that in principle one can combine in one material nontrivial electric and magnetic properties. But this was just a general declaration, without any specific physics involved. After the suggestion by Dzyaloshinskii (following a short remark in [19]) that in particular magnetic systems there may exist magnetoelectric effect — electric polarization induced by magnetic field, and the inverse effect — inducing magnetization by electric field, this
effect was very quickly discovered in $\text{Cr}_2\text{O}_3$ by Astrov [20]. Soon after that a rather rapid development followed. And people started to look not only at magnetoelectrics, in which one needs external fields to induce interesting effects, but also at materials which in the ground state, in the absence of external fields, would combine the properties of magnets and ferroelectrics. Such materials were dubbed multiferroics [21]. Besides purely scientific interest, these systems promise very important practical applications — at present the most important being probably the potential possibility to control magnetic memory in computer storage devices electrically, using effects such as gating, etc., i.e. avoiding using electric currents with their dissipation. This is still the main driving force of these investigations.

Especially active search and study of such systems at an early stage of these investigations was carried out in the former Soviet Union, mainly by two groups; by Smolenskii in Leningrad, now St. Petersburg, and by Venevtsev in Moscow, see e.g. [22] and [23]. In these two groups a number of multiferroics were discovered, but the practical use of these was restricted either by low temperatures at which such state existed or by relatively weak coupling between electric and magnetic subsystems — which even now remain the main obstacles to their wide practical use, although enormous progress has been achieved in recent years. But, besides the potential applications, the study of these materials presented some real challenges to the general physics. One of these was an early observation that in one of the biggest classes of materials — perovskites $AB\text{O}_3$, to which quite a lot of magnetic systems belong, including the famous colossal magnetoresistance manganites such as $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, but also most of the interesting and practically important ferroelectrics, starting from $\text{BaTiO}_3$, there exists a striking “mutual exclusion”: materials with transition metals $B$ with partially-filled $d$-shells are magnetic, whereas those with empty $d$-shell, with configuration $d^0$, are of course not magnetic but could become ferroelectric. And surprisingly there was practically no overlap between these two big classes of materials: either these were ferroelectric, or magnetic, but almost never both simultaneously. This $d^{n-}d^0$ dichotomy was noticed long ago, but did not attract attention for quite a while. I remember that at around 1996 I told this story, and in general of the attempts to combine in one material both (ferro)magnetic and ferroelectric properties, to a very good and extremely broad physicist George Sawatzky with whom we have both worked at that time in Groningen University, and his reaction was very characteristic: “But this is very interesting! Why don’t we know anything about it?” Couple of years later at the workshop on quantum magnetism in KITP
in Santa Barbara in 1998 we organized the brainstorming discussion of the topic of a possible coexistence of electricity and magnetism. Nicola Spaldin (at that time Nicola Hill) spoke at this meeting about her *ab initio* calculations of one such “suspicious” material, BiNiO$_3$ [24], and we discussed this empirical observation about mutual exclusion of ferroelectricity (with $d^0$ configuration) and magnetism ($d^n$ configuration).

The next important step happened in 2001 when Nicola organized a special session at the March meeting of American Physical Society, Session C21, devoted to the discussion of multiferroics [25]. One can say that this session actually has lead to the revival of interest in this problem, and “put multiferroics on the map.” And indeed, already in 2007 there were 7 special sessions on multiferroics at the APS March meeting — sessions, not talks! And in 2008 the term “multiferroics” was in the title of 12 sessions at the March meeting. And apparently the start was this first special session at the March meeting in 2001.

But of course the most important was the experimental progress in this field reached a bit later, mainly by three groups. Tsuyoshi Kimura and Yoshinori Tokura found striking MF behaviour in TbMnO$_3$ [26], and Sang-Wook Cheong with coworkers in Tb$_2$Mn$_2$O$_5$ [27]. This was actually the discovery of a novel type of multiferroics which one now calls type-II MF — the systems in which ferroelectricity appears and is driven by a particular type of magnetic ordering, in contrast to type-I multiferroics in which FE and magnetism appear independently and in which most often different subsystems and different ions are responsible for the two orderings. The third breakthrough was the synthesis by the group of Ramesh of thin films of the classical type-I multiferroic BiFeO$_3$ (BFO) [28], till now remaining the system with the best performance and probably the best perspectives for practical applications (if we speak of one material, not of composite systems such as e.g. multilayers of good FE and good ferromagnets). The films made by Ramesh have shown very spectacular properties, with much stronger effects than known until that time on the bulk BiFeO$_3$ (although now people reach such good performance also in the bulk BFO). These three experimental breakthroughs, together with the realization of some fundamental theoretical problems and challenges, have lead to the revival of common interest in multiferroics and to a rapid progress in this field. At present quite a lot of new MF systems are found, and we can probably say that the main physical mechanisms responsible for this phenomenon are already understood, although the constant progress in this field is still ongoing and novel materials and novel phenomena are found. And, besides multiferroics per se, the experience
and know-how we have gained by studying multiferroics can be applied to the discussion of related phenomena even in non-MF materials. In this paper I will try to summarise some of this novel development, although of course I cannot cover this very big field. There exist already quite a few general review articles on multiferroics, a section on multiferroics is included in the book [29], and a special book on these systems is published [30]. I will not discuss here these effects in details, but will rather concentrate on the “spin-offs” of these studies, on a relatively qualitative level, concentrating on what is going on “beyond multiferroics”. But I will start with a short synopsis of the main effects and mechanisms of multiferroics per se.

MULTIFERROICS: BASIC PHYSICS

Generally speaking, we can divide multiferroics into two big groups. The first one, which can be called type-I MF [11], is formed by materials in which magnetism and ferroelectricity appear independently and are due to different mechanisms, different subsystems in a material — although of course they are coupled. In these materials the values of critical temperatures of magnetic and FE transitions can often be quite high, with the FE transition occurring typically at higher temperatures. The best example of these type-I MF is the already-mentioned BiFeO$_3$ with $T_{\text{FE}} = 1100$ K and $T_N = 643$ K, or the hexagonal manganites RMnO$_3$ ($R$ is a rare earth) with $T_{\text{FE}} \sim 1000$ K and $T_N \sim 100$ K. The magnetic and FE degrees of freedom in these systems are of course coupled but this coupling is typically rather weak.

Another group of multiferroics, type-II MF, are those in which ferroelectricity is driven by a particular type of magnetic ordering. To these belong the two multiferroics of this class discovered first, TbMnO$_3$ [26] and TbMn$_2$O$_5$ [27]. The paramagnetic state in these systems is not FE, but a particular type of magnetic ordering can generate FE polarization. It is these new materials which have attracted the main general physical interest and which have lead to the emergence of several novel physical concepts. From a practical point of view these type-II MF may seem more promising — the intrinsic coupling of magnetism and ferroelectricity in those is of course very strong. But unfortunately most of these materials have relatively low values of critical temperatures, below which this coexistence of FE and magnetism is observed.

Two general mechanisms are involved to explain the appearance of electric polarization
in some particular magnetically-ordered states in type-II multiferroics. One of those is the usual magnetostriction: a particular magnetic ordering can break inversion symmetry, and the corresponding magnetostrictive distortion of the lattice in some magnetic structures can induce electric polarization, see e.g. [31]. This mechanism does not require the presence of spin–orbit interaction. Another, more widespread and more interesting mechanism of MF behaviour in type-II MF relies on the use of relativistic spin–orbit coupling and is more in spirit with the original mechanism of magnetoelectricity proposed by Dzyaloshinskii. There are several versions of this mechanism, see e.g. [32, 33]. The most popular and most important for going “beyond multiferroics” is the mechanism of the appearance of electric polarization in magnets with cycloidal magnetic structure. This mechanism was elucidated in a microscopic treatment in the paper of Katsura, Nagaosa and Balatsky [34], and it was obtained using the Landau expansion by Mostovoy [35]. According to this theory, if spins on two neighbouring magnetic ions are not collinear, there would appear for this pair of ions an electric polarization proportional to

$$P_{ij} = cr_{ij} \times [S_i \times S_j]$$  \hspace{1cm} (1)

where $c$ is some coefficient. For cycloidal magnetic structure shown in Fig. II(a), it would give a net polarization, i.e. the cycloidal magnets are intrinsically multiferroic.

The microscopic mechanism of the appearance of electric polarization was disclosed by Sergienko, Sen and Dagotto [36]. It is actually the inverse Dzyaloshinskii effect. According to [3], for some particular symmetries there exists for a pair of ions $i, j$ the antisymmetric
DM (Dzyaloshinskii–Moriya) exchange

\[ H_{\text{DM}} = -D_{ij} \cdot (S_i \times S_j) . \]  

(2)

If the Dzyaloshinskii vector \( D \) is nonzero, this interaction leads to canting of neighbouring spins. But vice versa, if spins are noncollinear for any reason, to gain this energy it may be favourable to distort the lattice and shift the ions so as to make \( D \neq 0 \). In typical cases, e.g. in perovskites, the nn exchange between magnetic ions \( M_i \) and \( M_j \) is carried out by superexchange via ligand (e.g. oxygen ions) sitting in between, Fig. 1(b). If such oxygen sits exactly at the middle of the \((ij)\) bond, by symmetry the DM interaction is zero \[37\]. To gain the DM energy (2) it is then better to shift such oxygen by some distance \( \delta \) perpendicular to the \((ij)\) bond, e.g. in the \( z \)-direction, Fig. 1(b). Then there would appear a nonzero DM interaction with the vector \( D \sim r_{ij} \times \delta \), so that now we gain the DM energy (2). But such shift of negatively charged oxygen ions away from the “centre of gravity” of positive charges of TM ions \( M_i \) and \( M_j \) would create an electric dipole, or a polarization in the \( z \)-direction. This is the inverse Dzyaloshinskii effect — the mechanism of the appearance of MF behaviour in many MF systems, and it is this mechanism which could also lead to an electric polarization of different magnetic textures — some domain walls, skyrmions, etc., which I will extensively “exploit” below.

**ELECTRONIC MECHANISMS OF COUPLING OF ELECTRICITY AND MAGNETISM**

Usually the appearance of electric polarization in magnetoelectrics and multiferroics is attributed to lattice effects, to the corresponding shifts of ions. However there also exists a purely electronic mechanism which can lead to such coupling. In particular this mechanism can operate in frustrated systems, the basic building blocks of which are triangles made of transition metal (TM) ions, Fig. 2. If one describes \( d \)-electrons in such a triangle by the usual Hubbard model,

\[ H = -t \sum_{\langle ij \rangle, \sigma} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} , \]  

(3)

one can show that for certain magnetic textures, for certain spin correlation functions, there may appear charge redistribution in such triangles, so that in contrast to the standard picture the charge at site \( i \) is not exactly 1 as in the usual Mott insulators, but can be more...
FIG. 2. The appearance of electric polarization in triangles with a particular spin pattern. The oval in (b) is the singlet state of spins $S_2, S_3$, with $S_2 + S_3 = 0$ and $S_2 \cdot S_3 = -\frac{3}{4}$, cf. Eq. (4).

or less than 1. Correspondingly, there would appear in this situation an electric dipole in such a triangle, the magnitude and direction of which is determined by the spin structure. That is, it is a purely electronic mechanism of multiferroic behaviour.

The treatment carried out in [38] (see also [39]) shows that when one treats the model (3) (for one electron per site and for strong correlations $t/U \ll 1$) to third order in $(t/U)$, the charge at site 1 is given by

$$n_1 \sim 1 + K \left[ S_1 \cdot (S_2 + S_3) - 2S_2 \cdot S_3 \right], \quad K = 8t^3/U^2$$

and similar expressions for $n_2, n_3$. I.e. if this spin correlation function entering (4) is nonzero, there would occur charge redistribution in such a triangle, and there would appear in it an electric dipole moment, see Fig 2. It seems that the effect here is rather small, $\sim t^3/U^2 \ll 1$.

But one can show that the same expression would also be valid in case when $t \sim U$, only with a different value for the coefficient $K$ in (4). Thus in general this purely electronic mechanism of the generation of electronic polarization by a particular magnetic texture may be quite significant. (Actually the contribution of lattice distortions would lead to the same expression for polarization in this case, with the same dependence on the spin structure [39].)

Interestingly enough, in the same situation there would appear for certain spin textures also spontaneous circular currents in such triangles, with the respective orbital moment. These currents exist for noncoplanar spins, and they are given by the expression [38]

$$j_{123} = C\kappa(123)$$

where $\kappa$ is the scalar spin chirality,

$$\kappa(123) = S_1 \cdot (S_2 \times S_3)$$
Here the coefficient $C$ in the nondegenerate Hubbard model is given by $C = 24et^3/hU^2$. Thus such noncoplanar spin texture will lead not only to the fictitious magnetic field due to Berry phase [40], but also to real orbital currents and orbital moments, proportional to scalar spin chirality (6). We will see the examples of this effect below.

The physical mechanisms described in this and in the previous sections can be used to predict or explain not only some features of multiferroics, but also some phenomena connected with the interplay between electric and magnetic degrees of freedom in other situations. This will be discussed in the following sections.

**DIPOLES ON MONOPOLES IN SPIN ICE**

One very interesting phenomenon, discovered recently in frustrated systems, is the appearance in some of them (so called spin ice systems) of excitations having the properties of magnetic monopoles [41, 42]. After this proposal was made theoretically, such monopoles were discovered and extensively studied experimentally, see e.g. [43–45]. Such monopoles were first predicted and observed in pyrochlore spin ice such as Tb$_2$Ti$_2$O$_7$, in which the main building blocks are metal (here Tb) tetrahedra with strong Ising ions, with moments pointing towards (or away from) the centre of tetrahedra, Fig. 3. In the usual spin ice ground state we have the situation with (2-in)–(2-out) state at each tetrahedron; such spin distribution is not unique and is highly frustrated. The monopole configuration which for the usual spin ice is an excited state, which can be excited at finite temperature but which can also be stabilised by the external magnetic field, corresponds to the (3-in)–(1-out) state (monopole $\mu$ with the magnetic charge inside a tetrahedron $+2Q$, where each spin is represented by a pair of magnetic charges $(+Q, -Q)$), or (1-in)–(3-out) state (antimonopole $\bar{\mu}$, with the charge $-2Q$). Such monopoles and antimonopoles can move in pyrochlore spin ice by flipping some spins and leaving a trail of reversed spins, but due to spin disorder inherent in spin ice state such strings have no tension (the energy does not increase linearly with the length of the string, as it would in a long-range ordered state), i.e. there is no confinement, and such monopoles and antimonopoles can live in a crystal as independent excitations.

This concerns the magnetic degrees of freedom of these systems. But using the treatment presented in the previous section, see Eq. (4), one can show that there would exist in this case electric dipoles attached to each magnetic monopoles [46]. Indeed, using Eq. (4) one
FIG. 3. The appearance of electric dipoles (thick green arrows) for monopoles (a) and antimonopoles (b) in metal tetrahedra — the building blocks of spin ice pyrochlores.

can see that there would be no net dipoles for the standard (2-in)–(2-out) states of spin ice (and neither for (4-in) or (4-out) states). But from this expression we immediately see that there would be a nonzero dipole (thick green arrow) in monopoles and antimonopoles, directed towards the “special” spin (red spin in Fig 3 — the out-spin in monopole, in-spin in antimonopole). (As Eq. (4) is even in spins, the reversal of all spins in going from monopole to antimonopole leaves the dipole unchanged.) Such electric dipoles at each magnetic monopole in the usual spin ice would be random and dynamic, and they would give extra electric activity in the state with monopoles. These dipoles and their consequences were indeed observed experimentally in Dy$_2$Ti$_2$O$_7$ and Tb$_2$TiO$_7$ [47, 48]. In a strong magnetic field $H \parallel [111]$ there would appear ordered monopolies and antimonopolies at every site in spin ice pyrochlore, and correspondingly there would be also dipole moments at every tetrahedron, ordered in an antiferroelectric fashion [46].

A similar effect should also exist in kagome spin ice, Fig. 4. In contrast to pyrochlores, here monopole ((2-in)–(1-out)) or antimonopole ((1-in)–(2-out)) configurations would exist at each triangle already in the ground state. Consequently there would exist, according to Eq. (4), electric dipoles at every triangle. Again, in the real spin ice ground state the spin configurations, i.e. in this case monopoles and antimonopoles, would be random, and with them the dipoles would also be random and fluctuating.

Using Eqs. (5), (6) one can show that there also exist spontaneous currents and orbital moments at magnetic monopoles in pyrochlores spin ice, Fig 5. These are random and dynamic in spin ice state with excited monopoles. (Note that, in contrast to dipoles, currents also exist in the usual (2-in)–(2-out) tetrahedra.)
FIG. 4. The appearance of an electric dipole of a magnetic triangle — the building block of kagome spin ice systems.

FIG. 5. Spontaneous current (thin circular arrow) and the resulting orbital moment (thick blue arrow) at a magnetic monopole in pyrochlore spin ice. As the expression (6) is odd in spins, currents and orbital moments in antimonopoles are opposite to those on monopoles, in contrast to electric dipoles

MOMENT FRAGMENTATION AND DIPOLES

An interesting twist in this story is the recent suggestion [49] that there could exist in spin ice systems a novel state — the state with spin, or magnetic moment fragmentation. This idea relies on the Helmholtz decomposition of magnetization into a divergence-free and divergence-full components, the later actually describing the distribution of magnetic monopoles — the sources of magnetic field in a system.

\[ M(r) = M_{\text{mono}} + M_{\text{ice}} = -\nabla \rho + \text{curl} \, A \]  

(7)

where \( \rho(r) \) is the density of magnetic charges ("monopoles"), and the second part is the divergence-free part of magnetization, corresponding to pure spin-ice configuration (with (2-in)–(2-out) states at every tetrahedron, with zero total magnetic charge inside every tetrahedron). (The decomposition (7) of course does not violate the Maxwell equations, in
particular $\text{div}\, B = 0$ as $B = H + 4\pi M$, and $\text{div}\, H = -4\pi \text{div}\, M$, see e.g. [50].) Here the most interesting feature is that there may exist a nontrivial partially-ordered state: the state in which monopoles and antimonopoles exist and are perfectly ordered in the ground state, whereas spins themselves are still disordered, see Fig. 6 for the kagome spin ice with moment fragmentation [51, 52].

A question arises, what would become with electric activity with dipoles in such a state. One can show [53] that dipoles would still exist in such states, but they would not be free independent dipoles but, rather, they would always be paired into $(d, -d)$ pairs, Fig. 6. That is, the transition to such moment fragmentation state would lead to the reduction of electric activity, e.g. microwave absorption, in such a state. The spontaneous currents and the corresponding orbital moments on monopoles are also paired, in this case in $(L, L)$ pairs with parallel orbital moments $L$ [53].

A very special feature of moment fragmentation state in spin ice is the coexistence in one spin system of both long-range ordered (monopoles) and disordered (spin themselves) components. This unusual situation determines also the unusual properties of defects and domain walls in these systems. Each time we have an ordered state we have to think of defects or excitations breaking this perfect order, and also of creation of domains and do-
FIG. 7. Electric dipoles on defects in moment fragmentation state in kagome spin ice (the situation in pyrochlores is similar [53]). (a) A typical situation for defects and domain walls: two neighbouring triangles both with monopoles, showing that at least one of them would have an unpaired dipole. (b) The “supermonopole” defect (3-in triangle). By recommuting spins one can get rid of unpaired dipoles. (c) The appearance of unpaired dipoles at a monopole in place of antimonopole. (d) One type of domain wall in monopole ordered state, showing the appearance of unpaired dipoles on it.
main walls \[54\]. The situation is of course the same here. We can create different types of point defects. These are for example the “reversed” monopole, i.e. monopole in place of antimonopole, or novel excitations, such as the state with (3-in) (“supermonopole”, magnetic charge $3Q$ inside a triangle) state in kagome ice, in which there are monopoles and antimonopoles (charges $\pm Q$) at every triangle, ordered in moment fragmentation state. One can also form domain walls in $\mu - \bar{\mu}$ two-sublattice ordered state (such as domain wall in an antiferromagnet). And all these defects or textures in the ordered monopole sector coexist with the still preserved spin disorder!

One can show that such defects or domain walls would also modify electric properties. At first glance it may seem that every such defect would lead to the creation of free dipoles: dipoles are paired into $(d, -d)$ pairs in the fragmentation state, and defects would remove one dipole for the pair, leaving another dipole unpaired. But actually it is not always the case. Thus the “supermonopole” ((3-in) state in kagome ice, or the (4-in) tetrahedron in a pyrochlore) would not create such free dipoles: using the remaining freedom of spins one can “recommute” those in such way that free dipoles are removed, Fig. 7(b). But other types of defects, e.g. $\mu$ in place of $\bar{\mu}$, would necessarily lead to the creation of unpaired dipoles (even three of them in kagome ice with spin fragmentation, Fig. 7(c) and four in moment fragmentation pyrochlores). A typical configuration of such defects is the neighbouring pair of two monopoles, see Fig. 7(a). One sees that in such situation there would be at least one unpaired dipole. This leads to the unpaired dipoles at the defect in Fig. 7(b). Also different types of domain walls would have unpaired dipoles, see e.g. Fig. 7(d). This probably can be used to control, orient and move such defects or domain walls by (inhomogeneous) electric field, which could potentially be useful for some applications.

**Dipoles at Magnetic Textures in Ordinary Magnets: Domain Walls, Skyrmions and All That**

We have seen in treating multiferroics that there should appear electric dipoles or polarization for certain spin configurations, e.g. at spin cycloids, Fig. 11(a). But the same local spin configuration can exist in many other situation, e.g. at the Néel domain walls in ordinary ferromagnets, Fig. 8. The magnetic structure of such domain wall can be visualised as part of a cycloid, i.e. according to the expression \[11\] it should also have nonzero electric...
polarization [35]. Consequently one could think of influencing such domain walls by electric field. This idea was proposed by Dzyaloshinskii in [14]. But already before that the team in Moscow University had the same idea, and they carried out corresponding experiments [55]. Using films of the standard magnetic garnet — a good insulating ferromagnet with $T_c$ above room temperature — the authors of [55] managed to see the motion of Néel domain walls when they applied an inhomogeneous electric field to the sample (created simply by applying a voltage pulse to a sharpened copper wire close to the film), see Fig. 9: the electric dipoles existing at Néel domain walls were attracted to the region of stronger electric field close to the tip. This is indeed a conceptually very simple but beautiful experiment confirming the main physical ideas developed first in treating multiferroics but which can be now applied to many other situations.

Yet another experiment which can be explained by the same physics — the coupling of magnetic and electric degrees of freedom at particular magnetic textures — is the observation of the creation of spiral magnetic structures at thin magnetic layers on the surface of nonmagnetic metals. It was discovered by the group of Wiesendanger in Hamburg that when one makes a monolayer of Mn on top of W, the magnetic structure of Mn becomes actually cycloidal instead of the expected collinear structure [56]. This effect was explained in [57] using microscopic treatment with the inclusion of the Dzyaloshinskii–Moriya interaction which should be present at the surface (the surface, of course, breaks inversion symmetry). But this effect can be explained very simply in the same picture of the appearance of electric polarization in cycloidal structures. As is shown in Fig. 10(a) the cycloidal structure leads to the creation of electric polarization directed in the plane of the cycloid and perpendicular to its direction. But, inversely, if there exists in a system an intrinsic polarization, or intrinsic electric field, it would lead to the creation of a cycloidal magnetic structure. Such electric
FIG. 9. The scheme of the experiment of Ref. [55] illustrating the motion of the Néel domain wall in an ordinary insulating ferromagnet when a voltage pulse is applied to a metallic tip, creating an inhomogeneous electric field in the sample. Red arrows are spins, thick green arrows are electric dipoles at the domain wall.

Field always exists at the surface of a metal — there exists there a double layer, or potential drop (the work function) and the corresponding electric field perpendicular to the layer. Consequently one may expect that instead of a collinear magnetic structure there would appear in this case a cycloidal structure with a particular sense of spin rotations. This is exactly what was observed in [56]. Such spin rotation works against spin anisotropy, thus if there exists too strong anisotropy in the film, e.g. an easy plane (which is often the case in magnetic films), this anisotropy can suppress the creation of such cycloids. But if such anisotropy is not too strong, cycloids can form in such situations. And, besides magnetic spirals, one can also stabilize magnetic skyrmions in similar situations. This was observed by the same group in a double layer of Fe on Ir [58].

Yet another interesting effect, connected with the same physics, was observed by the Hamburg group when they applied an inhomogeneous electric field to a system. In spirit this experiment is similar to the experiment of the MSU group described above. The authors of [59] (see also [60]) found that they can create skyrmions under the tip with electric voltage. And such skyrmions were created for one polarity of the field, but not for the other. Again,
FIG. 10. Electric dipoles at skyrmions. Spins are assumed to point up in the bulk and down at the centre of the skyrmion. In approaching the centre the spins (red arrows) are rotating, and we show the spin direction at the “middle” ring where the spins lie in the $xy$-plane. (a) Bloch skyrmion. (b) Néel skyrmion. In both cases, according to Eq. (1), there would exist electric dipoles directed radially (thick green arrows).

at least qualitatively one can explain this observation by the same physics as described above. There exist two types of skyrmions: the ones in which spins are rotating as in Bloch domain walls, so that in the middle of the skyrmion the spins are oriented along the circle, Fig. (a), and Néel skyrmions, in which the spins at the “middle” circle point away (or towards) the centre, Fig. (b). Using the expression (1) one can show that there would appear local electric dipoles in such textures: in both cases the dipoles will be directed radially, the green arrows in Fig. (For Néel skyrmions there would also appear net polarization perpendicular to the skyrmion plane.) When we apply voltage to the tip, we create an inhomogeneous electric field. Its radial component would interact with the radial dipoles at the skyrmions and, depending on the polarity, it would lead either to energy gain or to energy loss. Therefore one could indeed expect that the field of one polarity would stabilize the skyrmions under the tip, and the opposite polarity would work against it. This simple picture can thus explain experimental observation of.
DIPOLES AT SPIN WAVES

The same picture as used in the previous section can also predict a nontrivial effect even for ordinary spin waves in ferromagnets. Long ago Bogolyubov posed the question [62]: is there any nontrivial electric effect connected with spin waves? He actually considered what later on became known as the Hubbard model and treated it in perturbation theory, anticipating in particular the much later treatment of superexchange by Anderson, Goodenough and others. And he asked a question whether (when treated not in a purely magnetic models but going back to original electronic description) magnons can carry small electric charge. In a sense the treatment in [38] is in spirit similar to this old approach of Bogolyubov. His conclusion was somewhat ambiguous: actually he did not obtain any real current carried out by magnons, but the expressions equivalent to the currents due to spin chirality were in fact contained in his results. Recently Morimoto and Nagaosa [63] addressed the same question but for a specific situation with shift currents in multiferroics, and obtained that in particular situations there may indeed exist nontrivial electric effects on magnons.

Using the physics described above, in particular the same expression (1) for the electric polarization for canted spins, one can give the arguments that the ordinary spin waves e.g. in ferromagnets will have electric activity, however they will carry not electric charge, but electric dipoles [11]. Indeed the quasiclassical picture of a magnon is that shown in Fig. 10: in a magnon the spins are somewhat tilted away from the average magnetization direction $z$ and precess around it, so that the snapshot is that shown in Fig. 11. And in a spin wave this spin structure “moves” with a certain velocity.

When we look at the structure of Fig. 11 we immediately realize that whereas the $z$-component of magnetization is constant (and is just slightly less than $M_{\text{max}}$), the perpendicular $xy$-components of magnetization form exactly the cycloid of Fig. 11. Therefore, by
the same expression \( \text{(1)} \) we should expect that there would appear electric dipoles at the usual spin waves, shown in Fig. \( \text{11} \). And if we make a spin wave packet, then when it moves in a sample it would carry with it not only magnetization, but also a perpendicular electric dipole. I do not have a good idea could be the experimental consequences of the presence of such dipoles; maybe they can contribute, for example, to Raman scattering on magnons, changing selection rules, or could lead to some other similar effects.

**MONOPOLES ON CHARGES IN MAGNETOELECTRICS**

In the previous sections we have discussed several situations in which particular magnetic textures generate electric dipoles or currents. There is however also an opposite effect — in certain cases electric charges can generate magnetic response, and in particular lead to the creation of magnetic monopoles \([64], [16]\). This is the situation in magnetoelectrics — the field initiated by Dzyaloshinskii in 1959. Indeed if we have a magnetoelectric with diagonal ME tensor \( \alpha_{ij} \), the charge placed in such material will create a radial electric field, but due to the magnetoelectric effect there will also appear magnetization

\[
M_i = \sum_j \alpha_{ij} E_j
\]

For the diagonal magnetoelectric tensor \( \alpha_{ij} \) this magnetization will also be radial and have the shape of an ellipsoid. By the same Helmholtz decomposition \( \text{(7)} \) we see that it will have a divergence-full component (the spherical part of moments, shown in Fig. \( \text{12} \)) and a divergence-free quadrupole-like component. And the first one is equivalent to having magnetic monopole at the position of a probe charge.

The existence of such monopoles can lead to several nontrivial consequences, in particular in the transport properties of such systems \([64]\). One can also have measurable effects when one places and moves charges above the surface of such magnetoelectrics. Image charge created in this case inside the magnetoelectric would create a magnetic monopole, the magnetic field of which outside the sample can be measured experimentally. Such measurements indeed confirmed this picture \( \text{[17]} \) — probably the latest paper in which I. Dzyaloshinskii is a coauthor. I suppose this story will be described in details in another article in this volume, by N. A. Spaldin.
FIG. 12. The appearance of magnetic monopole (radial magnetic field) at a charge in a magneto-electric material (with nonzero diagonal components of the magneto-electric tensor $\alpha_{ij}$).

In conclusion, we can say that there exist indeed diverse and very interesting electric effects at different magnetic textures. This is yet another manifestation of the nontrivial interplay of magnetic and electric degrees of freedom in solids — the field pioneered by Dzyaloshinskii more than 50 years ago, and which still produces more and more new surprises.

ACKNOWLEDGEMENTS

I am grateful to many people with whom I collaborated over many years on the topics discussed in this article. This work was supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Project number 277146847 – CRC 1238

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