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APPLICATION

Applications of magnetoelectrics

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A short review of the development of magnetoelectrics is presented, primarily of recent work (2009–2012). Emphasis is upon the different applications for different generic types: for example, single-phase thin films for magnetic tunnel junctions but thick sandwich-structure multilayers and nano-composites for weak magnetic field detectors.

I. Introduction

The development of magnetoelectric materials and devices is in part an outgrowth of the development of high-quality ferroelectric thin films (Fig. 1). In the early 1970s Esaki pointed out[1] that ferroelectrics would make the ideal tunnel junction, with different transport characteristics useful for storing information in a memory device. Esaki's basic tunneling device has no memory without a ferroelectric layer. However, tunneling generally requires[1] a thickness d of <10 nm, and the conventional and heavily cited (but totally wrong) opinion in the science community (Basceri *et al.*,[2] Uchino[3]) was that ferroelectric thin films lost their ferroelectricity at thicknesses somewhat below a micron, due to depolarization fields at their surfaces. In fact, this loss of ferroelectric properties in thin films turned out to be due to non-stoichiometry in thin films, arising from such things as near-surface oxygen vacancies. Progress in quantifying this was led by work by scientists including Dawber,[4] Junquera and Ghosez[5] and most importantly, the group of Gregg in Belfast.[6–8] We now understand that in a typical perovskite oxide ferroelectric the polarization remains stable down to d = 2.4 nm, about 6 unit cells. However, it is not an exaggeration to point out that the incorrect interpretation of thin-film data delayed the development of ferroelectric tunnel junctions by a decade or more.

The initial goals were to switch magnetization M *via* an applied electric field E, and to switch polarization P with an applied magnetic field B (or H). Further, these switched states should remain (a "persistent" switch, not a transient), and be fast.[13a] Typical results are shown in Fig. 2 (direct P *vs.* H effect); Fig. 3 converse M *vs.* E effect; Fig. 4 switching for single-crystal bismuth ferrite.

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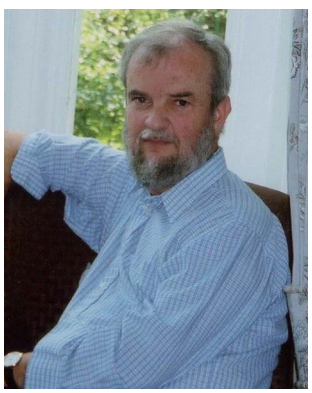

James F. Scott

James F. Scott received his degrees in physics from Harvard (A. B., 1963) and Ohio State (Ph.D., 1966). Best known for his work on ferroelectric thin films, he has coauthored 800 journal publications and five books. Since 1999 he has been professor of ferroics and director of research at the Cavendish Laboratory, Cambridge University. In 2008 he was awarded the MRS Medal. Other prizes include a Humboldt Prize, the Jozef Stefan medal, and a Monkasho Award. He is a Fellow of the Royal Society and a member of the National Academy of Sciences of Slovenia.

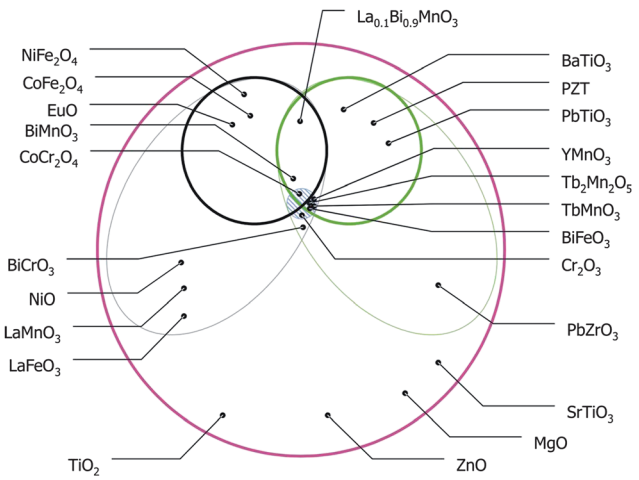

Fig. 1 Venn diagram for magnetoelectric and multiferroics after Eerenstein *et al.* and with representative oxides shown (Bibes *et al.*[11a]). The upper left circle contains ferromagnetic materials, whereas the upper right has ferroelectric materials; the overlap is multiferroic, which is neither necessary nor sufficient for magnetoelectricity (blue cross-hatched region). The larger circle contains antiferromagnets, antiferroelectrics, ferrimagnets, and in general magnetically or electrically ordered crystals (ellipses).

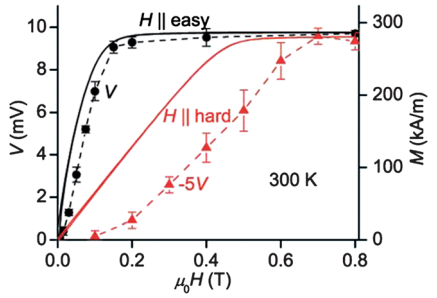

Fig. 2 Changes for multilayer superlattices in polarization with magnetic field P(H).[13a]

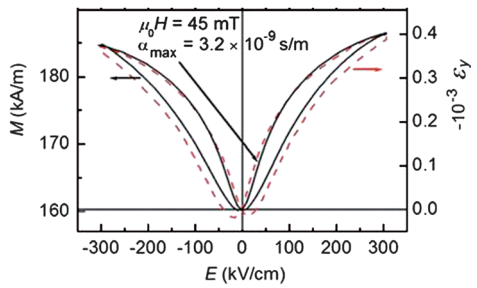

Fig. 3 Changes of magnetization M(E) with electric field.[13c]

II. Generic types of magnetoelectrics

A. Single-phase crystals

1) Multiferroics. There are at least four generic kinds of magnetoelectric materials. Of greatest physics interest are single-phase multiferroic crystals, in which the ferroelectricity and ferromagnetism are created from the same structural arrangement. Sometimes this is from the same ion (*e.g.*, Fe^{+3}) and sometimes the ferroelectricity arises primarily from one ion (*e.g.*, the unpaired electron in Pb or Bi), while the magnetism arises from a second ion (*e.g.*, Fe in $BiFeO_3$). For memory device purposes it is desirable that the materials be highly insulating, function at room temperature, and have a large switched charge (remanent polarization >1 μC cm^{-2}). Most multiferroics do not meet these requirements. Usually the operating temperature is too low and the conductivity is too high. In addition to bismuth ferrite, CuO functions up to 230 K[1e,f] but has very small polarization (too small to be read by a sense amplifier for a switched capacitor random access memory RAM); similarly, $Sr_3Co_2F_{24}O_{41}$ is multiferroic at room temperature under small magnetic bias fields,[1g] but its polarization is also very small. Single-phase mixtures of $PbFe_{1/2}Ta_{1/2}O_3$ and $PbZr_xTi_{1-x}O_3$ work well, however.[1h]

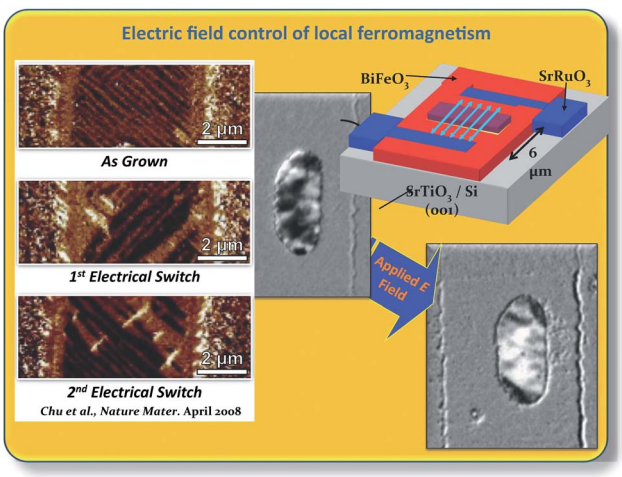

Fig. 4 Switching in single-crystal bismuth ferrite (Ramesh *et al.*[16i]).

2) Non-multiferroics. In addition to multiferroics (we use the term to designate crystals with ferroelectric and ferromagnetic ordering), there are magnetoelectrics which are antiferroelectric and/or antiferromagnetic (Fig. 1). Magnetoelectricity is defined simply as a bilinear term $\alpha_{ij} P_i M_j$ in the free energy, where P is the polarization tensor and M, the magnetization. It is neither necessary nor sufficient for magnetoelectrics to be multiferroic (for example, they can be antiferroelectric and antiferromagnetic). The first magnetoelectric discovered was in fact chromia, Cr_2O_3, which is neither ferromagnetic nor ferroelectric.[9] The main advantage of chromia as a magnetoelectric device is its operating temperature, which extends above 40 °C. However, no one has yet produced a commercial device from this material, despite recent efforts from Duisburg[9b,c] with industry support. There are only a few other non-multiferroic magnetoelectrics: The paramagnetic ferroelectric $Tb_2(MoO_4)_3$ is magnetoelectric but not multiferroic. Its behavior is discussed by Eerenstein *et al.*[1i] And the hexaferrite $Ba_{0.5}Br_{1.5}Zn_2Fe_{12}O_{22}$ has a polarization P induced by an applied magnetization M.

Conversely, there are multiferroics that are not magnetoelectric. A good example is $BaCoF_4$, which has **P** and **M** both along the a-axis, but no diagonal α_{aa} term that can couple them.[1j]

3) Applications for single-crystal magnetoelectrics. The greatest effort in technology transfer for single-crystal magnetoelectrics has come from France, with magnetic tunnel junctions that can be controlled by electric fields. The Thales group of Gajek and Bea,[10] Barthelemy,[10b] Bibes[11] has been extremely productive, with frequent collaborations with Cambridge (Mathur[13]), as has the group of Cheong at Rutgers.[12] Tunnel junction results for Fe/$BaTiO_3$/LSMO are shown in Fig. 5.

The present state of play is to try to make more highly insulating single-phase magnetoelectrics, with operating temperatures above ambient (to meet both military and automotive specifications), and with larger magnetization (all room-temperature materials examined thus far are weak canted ferromagnets or weak cluster ferromagnets, and switched M is too small). For a magnetoelectric RAM, one would like to combine the ultrafast (250 ps) electrical WRITE operation with the non-destructive (no reset) magnetic READ operation, thus combining the best qualities of FRAM and MRAM. But thus far, the magnetization is too small for effective READ.

B. Laminar sandwiches (bilayers or superlattices; *e.g.*, terphenyl-d and PZT)

Unlike the work on single-crystal magnetoelectrics, the discovery of suitable materials for composite devices is not a challenge:

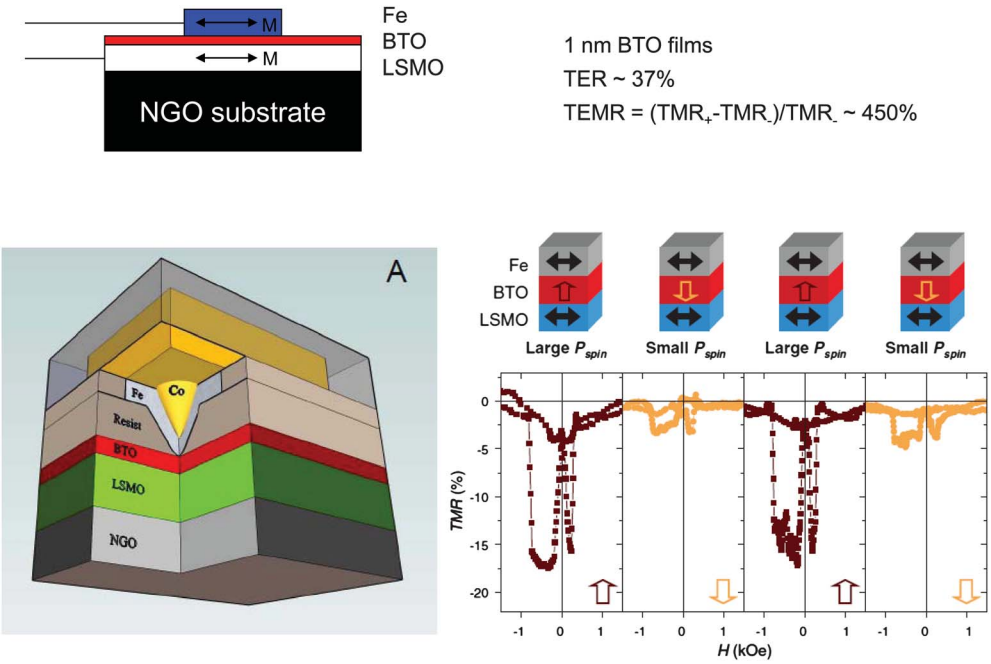

Fig. 5 Magnetoelectric tunnel junction (Mathur *et al.*). Ferroelectric voltage control of spin polarization, with 1 nm barium titanate films with a 450% change in tunneling electrical resistance.[11e]

Placed in close proximity, a strongly magnetostrictive material and a strong (poled) piezoelectric will provide a voltage output when a magnetic field is present and a magnetization in the presence of an electric field. Thus in this case the problem is engineering design and materials science optimization, rather than pure physics.

The main application is for SQUID (superconducting quantum interference device) replacement for weak field detection. Three main groups have pioneered this field and its commercial technology transfer: Viehland[14] at Virginia Tech.; Srinivasan[15] at Oakland University in Michigan; and Fetisov[16] at MIREA in Moscow. The most common materials used are probably Terphenol-D as the magnetostrictor and lead zirconate-titanate (PZT) as the ferroelectric. However metalic glasses ("metglas") and galphenol are also used. Fig. 6–8 illustrate the metglas piezofiber structure, the magnetoelectric coefficient α *versus* H for this structure, and the noise levels (10^{-12} $THz^{-1/2}$) achieved.[14a]

Typical structure is illustrated in Fig. 9 for laminar sandwiches.[16i] The idea is to make very weak magnetic field detectors which do not require cryogenic cooling. Sub-nanotesla sensitivity has been achieved by using ac resonant frequency techniques, but dc detection is also possible. The initial applications are for systems that must be robust. Military applications, such as magnetic mines in harbors are a probable priority, but portable magnetic detectors for geological field work might also be practical outcomes. Medical applications to MRI imaging systems are a longer term but potentially very important area.

There is also a bit of serendipity in this field. About a decade ago the multilayer ceramic industry moved from Ag-Pd electrodes to base metal electrodes (generally Ni) just to save costs, as Pd became very expensive. Of course Ni is magnetic, and the resulting multilayer capacitors are unintentionally magnetoelectric devices.[17] They can be >200 layers, are smaller than a small coin, and cost about US$0.01 each (Fig. 9). At Cambridge we use them for first-year undergraduate laboratory classes, demonstrating piezoelectricity (with tweezers), pyroelectricity (with hot plates), and magnetoelectricity (with bar magnets). These $BaTiO_3$:Ni devices are printed from "ink" solutions in a manner similar to making postage stamps and then furnace-annealed. Many corporations, including AVX,

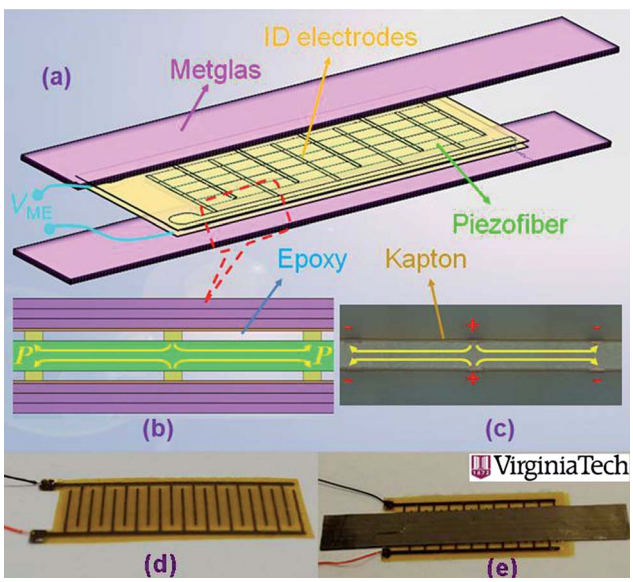

Fig. 6 Metglas on piezofibers,[14a] consisting of an interdigital electrode/PMN-PT core composite, and symmetric 3-layer metglas actuators on the bottom and top of the core composite.

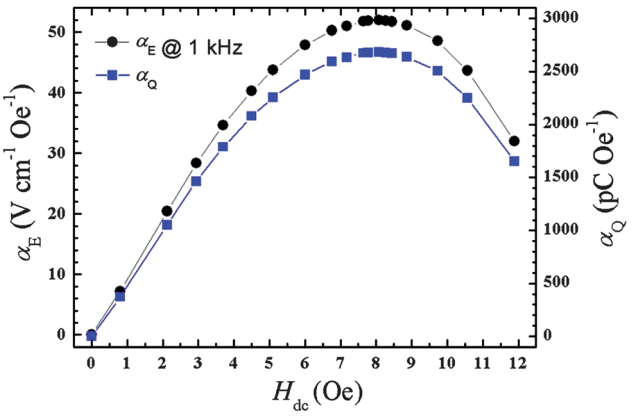

Fig. 7 The magnetic field dependence coefficient of a Metglas/piezofiber sensor: α *versus* H for this metglas system, and the magnetoelectric charge.[14a]

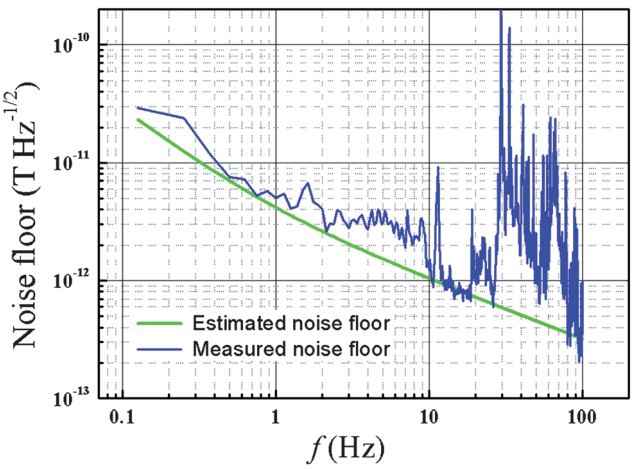

Fig. 8 Measured and estimated equivalent magnetic noise of a Metglas/piezofiber, showing the noise levels achieved.[14a]

Fig. 9 One-cent magnetoelectric multilayer capacitor (Israel *et al.*): photograph, emphasizing size and cost (both M US$0.01).[17a]

TDK, and Kyocera produce them in volumes of hundreds of millions. A clever scheme of contacts[13c] has produced a temperature-independent device in which the pyroelectric voltage in two halves of the device is canceled out, but the magnetoelectric voltages add (Fig. 10).

Magnetic detectors are not the only applications actively investigated: Voltage-driven phase shifters for the 5–20 GHz and 100 GHz–1 THz regime are also attractive.[18] These phase shifters have two important requirements: First, they must have a large frequency shift with applied voltage; and second, their insertion losses must be very small. The insertion loss has been the biggest technology-transfer problem. The main application may be for phased-array radar, where thin film phase shifters will be smaller and weigh less than conventional devices. Fig. 11 shows a multiferroic band-pass filter. The schematic diagram shows a magneto-electric microwave band-pass filter consisting of a bilayer of yttrium iron garnet (YIG) film on a gadolinium gallium garnet (GGG) substrate and lead zirconate titanate (PZT) in a micro-stripline device structure. The input power is coupled to the output under ferromagnetic resonance (FMR) as shown in Fig. 12.Traditional YIG filters are tuned with a magnetic field. But with the YIG-PZT composite in the device, an electric applied to PZT produces a mechanical strain that manifests as an induced magnetic field in YIG, leading to tuning of FMR and the pass-band as shown.

Fig. 13 shows a multiferroic microwave phase shifter: The diagram shows a stripline phase shifter with a YIG-PZT bilayer. A static magnetic field applied to the device biases the device close to FMR in YIG. An electric field applied to PZT produces a strain induced magnetic field in YIG and therefore a rapid change in the permeability and a phase shift (Fig. 14). The data show a linear dependence of the differential phase shift on the electric field E.

C. Nano-composites

A different approach to the required intimate contact between magnetic material and ferroelectric has been to use nano-composites. A typical device[16i] uses PZT-$CoFe_2O_4$ (Fig. 15) and significant development was made in Maryland by Ramesh (now at Berkeley)[19] and Roytburd[20] and their coworkers there,

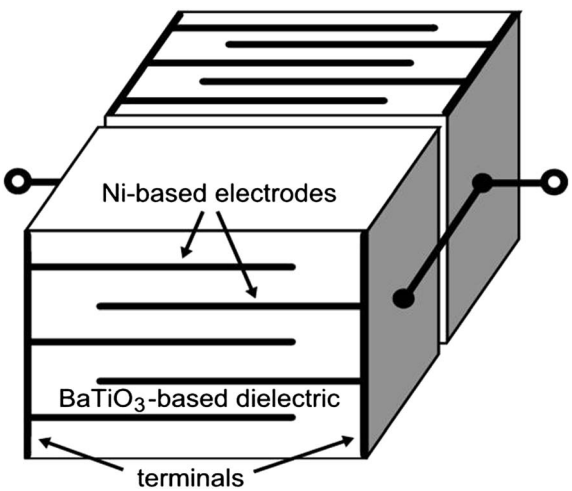

Fig. 10 Temperature-compensated device,[13c] as in Fig. 9.

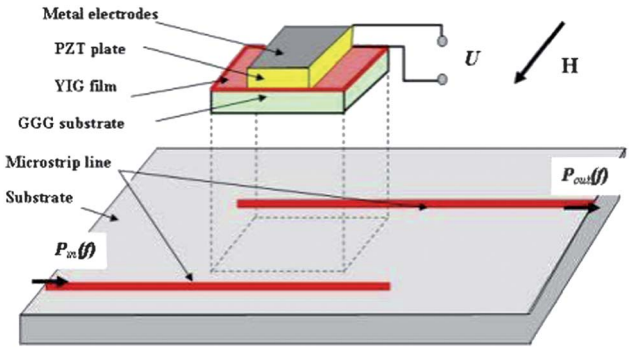

Fig. 11 Magnetoelectric GHz bandpass filter (schematic).[15]

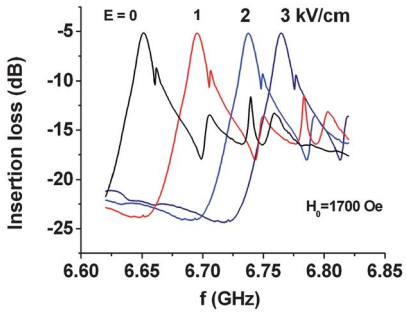

Fig. 12 Response at different electric fields.[15]

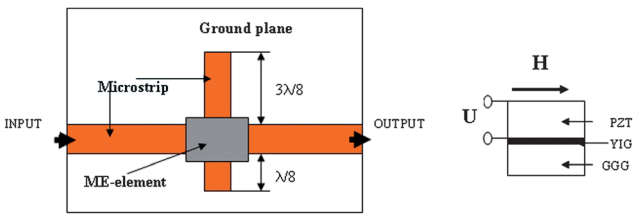

Fig. 13 Multiferroic microwave phase shifter.[15]

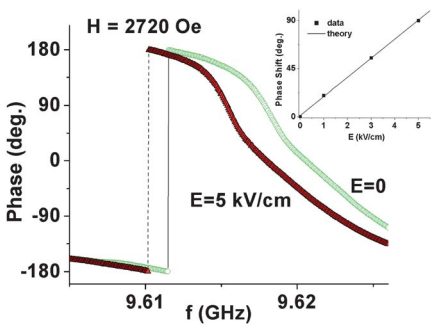

Fig. 14 Phase shift *versus* applied electric field E.[15]

including the use of polymers.[20e] In this case a main problem is achieving epitaxial or grain-ordered nano-structures such that the induced voltages, strain, and/or magnetizations lie in a particular direction.

As with laminar bilayers and superlattices, the main application is detectors.

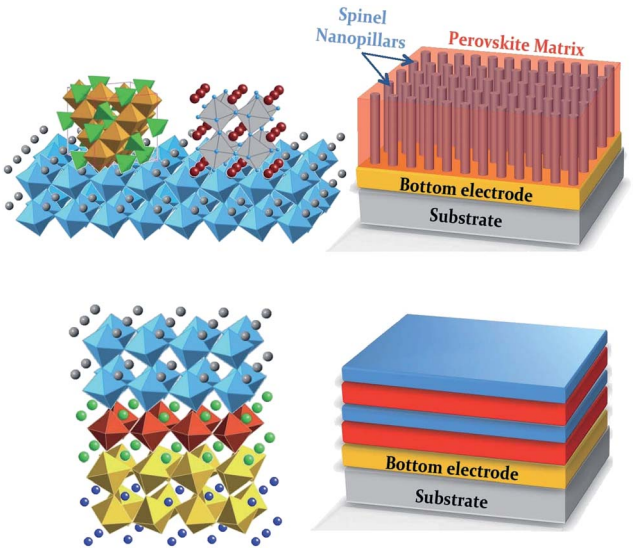

Fig. 15 Nanostructures in PZT-$CoFe_2O_4$ two-phase systems.

D. Clustered ferromagnets

Finally, the last type of magnetoelectric material to be discussed are crystals in which the weak ferromagnetism comes not from Dzyaloshinskii-Moriya canting of antiferromagnetic ordering, but from ferromagnetic clustering of spins. Good examples exist in perovskites[21a–i] including $PbFe_{1/2}Ta_{1/2}O_3$, $PbFe_{1/2}Nb_{1/2}O_3$, and $PbFe_{2/3}W_{1/3}O_3$ and their single-phase mixtures with PZT. Of these, the Fe/W compounds are birelaxors, with no long-range magnetic or ferroelectric ordering at any temperature, but strong short-range ordering coupled *via* strain,[21j,k] whereas the Fe/Ta compounds have long-range order. $PbFe(0.17)Ta(0.17)Zr(0.33)Ti(0.33)O_3$ is in fact a good switching ferroelectric up until 800 K, making it probably the highest-temperature perovskite oxide ferroelectric known. It becomes cubic at 1123 K but centric and quasi-cubic around 850 K. As with the nonmagnetic prototype, $PbSc_{1/2}Ta_{1/2}O_3$, the degree of ordering at the perovskite B-site is highly dependent upon chemical processing, and especially annealing temperature, producing a wide range of structures from fully disordered to perfectly ordered.[22]

E. Domain wall devices

One of the most exciting mew directions in magnetoelectrics is research in the area of domain wall nano-electronics. Stimulated in part by two key observations – first, by Daraktchiev *et al.* that domain walls can be ferromagnetic when the surrounding domains are antiferromagnetic,[23a,b] and second, by Seidel *et al.* that domain walls can be electrically conducting even in rather insulating materials[24] – this has led to work on devices in which the walls themselves are the functional parts. A review has recently been published by Catalan *et al.*,[25] and the most recent developments involve vortex domains. Vortex domain walls have been reported in different contexts lately[26a–d] (Gruverman, Gregg, Pan, Jia), and significantly have been shown to be conducting even when the adjacent domains are not.[27]

F. Conclusion

In summary we see that different kinds of magnetoelectrics are being actively studied, including single-phase crystals, laminar sandwich-like superlattices, nano-composites, and cluster magnets. However, each of these has its own niche application area, with tunnel junctions the main target for single-phase epitaxial thin films and sensors for weak magnetic fields the object of bimorph prototypes. The 100 GHz–1 THz phase shifter also merits further attention.

What is the future? Firstly, it has been thought for many years, since the early work of Brown, Hornreich and Shtrikman,[28] that there was a physical upper limit imposed by nature on the magnitude of magnetoelectric tensor components: $\alpha_{ij} < \chi_i(M)\,\chi_j(E)$, where χ_i (M) is the magnetic susceptibility and χ_j (E) the electric susceptibility (dielectric constant). However, recently Dzyaloshinskii has shown[29] that a may actually diverge without limit in some cases (an example would be if Cr_2O_3 became ferromagnetic at a lower temperature). The reason is that the Brown-Hornreich-Shtrikman theory is a linear response model, whereas things become very nonlinear near phase transitions; so one might hope that very large magnetoelectric coupling can yet be discovered. The second thing to note is that quadratic coupling can be very important for devices. As Bloembergen and Hou showed long ago for nickel sulphate hexahydrate,[30] coupling of form $\beta_{ijk}\,P_i\,M_j\,M_k$ can be very large in crystals. Note that linear-quadratic coupling of this form remains nonzero and often very large for planar magnets well above the three-dimensional Neel temperature, since $<M^2>$ is very large; in $BaMnF_4$ this term gives large dielectric anomalies up to approximately $3T_N$ (*ca.* 80 K).[31a,b] And more recently Kumar *et al.* and Blinc *et al.*[21j,k] have shown that biquadratic coupling of form P^2M^2 can also be large and hugely frequency-dependent; the latter term is always present in magnetic ferroelectrics, since it is a scalar independent of crystal symmetry. Of similar interest is trilinear coupling[31c,d] of form $\mathbf{P}\cdot(\mathbf{L}\times\mathbf{M})$, through which ferroelectricity causes weak ferromagnetism by canting the antiferromagnetic spins *via* a Dzyaloshinskii-Moriya anisotropic exchange. This could be used as a voltage-driven magnetic memory element.

Another problem to overcome is that all room-temperature multiferroics discovered to date are weak ferromagnets. For device application strong ferromagnetism is preferred. The sole strong ferromagnet multiferroic reported is $EuTiO_3$ under stress,[1e] but it functions at only cryogenic temperatures. Thus, further efforts should be made on strongly ferromagnetic multiferroics and on room-temperature or near-ambient materials such as CuO, $Pb(Fe,Ta,Zr,Ti)O_3$, $Pb(Fe,W,Zr,Ti)O_3$ or $Sr_3Co_2F_{24}O_{41}$ and its family.[1e–h] Similarly, for switched capacitor memory devices a swuitched polarization $P > 1\ \mu C\ cm^{-2}$ is required; readers should note that many magnetoelectrics have P of a few $\mu C\ m^{-2}$ or even $nC\ cm^{-2}$, three or four orders of magnitude too small for practical devices, but these units are not mentioned, let alone emphasized by the authors.

From a device point of view the rather elegant THz emission studies of Tonouchi *et al.*[32a,b] on bismuth ferrite should be extended to other multiferroics.

Other new directions would include replacing the PZT in bimorphs or superlattice composites with a polymeric ferroelectric such as PVDF (polyvinylidene difluoride) to achieve larger areas or non-planar structures. An additional benefit might be obtained from the fact that PVDF is strongly flexoelectric.[20e,33a,b] Other magnetic thin films with giant flexoelectricity (voltage output from inhomogeneous strain or *vice versa*) include $HoMnO_3$.[33b] There are of course many magnetoelectric materials that are not oxides, but in fairness the oxides have received the most attention thus far. Two extensive families of fluorides include $K_3Fe_5F_{15}$ and $Pb_5Cr_3F_{19}$.[34a–g] They are very high-temperature ferroelectrics, and are interesting because the former involves 2 Fe^{+3} ions and 3 Fe^{+2} ions per formula group. K can be substituted for by other alkali metals, and Pb by alkaline earths, with a complete range of magnetic ions (Mn, Ni, Co, *etc.*) for Fe or Cr. However, none of these function as multiferroics at room temperature, and $K_3Fe_5F_{15}$ has the highest magnetoelectric temperature of 123 K. Some *ab initio* modeling has been reported[34f] in 2010, but it does not seem fully compatible with the experiments done one or two years earlier,[34b,g] which were unfortunately apparently unknown to the authors of the theories.

Although military devices such as magnetic mines may find immediate application for laminar composite magnetoelectrics, let us end this review on a more benign note: There are equally attractive flexoelectric devices in the health and medical areas, such as refreshable Braille displays, which might benefit from magnetic properties;[35] and use in magnetic resonance imaging (MRI) will probably be forthcoming as well. The availability of room-temperature devices at less than $0.01 apiece[17] may permit applications in new areas such as toys and in pedagogical teaching devices.[36]

Acknowledgements

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