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Review

Functional Requirements of Multiferroic and Magnetoelectric Coupled Materials: Structural, Electronic, and Application-Driven Perspectives

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Abstract:

Multiferroic and magnetoelectrically (ME) coupled materials have attracted considerable attention because they simultaneously support at least two ferroic orders and, in the ME subclass, allow one order to be controlled by a conjugate field of the other. Translating these coupled phenomena into viable device technologies imposes a precise and demanding set of material requirements that extend well beyond the simple coexistence of ferromagnetism and ferroelectricity. This review articulates those requirements in a systematic manner, addressing crystal-structural prerequisites, electronic-structure constraints, thermal and chemical stability benchmarks, and the interface engineering criteria relevant to composite ME heterostructures. Bulk single-phase systems, composite laminates, nanostructured films, and emerging two-dimensional (2D) van der Waals multiferroics are each examined through the lens of what the material must deliver, rather than merely what has been observed. Special attention is given to the figures of merit that govern sensor, energy-harvesting, memory, and neuromorphic device applications, because the requirements hierarchy shifts substantially depending on the end use. Ongoing challenges in reconciling contradictory structural demands, suppressing leakage currents, and characterizing ME coupling quantitatively are discussed, and pathways toward materials that meet multiple simultaneous requirements are proposed. The article draws on a broad literature base spanning theoretical prediction, synthesis, and functional characterization of representative systems including $BiFeO_3$-based perovskites, $BaTiO_3/CoFe_2O_4$ composites, layered $Cr_2Ge_2Te_6$ and $CuCrP_2S_6$, and strain-engineered thin films.

Keywords: multiferroics; magnetoelectric coupling; ferroelectric; ferromagnetic; ME coefficient; composite heterostructure; thin film; 2D multiferroics; device requirements.

1. Introduction

The concept of multiferroicity, understood as the simultaneous presence of two or more primary ferroic orders in a single phase or engineered composite, was placed on a rigorous theoretical footing by Schmid in 1994, yet its practical implications remained largely academic until the rediscovery of strong ME coupling in $BiFeO_3$ thin films around two decades later.[1,2] Since that inflection point, the field has expanded rapidly, driven by the prospect of non-volatile memories that write magnetically and read electrically, ultralow-power sensors, and energy harvesters that exploit both piezoelectric and magnetostrictive transduction within the same structure.[3,4]

Despite this progress, a persistent gap exists between what is desired from a multiferroic material and what any single known compound or composite actually provides. The demands placed on a material by a practical application are multidimensional: a room-temperature ME coefficient large enough to produce a measurable voltage from a modest magnetic field, a coercive electric field low enough to switch polarization without breakdown, a spontaneous magnetization sufficient for read-out, mechanical compliance matched to a substrate, and chemical stability across decades of operation, to name only the most obvious.[5,6,7] Single-phase multiferroics rarely satisfy even half of these simultaneously, while composite ME systems can achieve larger coupling but introduce interface and processing constraints of their own.

This article is organised around the question of requirements rather than achievements. Section 2 surveys the structural and electronic prerequisites at the atomic scale. Section 3 addresses thermodynamic and kinetic stability requirements. Section 4 examines the specific demands imposed by composite ME architectures. Section 5 relates the material requirements to device-level figures of merit for the major application classes. Section 6 discusses unresolved conflicts between simultaneously required properties, and Section 7 concludes.

2. Structural and Electronic Prerequisites

2.1 Symmetry Requirements

The most fundamental criterion for single-phase multiferroicity is that the crystal structure must belong to a space group that simultaneously allows a polar distortion (breaking inversion symmetry) and long-range magnetic order (breaking time-reversal symmetry).[8,9] Of the 122 magnetic point groups, only 13 satisfy both conditions, which immediately explains the rarity of room-temperature single-phase multiferroics. In practice, the structural chemist seeking a new multiferroic must identify a motif in which the ion responsible for off-centring, typically a lone-pair cation such as Bi^{3+} or Pb^{2+}, coexists with a magnetically active sublattice of partially filled d-orbital transition metal ions without the two sublattices electronically quenching each other.[10,11]

The d^0-ness rule, which predicts that B-site transition metal ions in perovskites prefer a centrosymmetric configuration when the d-shell is partially filled, represents a direct structural conflict. This rule has been circumvented in systems such as $BiFeO_3$ by exploiting A-site lone-pair activity rather than B-site displacement, in $EuTiO_3$ under strain where the magnetic Eu^{2+} and off-centre Ti^{4+} occupy separate sublattices, and in hybrid improper ferroelectrics where two non-polar distortion modes combine to generate net polarisation without requiring an off-centred d^0 ion.[12,13]

2.2 Electronic Structure and Bandgap Requirements

For a ferroelectric to function without leakage, the material must be insulating under the applied electric fields used during poling. This requirement translates into a minimum optical bandgap of approximately 2.5 to 3.0 eV for most device contexts, yet simultaneously, the magnetically active sublattice needs partially filled d or f states near the Fermi level to support long-range spin order.[14,15] These two demands are partially contradictory: partially filled d states tend to introduce mid-gap levels that promote polaron hopping and electronic conduction. In $BiFeO_3$, the experimental bandgap of approximately 2.67 eV is marginally acceptable, but the actual leakage in undoped films often exceeds what is tolerable for practical switching because of oxygen vacancies and the mixed-valence character of iron.[16,17]

Charge compensation strategies, including isovalent and aliovalent substitution on both A and B sites, are routinely employed to suppress leakage. La^{3+} substitution on the Bi site in $BiFeO_3$ concurrently reduces Bi volatility, pins the Fermi level, and modestly alters the magnetic structure.[18] Mn^{4+} or Ti^{4+} substitution on the Fe site suppresses the formation of Fe^{2+} centres that otherwise act as electron donors.[19] However, any substitution that modifies the d-occupancy of the magnetic sublattice will alter the superexchange interactions and hence the magnetic ordering temperature, illustrating how tightly the electronic, structural, and magnetic requirements are coupled.

2.3 Magnetic Order Requirements

For device relevance, magnetic ordering should persist well above room temperature (T_n or $T_1 > 500$ K preferred to allow a reasonable operational margin) with a coercive field low enough to permit field-driven switching in the intended application.[20,21] In sensors and energy harvesters, a large piezomagnetic coefficient $d\lambda/dH$ at low fields is more important than a large saturation magnetisation, whereas in ME memory concepts that rely on electric-field-written magnetisation states, a square hysteresis loop with well-defined remnant magnetisation is critical.[22,23] Single-phase multiferroics are generally antiferromagnetic or weakly ferromagnetic at room temperature, which limits their direct utility in devices that require substantial net magnetisation. Composite architectures address this by combining a magnetostrictive ferromagnetic phase such as Terfenol-D, $CoFe_2O_4$, or Ni with a piezoelectric phase, effectively decoupling the magnetic and polar order requirements between two different materials.[24,25]

3. Thermodynamic and Kinetic Stability Requirements

3.1 Phase Stability and Processing Windows

A multiferroic compound intended for thin-film integration must be thermodynamically stable or, at minimum, kinetically persistent within the processing conditions imposed by substrate compatibility, which typically means deposition temperatures between 550 and 750°C in an oxidising atmosphere and post-deposition cooling that avoids decomposition or phase segregation.[26] $BiFeO_3$ is metastable with respect to $Bi_{25}FeO_{39}$ and $Bi^2Fe_4O_9$ below approximately 770°C, meaning that extended annealing at lower temperatures leads to parasitic phase formation that interrupts polarisation switching and introduces magnetic impurities that complicate interpretation.[27,28] Tight control of oxygen partial pressure, film thickness, and heating/cooling rates are therefore requirements rather than options in $BiFeO_3$ processing.

Multicomponent systems such as $(1\text{-}x)BiFeO_3\text{-}xBaTiO_3$ or $Ba(Fe0.5Ta0.5)O_3$ solid solutions widen the phase-stable composition window while simultaneously shifting the magnetic ordering temperature, allowing some degree of independent optimisation.[29] In bulk ceramics prepared by solid-state reaction or sol-gel auto-combustion, the sintering atmosphere and duration must suppress both Bi_2O_3 evaporation and Fe^{3+} to Fe^{2+} reduction, both of which introduce conducting defects incompatible with stable polarisation measurements.[30]

3.2 Thermal Stability of Coupling

Even when the ferroic orders individually survive to high temperature, the ME coupling tensor $\alpha^{I}{}_{j}$ may vanish or change sign at temperatures well below either the magnetic or ferroelectric Curie point if the coupling mechanism involves a thermally fragile intermediate such as a spin-lattice interaction of second order.[31] This is particularly evident in type-II multiferroics, where ferroelectricity is induced by non-collinear spin spiral ordering: the polarisation disappears when the spiral unwinds under modest applied fields or temperature increases, setting a much lower operational ceiling than the nominal magnetic ordering temperature.[32] Requirements for practical devices thus demand that the coupling mechanism be robust, meaning it should originate from direct overlap integrals between magnetic and polar distortion modes rather than from fragile spin-current or exchange-striction mechanisms that are easily disrupted.

4. Requirements for Composite ME Architectures

4.1 Interface Quality

In two-phase composites, whether laminate, particulate, or vertically aligned nanostructured, the ME coupling is mediated entirely through mechanical strain transfer across the interface between the magnetostrictive and piezoelectric phases. This immediately establishes interface quality as the dominant materials requirement, eclipsing even the intrinsic properties of the individual phases.[33,34] A well-bonded, chemically clean interface with minimal interdiffusion is needed to ensure efficient strain transmission. Any interfacial layer, whether an amorphous interdiffusion zone, a secondary phase formed by elemental exchange, or an oxide barrier grown during high-temperature processing, acts as a mechanical compliance buffer that attenuates the transmitted strain and proportionally degrades the effective ME coefficient.[35]

Quantitative studies on $BaTiO_3/CoFe_2O_4$ bilayers and $Pb(Zr,Ti)O_3$/Terfenol-D laminates consistently show that the experimentally measured longitudinal ME coefficient α_{ll} can be two to five times smaller than theoretical predictions based on the single-crystal elastic and piezoelectric constants of the constituent phases alone, with the discrepancy attributable primarily to interfacial non-ideality and partial depoling of the piezoelectric layer during magnetostrictive cycling.[36,37] This finding implies a practical requirement: the processing route must be designed to minimise interfacial reaction while maximising adhesion, often a conflicting pair of goals that must be balanced through careful selection of deposition sequence, temperature, and interlayer materials.[38]

4.2 Phase Connectivity and Geometry

The BulkHagen notation for composite connectivity (0-3, 1-3, 2-2, and 3-3) captures a fundamental geometric requirement: the dimensionality through which each phase is continuously connected governs both the strain-transfer efficiency and the electrical percolation threshold.[39] A 2-2 laminate with full-area bonding provides the highest mechanical coupling for a given volume fraction, which is why laminates based on PMN-PT single crystals bonded to Metglas amorphous ribbons have achieved ME coefficients exceeding 20,000 mV cm⁻¹ Oe⁻¹ under resonant conditions.[40,41] However, such laminate structures require manual assembly and are not compatible with thin-film microfabrication. Epitaxial vertical nanostructures (1-3 connectivity) offer a substrate-compatible alternative but are subject to substrate clamping that severely suppresses in-plane strain and reduces the effective ME response.

4.3 Resistivity Matching

A frequently overlooked composite requirement is that the electrical resistivities of the two phases must be matched in a way that prevents the low-resistivity magnetic phase from electrically shorting the piezoelectric phase. In $CoFe_2O_4/BaTiO_3$ composites, the spinel ferrite is sufficiently insulating to allow standard impedance measurements without shorting, but in composites incorporating metallic magnetostrictive alloys (Terfenol-D, Galfenol), an electrically insulating adhesive or bonding layer must be inserted, which simultaneously introduces the compliance mismatch discussed above.[42,43] This resistivity mismatch requirement becomes especially demanding in thin-film geometries where conventional oxide ferrites are the preferred magnetic partner because their resistivity can vary by several orders of magnitude depending on stoichiometry and processing atmosphere.[44]

5. Application-Specific Requirements

5.1 Magnetic Field Sensors

In a passive ME magnetic sensor, the relevant figure of merit is the voltage sensitivity $S^{\gamma} = \alpha^{I}{}_{j} / \varepsilon r\ \varepsilon_0$, where $\alpha^{I}{}_{j}$ is the ME coefficient and ε_r is the relative permittivity.[45] A high ME coefficient is necessary but not sufficient: a material with very large permittivity will dilute the voltage output even if α is respectable. This places a simultaneous requirement on low-loss, moderate-permittivity piezoelectric behaviour, which generally favours hard PZT or AlN over relaxor ferroelectrics that exhibit large ε_r but also large dielectric loss.[46] Beyond the ME coefficient, the sensor noise floor is governed by the mechanical quality factor of the resonant mode used for detection, the 1/f magnetic noise of the magnetostrictive layer, and the electronic noise of the readout circuit. The material requirement translates into a high-Q mechanical resonance, meaning low acoustic damping in both phases, combined with a magnetostrictive layer that shows minimal magnetic viscosity and domain-wall pinning noise.[47,48]

5.2 Energy Harvesters

Vibration-based ME energy harvesters convert ambient mechanical oscillations into electricity via the magnetostrictive-piezoelectric coupling pathway. The power output scales as the square of the effective ME coefficient and as the operating bandwidth around mechanical resonance.[49] The material requirements for this application are therefore a large dλ at low field amplitudes (to generate sufficient strain from ambient vibrations), a high piezoelectric charge coefficient d_{33} or d_{31} in the coupled phase, a mechanical Q factor high enough to amplify strain at resonance but not so high as to make the device excessively narrowband, and a damping loss small enough that the harvested power exceeds internal dissipation.[50,51] Because ambient vibration spectra are broad and time-varying, there is growing interest in broadband harvesters achieved through nonlinear bistable configurations or arrays of differently tuned resonators, but these solutions impose additional mechanical requirements on fatigue resistance and long-term stability of the ME bond under cyclic loading.[52]

5.3 Non-Volatile Memory

ME-based non-volatile memory concepts rely either on the electric-field-written control of magnetisation direction (ME-RAM) or on the magnetically assisted switching of ferroelectric polarisation.[53,54] The requirements for ME-RAM are among the most stringent in the field: the ME coupling must be large enough that a voltage of a few volts across a nanometre-scale film can deterministically reverse the magnetisation of an adjacent ferromagnetic layer, the magnetisation must remain stable between write events (high thermal stability energy K_uV/k_1T), and the ferroelectric switching must be repeatable to at least 10^{10} cycles without fatigue or imprint.[55] No known single-phase multiferroic simultaneously meets all three of these requirements at room temperature, which has led to composite strategies in which an ME interface layer couples a separate ferroelectric gate to a ferromagnetic storage layer. Even in this composite geometry, the requirements are demanding: the gate dielectric must be robust, the ME interface coupling must be reproducible, and spin-orbit torque must be sufficient to achieve complete reversal.[56,57]

5.4 Neuromorphic and Reconfigurable Devices

Emerging neuromorphic computing architectures propose multiferroic tunnel junctions and ME synaptic elements as analogue weight elements in artificial neural networks.[58] The material requirements here differ substantially from binary memory: rather than a sharp two-state switching behaviour, a gradual and controllable variation of tunnelling conductance as a function of applied voltage pulse is needed, mimicking the potentiation and depression of biological synapses.[59] This places a requirement on a smooth polarisation gradient across the tunnel barrier, controlled by partial switching or domain nucleation dynamics, rather than on complete polarisation reversal. The reproducibility of the intermediate states under repeated programming and the retention of those states over time are the relevant figures of merit, and meeting them simultaneously requires precision control of defect density, domain-wall pinning energy landscape, and barrier homogeneity at the nanometre scale.[60]

6. Challenges

6.1 Simultaneously Demanded but Contradictory Properties

Perhaps the central intellectual challenge in multiferroic materials science is that many of the most strongly desired properties are intrinsically in tension with one another. Table 1 summarises some of the most commonly encountered conflicts:[61,62]

(i) High spontaneous polarisation versus low leakage current: large polar distortions tend to create a narrow bandgap and a large density of polar phonons that facilitate polaron hopping; (ii) high magnetic ordering temperature versus weak magnetoelectric coupling: materials stabilised by strong superexchange, such as antiferromagnetic oxides, rarely show the large spin-lattice coupling needed for a practical ME response; (iii) large magnetostriction versus mechanical hardness: highly magnetostrictive materials such as Terfenol-D are mechanically brittle and prone to cracking under the cyclic stress of resonant operation; (iv) epitaxial strain enhancement of coupling versus substrate clamping: misfit strain in thin films can stabilise polar phases but simultaneously clamps the in-plane piezoelectric response.[63,64,]

6.2 Quantification Challenges

A persistent methodological challenge that compounds the difficulty of meeting application requirements is the absence of standardised measurement protocols for the ME coefficient α^{I}_{j}.[65] Dynamic ME measurements using a small AC magnetic field superimposed on a DC bias field are the most common, but the extracted α depends sensitively on the frequency, AC field amplitude, DC bias, sample geometry, and whether the sample is mechanically clamped or free-standing. Resonant enhancement near the electromechanical resonance frequency can yield α values three to four orders of magnitude larger than the quasi-static result, creating a literature in which comparisons across reports are often misleading.[66,67] Establishing a set of standard test conditions, analogous to those adopted for piezoelectric characterisation, is itself a requirement that must be met at the community level before the materials-level requirements discussed in earlier sections can be reliably benchmarked.[68]

7. Conclusions

The foregoing analysis demonstrates that the path from a multiferroic curiosity to a deployable device is governed not by a single exceptional property but by the simultaneous satisfaction of a hierarchy of coupled requirements. At the atomic scale, the material must reconcile the symmetry conflict between polar distortion and magnetic order, suppress electronic leakage without sacrificing magnetic exchange strength, and maintain both ferroic transitions well above the intended operating temperature. At the mesoscale, composite architectures must deliver mechanically efficient, chemically stable interfaces whose strain-transfer fidelity is not degraded by interdiffusion, clamping, or resistivity mismatch between constituent phases. At the device level, the relevant figure of merit shifts substantially with application: voltage sensitivity and low noise for sensors, large piezomagnetic coefficient and mechanical Q for harvesters, fatigue-free cycling and sharp switching for memory, and controllable intermediate polarisation states for neuromorphic elements. No currently known material or composite meets all of these requirements simultaneously, which explains why the field continues to expand despite two decades of intensive research. Future progress will depend as much on the establishment of standardised measurement protocols for the ME coefficient as on the discovery of new compounds, since reliable benchmarking across laboratories is a prerequisite for rational materials optimisation. The requirements framework presented here is intended to serve as a practical reference for directing synthesis, characterisation, and device integration efforts toward targets that are both physically justified and application-relevant.

Author Contributions

A.M, B.G.H and S. M. contributed to the data analysis, wrote, conceived, contributed to the text in the manuscript, commented on the manuscript, and approved the final version.

Declaration of Competing Interest

Authors don't have any conflict of interest.

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