The estimation of physical properties in textured polycrystals is reviewed. “Principal” properties, which relate actions and responses within the same subsystem (electric, elastic, ...), as well as “coupling” properties (e.g., piezomagnetism), linking actions, and responses associated with various subsystems (magneto-elastic, thermo-electric, ...) are analyzed. Tensor ranks from 1 to 4, with polar and axial characteristics are considered. Virtual-time inversion (the case of magnetostriction) is taken into account. Matrix and surface representations are considered. Significant differences in the effect of texture on properties arise from the diversity of properties tensors ranks and polar/axial natures. To predict the effective values of coupling properties, precautions required for application of the Voigt, Reuss, and Hill approximations are pointed out. At all stages of the proposed methodology, a symmetrized spherical harmonics treatment of the orientation distribution functions, the inverse pole figures and (single- and polycrystals) physical properties is applied. For the case of magnetostriction, a functional program for estimating polycrystal performance is included as Supporting Information. The input data are the single-crystal property coefficients and the polycrystal inverse pole figure parameters. The coincidence of predicted magnetostriction coefficients with experimentally measured values is satisfactory. Recently established considerations regarding the characterization of coupling properties in complex materials are divulged.

1. Introduction

“Coupling” properties, in a given material, link one subsystem (say, elasticity) of the investigated object to a different subsystem (say, magnetism). Piezoelectricity, magnetostriction and magnetoelectricity are examples of coupling properties.\[1\text{–}3\]

Current technological requirements of high-performance transducers (sensors, actuators) demand deep and systematic studies of coupling properties.\[4\text{–}6\] Present work is part of a current research trend oriented to characterize the complex materials that are used in the manufacture of diverse sensors and actuators. Recent research in ferro-piezoelectric,\[7\text{–}10\] multiferroic,\[5,6\] and magnetostrictive\[11,12\] materials illustrates the level of performances and scientific challenges that characterize the current state of the art. The introduction of composite materials opens new possibilities of diversification of the useable physical effects and/or increases the values of technologically interesting parameters.\[13,14\] This has led to the development of the theory of homogenization,\[15,16\] which makes possible the prognosis of properties for complex multicomponent materials.\[17,18\]

In the mentioned scenario, the present article is devoted to review the influence of crystallographic textures on electromagnetic coupling properties. In texture research, the influence of preferred orientation on “principal” properties (action and effect on the same subsystem) has been far more investigated and applied than the role of texture on coupling properties.\[19,20\] Knowledge about texture and mechanical properties has grown impressively.\[21,22\] In what follows, textured polycrystals coupling properties are characterized. The case of electromagnetic interactions is analyzed in detail. Some criteria and methods for estimating effective macroscopic properties are given. An open source computer program for a predictive estimation of magnetostriction in axially textured polycrystals is provided. Throughout, the article attention is focused on different coupling interactions to discuss different components of the general theme considered in the work.

2. The Representation of Physical Properties: Principal and Coupling Interactions

2.1. Properties Tensors

A linear description of materials physical properties is expressed by the tensor constitutive Equation 1:

\[
\begin{align*}
\text{Principal Properties:} & \\
\text{Coupling Properties:} & \\
\end{align*}
\]
Y = KX.  \hspace{1cm} (1)

X characterizes an action on a sample; Y is a measure of the specimen response, and K represents the investigated material’s property. The \((X, Y, K)\) respective tensors ranks are \(m\), \(n\), and \(m + n\). Besides rank, the polar or axial nature of the considered magnitudes, as well as their behavior under space and time inversion, play important roles on materials properties.

Figure 1 represents the thermo–elasto-electro-magnetic equilibrium interactions of electrically isolating materials, under the linear approximation. The spheres in the external tetrahedron represent actions (“causes”, independent variables) and the ones in the internal tetrahedron describe the material responses (“effects”, dependent variables). Responses are linked to actions through linear constitutive relations, for example, \(D = \kappa \cdot E\).

Table 1 summarizes the definitions of the magnitudes and properties presented in Figure 1. Cause-effect couples in a given vertex of Figure 1 belong to the same sub-system (thermal, electric, magnetic) in the investigated material. They represent the principal interactions. Links between magnitudes in different vertices characterize the mixed interactions or coupling properties.

The properties in Figure 1 and in Table 1 display the wide spectrum of mathematical characteristics of physical properties. Tensors ranks scan from 0 (heat capacity) to 4 (elasticity). Polar (electric) and axial (magnetic coupling) magnitudes are present and space-time inversion differences also appear.

Working with constitutive relations, the independent variable (the “action”) is assumed invariant in the sample volume. For a homogeneous sample, (say a single crystal) this condition is automatically fulfilled. For a textured polycrystal (the situation of interest in the present paper), this is not always the case. The independent variables in Figure 1 and in Table 1 are suitable for the geometric configuration known as “the Voigt condition”. This point is discussed below.

The formal characterization of tensor properties is by means of their representative components \(K_{ij...n}\). The number of indexes equals the tensor rank \(r\). For tensor ranks \(r \geq 3\), besides a \(r - \) dimensional array, the so-called matrix notation is widely used. The dimensionality of the representative matrix is lowered. Third rank tensors (e.g., piezoelectricity) are described by \(3 \times 6\) matrices and fourth rank tensors (e.g., elasticity) by \(6 \times 6\) ones. Details of the tensor → matrix transformation can be found, for example, in the classic book by Nye.\(^{[23]}\)

A tendency in properties characterization is the display of the so-called longitudinal surfaces representation of tensor properties. In the mentioned surfaces, the distance from the reference frame origin to the surface represents the projection of the property in the considered direction. Longitudinal surfaces represent the anisotropy of physical properties in a vivid manner.

As we have mentioned, the present treatment is based on the linear approximation of interactions and properties. This characterization has high practical value, but also has limitations. From the classical works of Preisach\(^{[24]}\) on magnetic hysteresis to the recent treatment of the elasto-magneto-electric interaction with homogenization tools,\(^{[25]}\) the consideration of non-linearity has been systematically gaining importance. Non-linear modeling of ferroelectrics has been discussed by Landis.\(^{[26]}\)

![Figure 1](https://www.advancedsciencenews.com/doi/abs/10.1002/adem.201700827/fig1?pp=1700827)  
Figure 1. Principal and coupling interactions and properties. Adapted under the terms of the Creative Commons Attribution License\(^{[3]}\) 1998, Hindawi.

2.2. Properties Databases

The classical work of Landolt-Bernstein\(^{[27,28]}\) is the historical and most representative collection of materials properties numerical data. It can be accessed via Internet (http://materials.springer.com/) and is extraordinarily useful. A very interesting material properties database, the Materials Project, is being developed at UC-Berkeley. This is a
supercomputer materials prediction project.\cite{29,30} They have already calculated from first principles the structure and several physical properties of thousands of hypothetical and real materials. Obtained theoretical results are acceptably close to experimental ones. It is worth navigating their web page (https://materialsproject.org/). Another internet-based properties database is the Material Property Open Database MPOD, a France-Mexico-Italy-Lithuania project (http://mpod.cimav.edu.mx).\cite{31,32} MPOD collects and delivers as open-access information the published experimental values of properties matrices of a growing number of crystalline materials. A consistent dictionary with a system of properties definitions and references to the original sources are provided.

The Materials Project and MPOD display in color the properties surfaces. The surface color in MPOD is a function of the property value. MPOD also generates “stl” files, suitable to produce 3D-prints of any property surface. Figure 2 shows representative computer display of the piezoelectric tensor e of a quartz crystal.\cite{33} Additional examples of MPOD outputs are shown in the Supporting Information (Figures S1 and S2).

### Table 1. Constitutive equations and tensor types for thermo-elasto-electro-magnetic equilibrium properties.

<table>
<thead>
<tr>
<th>Property and constitutive equations</th>
<th>Related magnitudes</th>
<th>Tensor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Principal</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat capacity, C: ( S = \rho C \frac{\Delta T}{T} )</td>
<td>Entropy S (P0)/Temperature T (P0)</td>
<td>P0</td>
</tr>
<tr>
<td>Elastic stiffness, C: ( \sigma = \varepsilon \varepsilon )</td>
<td>Stress ( \sigma ) (P2)/Strain ( \varepsilon ) (P2)</td>
<td>P4</td>
</tr>
<tr>
<td>Electric permittivity, ( \kappa ): ( D = \kappa E )</td>
<td>Electric displacement ( D ) (P1)/Electric intensity ( E ) (P1)</td>
<td>P2</td>
</tr>
<tr>
<td>Magnetic permeability, ( \mu ): ( B = \mu H )</td>
<td>Magnetic induction ( B ) (A1)/Magnetic intensity ( H ) (A1)</td>
<td>P2</td>
</tr>
<tr>
<td>Coupling</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thermal expansion, ( \eta ): ( \sigma = \eta T ), ( S = \eta \varepsilon )</td>
<td>Stress ( \sigma ) (P2)/Temperature T (P2)</td>
<td>P2</td>
</tr>
<tr>
<td>Pyroelectricity, ( \rho ): ( D = \rho T ), ( S = \rho E )</td>
<td>Elect. displacement ( D ) (P1)/Temperature T (P0)</td>
<td>P1</td>
</tr>
<tr>
<td>Pyromagnetism, ( \beta ): ( B = \beta T ), ( S = \beta H )</td>
<td>Magnetic induction ( B ) (A1)/Temperature T (P0)</td>
<td>A1</td>
</tr>
<tr>
<td>Piezoelectricity, ( \alpha ): ( D = \alpha \varepsilon ), ( \sigma = \alpha E )</td>
<td>Electric displacement ( D ) (P1)/Strain ( \varepsilon ) (P2)</td>
<td>P3</td>
</tr>
<tr>
<td>Piezomagnetism, ( \beta ): ( B = \beta c ), ( \sigma = \beta H )</td>
<td>Magnetic induction ( B ) (A1)/Strain ( \varepsilon ) (P2)</td>
<td>A3</td>
</tr>
<tr>
<td>Magnetoelasticity, ( \alpha ): ( B = \alpha E ), ( D = \alpha H )</td>
<td>Magnetic induction ( B ) (A1)/Electric intensity ( E ) (P1)</td>
<td>A2</td>
</tr>
</tbody>
</table>

Legend: Tensor characteristics: P: Polar; A: Axial; \( r = 1, \ldots, 4 \): tensor rank. Scalars are in italics, vectors and tensors in bold.

3. Symmetry and Physical Properties — General

The physical basis for establishing symmetry-based selection rules for materials properties is the Neumann Principle: *Effect’s symmetry is always, at least, equal to cause’s symmetry* . In electromagnetic theory, fields’ symmetry cannot be lower than charges and currents symmetry. In crystal physics, properties symmetry must be, at least, equal to the structure symmetry. In group-theoretical argot, for single-crystal macroscopic properties: the point symmetry group of any physical property contains, as a subgroup, the structural point group of the considered crystal.

The Neumann Principle, in representation theory,\cite{34} leads to the following selection rule: For the physical quantities X and Y to be related by a nonzero property K, invariant under the structure point group (P), the bases representing both magnitudes (X and Y) must belong to the same irreducible representation (“irrep”) of (P). X and Y are, respectively, components of action and response tensors X and Y considered hitherto.

The irreps methodology has allowed the compilation of tables describing the detailed structure of properties matrices in a systematic scan of tensors ranks. Mentioned tables can be found in several crystal physics books.\cite{35,36} The peculiar case of magnetoelasticity is considered below.

Electric and magnetic magnitudes behave differently under symmetry operations and they fulfill the Neumann Principle in interestingly distinct manners. For a perovskite crystal ferroelectricity is impossible if the crystal structure is centrosymmetric. But for an α-iron sample, with centrosymmetric BCC crystal structure, ferromagnetism is allowed. A horizontal electric current ring (cause) is symmetric with

![Figure 2](image-url). Piezoelectric longitudinal modulus e (h). Quartz single crystal. Structure point group: 321 = D3. Property point group: \(-6\infty = D_{6h}\). Scale bar units: C/N.
respect to a horizontal mirror. But its magnetic field (effect) shows opposite (North and South) poles up and down the mentioned mirror.

These paradoxes find their solution in the different mathematical natures of electric and magnetic vectors. Electric vectors (E, P, D) are polar, they invert themselves under spatial inversion and are invariant under (virtual or real) time inversion. Magnetic vectors (B, M, H) are defined as vector products, they are axial, invariant under spatial inversion, and invert themselves under time inversion. Polar and axial vectors differ consequently under anti-symmetry operations.

Figure 3 shows, by means of an example, the different behaviors of polar and axial magnitudes. The electric dipole in (a) is surrounded by a polar electric field. The magnetic field around the magnetic dipole in (b) is axial. The equatorial (horizontal) plane is an anti-symmetry element in the electric case and a symmetry mirror for the magnetic dipole. In both cases, the charge/current → electric/magnetic fields relationships follow the cause → effect Neumann Principle for color symmetry.

Properties, for example, piezoelectricity, that transform a polar action (stress or strain) into an axial response (magnetization) are axial. The classification in Table 1 was established in accordance with the aforementioned criterion.

The different behavior of magnetic magnitudes under symmetry operations, especially under space and time inversion transformations, has generated the generalization of the symmetry concept, leading to the magnetic (color) symmetry groups. In this conceptual frame, the number of symmetry point groups goes from 32 to 122. There are 32 ordinary crystallographic point groups, 32 gray and 58 "black and white" point groups.

4. Electromagnetic Coupling: Representative Tensors

4.1. Magneto-electricity

The characterization of magneto-electricity is analyzed next. The applied methodology follows the approach given by Nowick. LiCoPO₄ is taken as a case study. The (ordinary) point group of this material’s atomic distribution is D₂h = mmm. Table 2 shows the irreps of this group. According to the axial nature of the magnetoelectric tensor, to find the nonzero elements of a we must identify the components of polar vectors (x, ...) and axial ones (Rx, ...) that share irreps. Inspecting the D₂h irrep column of the Vector bases field in Table 2, no single irrep shared by polar and axial vectors are found. Apparently, LiCoPO₄ could not be magnetoelectric. Our analysis, to be correct, must take into consideration the magnetic point group, not only the ordinary one.

According to Rivera, the magnetic point group of LiCoPO₄ is D₂h(C₃ᵥ) = mmm. This magnetic group is related to D₂h as follows. C₃ᵥ is a subgroup of D₂h, containing half of the D₂h elements. Denoting as Θ the time inversion operator, D₂h(C₃ᵥ) = C₃ᵥ + Θ(D₂h − C₃ᵥ). Combined with Θ, symmetry operations preserve their symmetry nature when acting on time-invariant vectors (E, P, and D). When acting on time-reversible vectors (B, M, H), Θ converts symmetry into anti-symmetry. The consideration of magnetic symmetry leads to the identification of the complementary irreducible representations (co-irreps), shown in the D₂h(C₃ᵥ) co-irreps column of Table 2. Inspection of the just mentioned column shows that (x; Rₓ) share the B₂u co-irrep and (y; Rᵧ) share B₃u. It follows that the magnetoelectric matrix for the investigated magnetic point group has the structure given in Equation 2.

\[
a = \begin{bmatrix}
0 & a_{12} & 0 \\
a_{21} & 0 & 0 \\
0 & 0 & 0
\end{bmatrix}
\]  

(2)

The numerical values of aᵢⱼ for the specific case of LiCoPO₄ at T = 4.2 K have been found by Rivera, Equation 3.

\[
a = \begin{bmatrix}
0 & 18.4 & 0 \\
30.6 & 0 & 0 \\
0 & 0 & 0
\end{bmatrix} \text{ (ps/m)}
\]  

(3)

Figure 4 represents the magnetoelectric longitudinal coefficient for the mentioned crystal. Due to the off-diagonal type of the magnetoelectric tensor, the coupling effect in this case is mostly transverse. The maximum magnetoelectric field appears in the y axis under an excitation along the x axis. The maximum...
longitudinal effect appears parallel with the diagonals of the \((x, y)\) axes. The vertical planes parallel to \(x\) and \(y\) axes \((y = 0\) and \(x = 0)\) are ordinary symmetry planes. They transform positive lobes into negative ones. The \(z = 0\) plane is an anti-mirror. Magnetic symmetry operates in an interesting way on the axial magnitudes.

Next, we briefly mention some topics beyond single-crystal linear magnetoelectricity. Second-order approximation to magnetoelectric effect is represented by Equations 4 and 5:

\[
P_m = a_{mn} H_n + \gamma_{mn} H_n E_l + \frac{1}{2} \beta_{mn} H_n H_l \tag{4}
\]

\[
\mu_0 M_m = a_{mn} E_n + \beta_{mn} E_n H_l + \frac{1}{2} \gamma_{mn} E_n E_l \tag{5}
\]

Table 2. Character and vector bases for \(D_{2h}\) and \(D_{2h}(C_{3v})\) irreducible representations (irreps) and complementary irreducible representations (co-irreps).

<table>
<thead>
<tr>
<th>(D_{2h})</th>
<th>(E)</th>
<th>(C_2(x))</th>
<th>(C_2(y))</th>
<th>(C_2(z))</th>
<th>(i)</th>
<th>(\sigma^x)</th>
<th>(\sigma^y)</th>
<th>(\sigma^z)</th>
<th>(D_{2h}) Irreps</th>
<th>(D_{2h}(C_{3v})) Co-irreps</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A_g)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>(B_{1g})</td>
<td>1</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>–</td>
<td>(R_x)</td>
</tr>
<tr>
<td>(B_{2g})</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>–</td>
<td>(R_y)</td>
</tr>
<tr>
<td>(B_{3g})</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>(R_z)</td>
<td>–</td>
</tr>
<tr>
<td>(A_u)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>(x)</td>
<td>(z)</td>
</tr>
<tr>
<td>(B_{1u})</td>
<td>1</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>(z)</td>
<td>(y_{x} R_{x})</td>
</tr>
<tr>
<td>(B_{2u})</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>(y)</td>
<td>(y_{y} R_{y})</td>
</tr>
<tr>
<td>(B_{3u})</td>
<td>1</td>
<td>–1</td>
<td>–1</td>
<td>1</td>
<td>1</td>
<td>–1</td>
<td>1</td>
<td>–1</td>
<td>(x)</td>
<td>(x_{z} R_{z})</td>
</tr>
</tbody>
</table>

Legend: \(x\), \(y\), \(z\): generic polar vector components; \(R_x\), \(R_y\), \(R_z\): generic axial vector components.

Longitudinal effect appears parallel with the diagonals of the \((x, y)\) axes. The vertical planes parallel to \(x\) and \(y\) axes \((y = 0\) and \(x = 0)\) are ordinary symmetry planes. They transform positive lobes into negative ones. The \(z = 0\) plane is an anti-mirror. Magnetic symmetry operates in an interesting way on the axial magnitudes.

Piezoelectric-piezomagnetic (or magnetostrictive) magneto-electric composites show high coupling parameters. Characterizing these systems is one of the most important challenges today in the field of magnetoelectricity. On theoretical grounds, if the interaction among composite particles is not taken into account, magnetoelectricity cannot be explained. Linear[18] and non-linear[25] homogenization treatments for the just stated problem have been developed. Figure 5 shows the calculated composition dependence of the magnetoelectric coefficient for a \(\text{BaTiO}_3–\text{CoFe}_2\text{O}_4\) composite.[18]

The link between magnetoelectricity and ferroic phenomena represents a topic of interest nowadays. The aforementioned group theoretical approach allows a systematic classification of the existing magnetic point groups regarding this correlation. Figure 6 shows in a Venn diagram the distribution of magnetic symmetry groups among the electromagnetic properties.
4.2. Magnetoelasticity: Piezomagnetism and Magnetostriction

Coupling between magnetization \( \mathbf{H} \), \( \mathbf{B} \), \( \mathbf{M} \), and strain \( \varepsilon \) may be linear or higher order. The first case is piezomagnetism and the second one is magnetostriction. For the selection of independent variables represented in Figure 1, the constitutive equations of piezomagnetism are: \( \sigma = \mathbf{b} \cdot \mathbf{H}; \ B = \mathbf{b} \cdot \mathbf{e} \). The piezomagnetic tensor, denoted \( \mathbf{b} \), is axial, third rank (A3). The piezomagnetic matrices, in the so-called “compact-matrix” notation, are \( 3 \times 6 \) matrices. The selection rules for non-zero and correlated matrix elements are derived by the group-theoretical methodology just outlined. Equation 6 represents the piezomagnetic tensor of a z-magnetized crystal of the Fe–Ga (galfenol) family. The galfenol crystal structure (ordinary point group) is magnetically tetragonal, with magnetic point group \( \text{point group } C_{4h} \text{ of } 4 \text{mm} \). The structure of the piezomagnetic matrix is the same as the piezoelectric matrix of a BaTiO\( _3 \) crystal (ordinary point group \( C_{4h} = 4 \text{mm} \)).

\[
\mathbf{b} = \begin{bmatrix}
0 0 0 & b_{15} & 0 \\
0 0 0 & b_{15} & 0 \\
b_{31} & b_{31} & b_{33} & 0 & 0
\end{bmatrix}
\] (6)

Magnetostriction: Quadratic effects in tensor representation lead to products of axial tensor components. This way, squared axial piezomagnetic components give up the fourth rank polar tensor magnetostriction. Equation 7 is the magnetostriction constitutive relation in tensor notation, valid for all symmetries. A usual practice in magnetostriction studies is to perform measurements under saturation magnetization and to select magnetization \( \mathbf{M} \) and deformation \( \varepsilon \) as independent and dependent variables, respectively.

\[
\varepsilon = \sum_{kl} N_{ijkl} M_k M_l (\text{general, tensor notation})
\] (7)

Magnetostriction in magnetostriction investigations usually goes up to saturation values, practically independent of crystal directions. The magnetostrictive deformation \( \varepsilon \) under saturation magnetization is generally denoted \( \lambda \). Regarding magnetostriction, galfenol crystals follow their cubic structural symmetry. The magnetostriction coefficients, in matrix notation, adopt the structure of Equation 8.

\[
\begin{bmatrix}
\varepsilon_1 \\
\varepsilon_2 \\
\varepsilon_3 \\
\varepsilon_4 \\
\varepsilon_5 \\
\varepsilon_6
\end{bmatrix} =
\begin{bmatrix}
N_{11} & N_{12} & N_{12} & 0 & 0 & 0 \\
N_{12} & N_{11} & N_{12} & 0 & 0 & 0 \\
N_{12} & N_{12} & N_{11} & 0 & 0 & 0 \\
0 & 0 & 0 & N_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & N_{44} & 0 \\
0 & 0 & 0 & 0 & 0 & N_{44}
\end{bmatrix}
\begin{bmatrix}
M_1^4 \\
M_2^4 \\
M_3^4 \\
M_2 M_3 \\
M_4 M_3 \\
M_4 M_2
\end{bmatrix}
\] (cubic, matrix notation) (8)

This last equation is analogous to the matrix expression for the elasticity of cubic crystals. The analogy is extended to the longitudinal surface representations. As with the Young modulus for cubic crystals, the longitudinal magnetostriction of a cubic crystal is represented by:

\[
\lambda = \lambda_{100} + 3(\lambda_{111} - \lambda_{100})(h_1^2 h_2^2 + h_2^2 h_3^2 + h_3^2 h_1^2)
\] (9)

\( \lambda_{111} \) and \( \lambda_{100} \) are materials coefficients and \( h_1 \) are direction cosines.

The surface representation associated with Equation 9 is included in the MPOD application. Several magnetostrictive systems are registered in the database. Figure 7 shows the anisotropic magnetostriction of terfenol and galfenol crystals. Cobalt ferrite and associated solid solutions have been widely investigated. Interestingly, small variations of composition lead to significant variations in the magnetostrictive parameters.\[40,41\]

5. Texture and Polycrystal Physical Properties

Texture is one of the most important factors determining the physical properties of polycrystals. Connectivity, so-called stereography\[42\] and other factors could also be mentioned. Texture acts as a modulating agent. In first approximation, polycrystal properties are single-crystal ones, averaged by the orientation distribution function (ODF). In this section, we first show how different properties, depending on tensor rank, and polar/axis nature are affected differently by texture. Next, we point-out the limitation of averaging as predictive tool and mention solution alternatives. Finally, current trends toward realistic predictions of polycrystals magnetoelastic properties are reviewed.
5.1. Qualitative Findings from Properties’ Averages

The following discussion applies to a single-phase polycrystal with a given distribution of its crystallites orientations (namely, with a given texture). As described by Bunge in his classical textbook,[43] the appropriate statistical descriptor of texture is the orientation distribution function (ODF). The ODF represents the relative population of crystallites in the neighborhood of a point in orientation (Euler) space. The ODF plays the role of a weighting factor for averaging single-crystal tensor properties (expressed as matrices as well as by surfaces). Averaging is carried out, in short, as follows. Single- and polycrystal properties are represented by their respective symmetrized spherical harmonics expansions. Polycrystal properties coefficients are linear combinations of single crystal ones’ times the ODF expansion coefficients. Specific algorithms for polycrystalline magnetostriction, consistent with the general theory,[44] are presented below in the description of the program **Magnetostriction_Texture.py**. The CIMAV Crystal Physics Group works to introduce in MPOD the option of estimating average properties of textured polycrystals using programs based on the Bunge method. The project progress can be followed on the web page (http://cimav.edu.mx/software).

**Figure 8**–**10** demonstrate the fundamental diversity of texture effect on different properties.

The dielectric constant (P2 tensor) of a BaTiO3 crystal is highly anisotropic. **Figure 8** shows the evolution of the dielectric surface for BaTiO3 fiber textures, as the hypothetical samples evolve from sharp texture to a random distribution of orientations. The dielectric constant changes from highly anisotropic to isotropic.

Quartz longitudinal piezoelectric surface (P3 tensor) preserves its shape, but collapses to a point as the ODF losses sharpness around the origin of Euler space, **Figure 9**.

The magnetoelectricity (A2 tensor) of LiCoPO4, **Figure 10**, behaves qualitatively in a manner similar to quartz piezoelectricity.

The origin of such a wide diversity of polycrystal behaviors lies in the multiplicity of mathematical natures of the considered properties.

5.2. Average Versus Effective Properties

Equation 10 describes the possible limitation of average properties as representative of the effective magnitudes that describe a material’s global performance:

\[
\bar{Y} = \bar{K} \cdot \bar{X} + \frac{1}{V} \int_{V} \Delta K \cdot \Delta X dV 
\]  

(10)

\(\bar{X}, \bar{Y}, \) and \(\bar{K}\) are average (mean) magnitudes in the investigated sample. As seen from equation (10), the mean \(\bar{K}\) represents the effective property if the independent variable remains invariant in the sample volume (\(\Delta X = 0\)). In our chemically homogeneous material,
effective polycrystal properties not only depend on the distribution of orientations, but also on crystallites’ shapes, sizes and relative positioning, that is, on sample’s so-called stereology.[42]

Polycrystals with crystallites arranged near to parallel or series configurations may be properly characterized, respectively, by the well-known Voigt (parallel) or Reuss (series) approximations. The Hill treatment, averaging Voigt, and Reuss results may be more realistic. Mentioned data treatments consist on suitable selection of the independent variable in the corresponding constitutive equations. The geometric mean approach[45] is an alternative that generates a unique and self-consistent result. In given cases, it is the best solution.

Dealing with coupling properties special precautions must be taken. The analysis in ref.[44] is summarized as follows. Consider the case of piezoelectricity. If one averages the “piezoelectric charge constant “d”, it works well from the point of view of the equation \( D = d \cdot \sigma \), if the system resembles a series configuration (constant \( \sigma \)). But \( d \) also appears in \( \kappa = d \cdot E \) and \( E \) is constant in parallel configurations, not in series. The solution to the mentioned paradox, for parallel (Voigt) configurations is the selection of independent variables shown in Figure 1. \( E, H \), and \( \kappa \) are invariant in this geometry and the corresponding properties may be averaged. For series (Reuss) association, the correct independent variables are \( D, B, \) and \( \sigma \). The thermodynamic magnitudes (\( T, S \)), due to their scalar nature, are not affected.

The Voigt, Reuss and Hill approximations, applied with proper precautions, lead to acceptable estimates in single phase materials. As general tendency, Voigt and Reuss values are taken as bounds for the estimation of effective properties.[46–48] Coupling properties in complex systems (say, composite magnetoelctrics) require more elaborated homogenization methods. The books by Milton[15] and Sihvola[16] are acknowledged references in the subject.

5.3. Recent Advances in Polycrystals Properties Prediction: The Magnetostriction Case

Since the discovery of giant magnetostrictive materials like Terfenol-D,[49] the importance of developing non-linear phenomenological models was noticed. Phenomenological models are suitable for the control of magnetostrictive materials due to their high accuracy and natural property of minor loops closure. The core of this theory is formed by so-called hysteresis operators, which describe hysteretic transducers as a mapping between function spaces. Three of the most representative models are the following:

1) The classical Preisach model,[24,50,51] where the hysteresis is modeled as an aggregate effect of all possible delayed relay elements which are parameterized by a pair of threshold variables.
2) The Krasnosel’skii-Pokrovskii model,[52] where the delayed relay elements have finite slopes.
3) The Prandtl-Ishlinskii model,[53] that is, a superposition of elementary play or stop operators.

Figure 9. Effect of texture on SiO₂ (quartz) piezoelectric longitudinal modulus. External surfaces: Single crystal; internal surfaces: Hypothetical textured polycrystal. a) Sharp texture (FWHM = 10°); c) Broad texture (FWHM = 30°).

Figure 10. Single- and polycrystal longitudinal magnetoelectric surfaces for LiCoPO₄. The external surface corresponds to the single crystal. The considered ODF is composed by a Gaussian component at the origin of the Euler space, with a FWHM of 30°. Reproduced with permission.[6] 2015, Elsevier.
Extensions of the classical Preisach model are available, including anisotropy effect, stress dependence, rate dependence, frequency dependence (hysteresis losses), and temperature dependence among other contributions.

Another approaches for non-linear dynamic models are based on finite element calculations. These models were first developed by Kvarnäs and Engdahl for a Terfenol-D rod. The magnetic and mechanical properties were given from static characterization, implying the need of an experimental set-up used for data acquisition of the material properties. Several other models were developed from this idea for Terfenol-D and for Galfenol as well. A limitation of finite element modeling is that it does not produce general solutions, but only numerical values for specific situations.

Microscopic or “physics-based” models are often grounded on the Jiles–Atherton model of ferromagnetic hysteresis. In these treatments, hysteresis is considered to arise from pinning of domain walls on defect sites. The use of a few physically related material parameters and the simplicity of use of these models made them very practical in engineering applications. The Jiles-Atherton model was extended for magnetostriction hysteresis by Sablik and Jiles and the effects of anisotropy and stress were later added. To characterize the eddy-current losses, Jiles modified the magnetization model by increasing irreversible magnetization. Wang et al. presented a nonlinear transient model with eddy current effect to describe effectively the influences of exciting frequency and temperature on the dynamic magnetostrictive effect. Figures 11 and 12 show some representative Wang’s theory results. More recently, a new model fully describing the effects of stress, frequency, and ambient temperature on the hysteresis characteristics of magnetostriction and magnetization has been formulated, on the basis of the thermodynamic theory, by Xiao. In this model, the Weiss molecular field and losses due to eddy currents are taken into account. Figure 13 shows the satisfactory level of theory-experiment coincidence in Xiao’s work.

Figure 11. Magnetostrictive strain hysteresis loops under different exciting frequency from 1 to 2000 Hz with fixed stress $-1500 \text{ psi}$ and temperature $T = 18 \degree \text{C}$. Reproduced with permission.

Figure 12. Magnetostrictive strain hysteresis loops under different temperature from 0 to 90 $\degree \text{C}$ with fixed stress $-1500 \text{ psi}$ for the exciting frequency of $f = 1000 \text{ Hz}$. Reproduced with permission.

Figure 13. Comparisons of magnetic hysteresis loops a) and magnetostrictive strain curves b) with and without Weiss molecular field effect under the exciting frequency $f = 1 \text{ Hz}$. Reproduced with permission.
distribution of magnetization energy states, that is, the result of crystal defects. The first proposed models were anhysteretic, but more recently, Daniel proposed an extension of them. This new irreversible model is fully multiaxial, allows the description of typical hysteresis and butterfly loops (Figure 15) and the calculation of magnetic losses as a function of external magneto-mechanical loadings.

6. The “Texture_Magnetostriction” Code

For saturation magnetostriction the magnetization vector is practically constant in the sample volume. The volume average of the magnetostriction constant gives a satisfactory first approximation to the magnetoelastic coupling coefficients. To calculate the texture-modulated $\lambda$ as a function of
orientation, the Bunge methodology\(^{[23]}\) has been systematized as a Python program. The developed code is given in the Supporting Information. The mathematics of the procedure is as follows:

The magnetostriction surface \(\lambda(h)\) for a cubic single crystal is represented as a symmetrized spherical harmonics expansion:

\[
\lambda(h) = \varepsilon_0 + \varepsilon_4 k_4(h) \quad (11)
\]

\(k_4(h)\) is the symmetric spherical harmonic of cubic symmetry corresponding to \(l = 4\) in the Bunge scheme:

\[
k_4(h) = n_4 \left[ 5(h_1^2 h_2^2 + h_1^2 h_3^2 + h_2^2 h_3^2) - 1 \right] \quad (12)
\]

\(h_i\) are the direction cosines of orientation \(h\) and \(n_4 = -0.646360\).

The expansion coefficients of \(\lambda(h)\) are:

\[
\varepsilon_0 = \frac{2}{5} \lambda_{100} + \frac{3}{5} \lambda_{111} \quad \varepsilon_4 = \frac{3}{n_4} (\lambda_{111} - \lambda_{100}) \quad (13)
\]

\(\lambda_{100}\) and \(\lambda_{111}\) are single crystal magnetostrictive coefficients.

Polycrystals are treated by an analogous formalism, with properties expansion coefficients determined by the texture. For fiber textures, the average magnetostriction shows axial symmetry \(\tilde{\lambda} = \tilde{\lambda}(\phi)\) and is characterized by a Legendre polynomials \([P_l(\phi)]\) expansion:

\[\tilde{\lambda}(\phi) = \tilde{\varepsilon}_0 + \tilde{\varepsilon}_4 P_4(\phi)\]  

(14)

with

\[P_4(\phi) = \frac{3\sqrt{2}}{16} (35\cos^4\phi - 30\cos^2\phi + 3)\]  

(15)

The polycrystal isotropic coefficient \(\tilde{\varepsilon}_0\) coincides with that of the single crystal, \(\tilde{\varepsilon}_0 = \varepsilon_0\). The anisotropic coefficient \(\tilde{\varepsilon}_4\) is calculated as follows. We describe the fiber texture by the inverse pole figure \(R(h)\) of the sample symmetry axis. \(R(h)\) is modeled as a Gaussian distribution around the preferred orientation \(h_0\).

\[R(h) = R_0 e^{-(\phi/\Omega)^2}\]  

(16)

\(\phi\) is the angular distance between the observation direction \(h\) and the reference \(h_0\). The parameter \(\Omega\) is linked to the “full width at half maximum” (FWHM) of the orientation distribution by the

---

**Figure 16.** Evolution of galfenol magnetostriction \(\lambda, \tilde{\lambda}\) with texture width FWHM. Single crystal parameters: \(\lambda_{100} = 212, \lambda_{111} = -13\). a) Single crystal; b) sharp texture (FWHM = 10°); c) broad texture (FWHM = 60°); d) random distribution of orientations.
relation \( \Omega = 0.6 \times \text{FWHM (radians)} \). \( R_0 \) is adjusted by the normalization condition:

\[
\int R(h) dh = 4\pi
\]  

(17)

The model inverse pole figure is expanded in spherical harmonics. The expansion coefficients of \( R(h) \) lead to the calculation of the coefficients of \( \tilde{\alpha} \), in our case of \( \tilde{c}_4 \). Mathematically:

\[
\tilde{c}_4 = \int R(h) k_4(h) dh
\]  

(18)

The integration is done by numerical quadrature with the dblquad function of the scipy library. The authors declare no conflict of interest.

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Supporting Information

Supporting information is available online from Wiley Online Library or from the author.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

galfenol magnetostriction, polycrystal magnetic symmetry, texture-properties relationship

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Figure S1: Young modulus. Quartz single crystal. a) View from a horizontal second order rotation axis. b) View from the vertical third order rotation axis. Structure point group: D₃. Property point group: D₃d.

Figure S2: 3D printing of quartz properties. a) Young modulus. b) Piezoelectric longitudinal modulus.
“Texture-Magnetostriction”

AN OPEN SOURCE PYTHON CODE

# -*- coding: utf-8 -*-
#
# MAGNETOSTRICTION IN AXIALLY TEXTURED CUBIC POLYCRYSTALS
#
import numpy as np
from numpy import sin, cos, pi  # makes the code more readable
from mayavi import mlab
import scipy.integrate as sq

def R1(azimut, polar):
    # Inverse pole figure. Preferred orientation = 0,0,1
    return np.exp(-(polar/omr)**2.0)*sin(polar)

def R(azimut, polar):
    # Normalized inverse pole figure
    return (4.0*pi/R0)*np.exp(-(polar/omr)**2.0)*sin(polar)

def R4(azimut, polar):
    # Integrand for texture coefficient C41. Explicit cubic symmetry spherical harmonic
    R41 = (4.0*pi/R0)*np.exp(-(polar/omr)**2.0)*sin(polar)
    k41 = 0.2423851 + 2.423851*cos(polar)**2.0 + 2.827827*cos(polar)**4.0 + \
          0.33799*sin(polar)**4.0*cos(4.0*azimut)
    return R41*k41

def Surface_representation(E, phi, beta):  # Graphic representation
    x1 = E*sin(phi)*cos(beta)
    y1 = E*sin(phi)*sin(beta)
    z1 = E*cos(phi)
    mlab.figure()
    mlab.mesh(x1, y1, z1, scalars=E)
    mlab.scalarbar(orientation="vertical")
    mlab.show()

if __name__ == "__main__":
    # Inicial data
    n4=-0.646360
    FWHM=10.0
    omgr=0.6*FWHM
    omr = omgr*pi/180.0

    # Cristina sample:
    L100=318.0*2.0/3.0; L111=-20.0*2.0/3.0
    # Terfenol-D:
    #L100=90.0; L111=1640.0
    # Hypothetical isotropic crystal:
    #L100=1.0; L111=1.0
# Spherical coordinates arrays:
phi, beta = np.mgrid[0.0:pi:180j,0:2.0*pi:360j]

# SINGLE CRYSTAL MAGNETOSTRICTION
# Cubic symmetry spherical harmonics
h1 = sin(phi)*cos(beta)
h2 = sin(phi)*sin(beta)
h3 = cos(phi)
Fi4 = h1*h1*h2*h2+h2*h2*h3*h3+h1*h1*h3*h3
k41 = n4*(5.0*Fi4-1.0) # Bunge expression

# Single crystal magnetostriction coefficients
em01 = 2.0*L100/5.0+3.0*L111/5.0;
em41 = 3.0*(L111-L100)/(5.0*n4)
print "em01,em41 =\n", em01, em41

# Single crystal magnetostriccion
E = L100+3.0*(L111-L100)*Fi4 # Trigonometric expression
#E = em01+em41*k41 # Spherical harmonics expression
Surface_representation(E, phi, beta)

# POLYCRYSTAL
# Inverse pole figure integral.
RR = sq.dbquad(R1,0.0,pi/2.0,lambda polar: 0.0, lambda polar: 2.0*pi)
R0=6.0*RR[0]
print "FWHM, Integral of R1 =\n", FWHM, R0

# Checking normalization condition.
RR = sq.dbquad(R,0.0,pi/2.0,lambda polar: 0.0, lambda polar:2.0*pi)
RR0 = 6.0*RR[0]
print "Integral of R, 4*pi =\n", RR0, 4.0*pi

# Texture coefficient C41:
C411 = sq.dbquad(R4,0.0,pi/2.0,lambda polar: 0, lambda polar: 2.0*pi)
C41 = 6.0*C411[0]
print "C41 =\n", C41

# Polycrystal magnetostriction expansion coefficients:
ep01 = em01
ep41 = (1.0/(4.0*pi))*np.sqrt(2.0/9.0)*em41*C41
#ep41 = 0.0 # Hypothetical random distribution
print "ep01, ep41 =\n", ep01, ep41

# Legendre polynomial:
P4 = (3.0*np.sqrt(2.0)/16.0)*(35.0*cos(phi)**4.0-30.0*cos(phi)**2.0+3.0)

# Polycrystal magnetostriction:
Ep = ep01 + ep41*P4
Surface_representation(Ep, phi, beta)