“Giant” Electrostriction

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The recent discovery of “giant” electrostrictors has re-ignited the interest in electrostriction, an electromechanical coupling existing in all dielectrics but overshadowed by its linear counterpart: piezoelectricity. In this review, after a reminder of “classical” electrostriction, we propose a definition of “giant” electrostriction based on two empirical relations (“Newnham” relation and one we propose). From this definition, we review previous reports on “giant” electrostrictors, to assess their nature. Focusing on the ones satisfying our definition, we compare their performances and characteristics. We also identify some of the hurdles to their adoption in the wide range of electromechanical applications, despite their fundamental and applicative interests.

I. INTRODUCTION

Electromechanical materials (piezoelectrics and electrostrictors) convert electric energy to mechanical energy (deformation or stress) and vice versa, thereby enabling sensors and actuators applications, for example in Micro-Electro-Mechanical Systems (MEMS) such as micro-machines, micro-switches, digital micro-mirrors, micro-valves and micro-pumps. Inverse piezoelectricity is the most common electromechanical coupling for actuators, where a linear strain or stress is induced by an electric field or electric displacement. Another electromechanical coupling that can be used for actuators is electrostriction, albeit with a strain quadratic in electric field or polarization. On the sensor side, direct piezoelectric effect is primarily used. If piezoelectricity can only exist in dielectrics with a non-centrosymmetric structure, electrostriction occurs in all dielectrics without exception.

The sign of the induced electrostrictive strain (positive, corresponding to stretching or expansion, or negative for compression) is independent of the field polarity. In most ceramics, electrostrictive stretching is observed in the direction of the external field, whereas polymers generally exhibit the opposite effect, i.e. compression.

The electrostrictive effect is usually very small compared to commercial piezoelectrics. However, in 2012, Gd-doped ceria thin films have been discovered to have an exceptionally large electrostriction coefficient, a behavior later also reported in ceramics. Nb,Y-stabilized bismuth oxide (Bi$_2$Nb$_{2-x}$Y$_x$O$_{2-2y/3}$) and La$_2$Mo$_2$O$_9$ (LAMOX) have been discovered to also exhibit “giant” electrostrictive performances. These giant electrostrictors generally have relative permittivity $\varepsilon_r \approx 30$, exhibit “giant response” at low frequency (around 10 Hz), and contain oxygen vacancies ($V_O$) as candidate materials for electrolytes in solid oxide fuel cells.

Lead halide perovskite single crystals methylammonium lead triiodide (MAPbI$_3$) have also demonstrated “giant” negative electrostrictive responses at 100 Hz.

In addition, soft nano-composites based on a polymer matrix and carbon nanotubes (CNT) and liquid-crystalline graphene (rGO) are another type of remarkable electrostrictors as they have been reported to have an “ultra-large” response for frequencies between 0.1 and 100 Hz.

Giant electrostrictors offer a promising orientation toward the development of electromechanical applications not only due to their superior electromechanical properties, but also because they could provide a replacement for lead-based PZT that is at the heart of the vast majority of piezoelectric applications.

Here, we shall review extensively the literature on giant electrostrictors to put their performances in perspective and enable the comparison with regular electrostrictors as well as assess their suitability for applications. The first part is devoted to classical electrostriction: the definitions, expressions, and relationships between the various electrostrictive coefficients as well as the universal empirical trends that characterises it. We shall demonstrate an equivalent trend for the electrostrictive coefficient, which is of greatest interest for applications. We shall also propose, from these two trends, a quantitative criterion to qualify an electrostrictive response as “giant”. The second part of the paper reviews extensively the “giant” electrostrictors reported so far, their performances, and whether they meet the criterion we have defined. The third part assesses their interest both on the fundamental level and for applications and underlines the major hurdles to overcome to fully benefit from this effect.

A. Definition of electrostriction

For linear electric and elastic materials, the polarization $P$ induced by an electric field $E$ is $P_i = \varepsilon_0 \chi_{ij} E_j$ (with $\varepsilon_0$ the permittivity of vacuum and $\chi_{ij}$ the elastic susceptibility) and Hooke’s law gives the strain $\epsilon$ induced by a stress $X$: $\epsilon_{ij} = s_{ijkl} X_{kl}$ with $s$ the elastic compliance tensor. Electromechanical properties relate strain/stress to electric field/electric displacement or vice-versa. There are therefore four electromechanical coefficients (tensors).

Electrostriction exists in all dielectrics, irrespective of the presence of a center of inversion or not (in contrast to piezoelectricity) and can be positive or negative. The
electromechanical coupling in a centrosymmetric dielectric can be expressed as the strain \( x_{ij} \) generated by an electric field or a polarization:

\[
\begin{align*}
-x_{ij} &= M_{ijkl} E_k E_l \\
-x_{ij} &= Q_{ijkl} P_k P_l
\end{align*}
\]  
(1)

where \( M_{ijkl} \) (\( m^2/V^2 \)) and \( Q_{ijkl} \) (\( m^4/C^2 \)) are respectively the field- and polarization-electrostriction tensors.

Similarly to the four piezoelectric tensors \( (d, g, e, \text{and} h) \), there are four electrostriction tensors. In addition to \( M \) and \( Q \), the \( m \) and \( q \) electrostriction tensors are defined as the stress \( (X_{ij}) \) generated by an electric field or a polarization, respectively:

\[
\begin{align*}
-X_{ij} &= m_{ijkl} E_k E_l \\
-X_{ij} &= q_{ijkl} P_k P_l
\end{align*}
\]  
(2)

Relations between the four piezoelectric \( (d, e, g, \text{and} h) \) and the four electrostrictive tensors \( (Q, q, M, \text{and} m) \) are illustrated in Fig. 1.

\[
\begin{align*}
d_{jk} &= \frac{1}{1+\kappa_k}\frac{d_{jk}}{e_{jk}^n}, \\
e_{jk} &= \frac{1}{1+\kappa_k}\frac{e_{jk}^n}{e_{jk}}, \\
g_{jk} &= \frac{1}{1+\kappa_k}\frac{g_{jk}}{e_{jk}^n}, \\
h_{jk} &= \frac{1}{1+\kappa_k}\frac{h_{jk}}{e_{jk}}, \\
d_{jk} &= \frac{Q_{ijkl}}{1+\kappa_k}\frac{M_{ijkl}}{Q_{ijkl}}, \\
e_{jk} &= \frac{Q_{ijkl}}{1+\kappa_k}\frac{g_{ijkl}}{Q_{ijkl}}, \\
g_{jk} &= \frac{Q_{ijkl}}{1+\kappa_k}\frac{h_{ijkl}}{Q_{ijkl}}, \\
h_{jk} &= \frac{Q_{ijkl}}{1+\kappa_k}\frac{m_{ijkl}}{Q_{ijkl}}, \\
&= Q_{ijkl}\frac{\chi_{ijkl}}{Q_{ijkl}}
\end{align*}
\]

FIG. 1. Relations among the four piezoelectric (left) and electrostrictive (right) tensors, via the elastic constant \( (c_{ijkl}) \) and compliance \( (s_{ijkl}) \) tensors, the electric susceptibility \( (\epsilon_{ijkl}) \), and its inverse \( (\eta_{ijkl}) \) in linear dielectric and elastic materials under isothermal or isentropic conditions.

Practically speaking, the two components of the electric field \( (E_k \text{ and} E_l) \) are most conveniently applied along the same direction. This leads to the quadratic relations between strain or stress and electric field or polarization. In addition, using Voigt notation, the expressions of the electrostrictive effects can be simplified:

\[
\begin{align*}
x_i &= M_{ik} E_k^2 \\
x_i &= Q_{ik} P_k^2
\end{align*}
\]  
(3)

with \( i = \pm 1 \) and \( k = \pm 3 \).

The difference between the effect described by the \( M \) and \( Q \) tensors appears under large fields. If both x-E curve and x-P curve are quadratic under small amplitude of the electric field (i.e. are described by Eq. (3)), the saturation of the polarization under large electric field in non-linear dielectrics leads to a saturation of the induced strain \( x \). The x-E curves is therefore not quadratic anymore, whereas the x-P curve remains quadratic. Expanding the expression of the polarization as a function of the field to higher terms in Eq. (3) leads to Eq. (4) where cubic, quartic etc. terms appear, whereas only the first (quadratic) term remains in linear dielectrics and leads to \( M_{ijll} = Q_{ijkl}(\epsilon_0\chi_{kl})^2 \).

\[
x = QP^2 = Q(c_0\chi_1 E + c_0\chi_2 E^2 + ...) \approx Q(c_0\chi_1)^2 E^2 + Q(2c_0^2\chi_1\chi_2)E^3 + Qc_0^2(\chi_1\chi_3 + \chi_2^2)E^4 + ...
\]

\[
= ME^2 + M^2E^3 + M^3E^4 + ...
\]  
(4)

This behavior is illustrated in Fig. 2(a) on a ferroelectric relaxor PMN-0.1PT \(((\text{PbMg}_2/3\text{Nb}_1/3\text{O}_3)_{0.9}\text{(PtTiO}_3)_{0.1})_7\) ceramic. The polarization-hysteresis loop (Fig. 2(a)) shows a saturation above \( \approx 1 \) MV/m. As a consequence the induced strain is only quadratic for low field (x-E curve, Fig. 2(c)), whereas the strain remains quadratic in polarization (x-P curve, Fig. 2(e)). In addition, the phase difference between the polariation and the electric field is more pronounced in the x-E curves than in the x-P curve. The evolution with frequency is also different. Figure 2(d) shows that the \( M \) coefficient decreases with increasing frequency (the maximum strain decreases whereas the maximum field is constant). On the contrary, the \( Q \) coefficient increases (a given strain occurs for lower polarisation values as the frequency increases), in agreement with the P-E loop shown in Fig. 2(b). Even though x-E loops (Fig. 2(c)) and x-P (Fig. 2(d)) are not archetypal parabola, fast Fourier transform enables to evaluate the \( M \) coefficient as electrostriction occurs at twice the electric field frequency.

Piezoelectricity and electrostriction are related. For example, piezoelectricity stems from electrostriction in ferroelectric materials. In addition, electrostrictors can exhibit an “effective” piezoelectric response when they are polarised by a dc field. The effective piezoelectric coefficient is proportional to the product of the electrostrictive coefficient \( M \) and bias field \( (E_{bias} \text{ in Fig. } 3(d)) \) in Fig. 3(a) shows the voltage profile with time that is applied on the sample, consisting of a dc voltage (2kV) inducing a static strain. In order to measure the effective piezoelectric behavior, a sinusoidal voltage of \( \pm 100 \) V or \( \pm 50 \) V is applied at a frequency of 10 Hz. The corresponding ac field causes an oscillating displacement of the top surface of the sample (measured by laser interferometry) at the same frequency and with an amplitude proportional to the ac field (see Fig. 3(b)). The proportionality and in-phase character result in a linear (ac) strain response to the applied ac field (as illustrated in Fig. 3(c)), akin the one of a true piezoelectric. The slope of this curve corresponds the effective piezoelectric coefficient \( (d_{eff}) \).

As illustrated in Fig. 3(d), the effective piezoelectric coefficient corresponds the derivative of the x=E curve around the strain induced by the dc field, provided the ac amplitude is small compared to the dc one. A large electrostriction coefficient \( M \) therefore contributes to large
FIG. 2. The electric-field induced polarization for PMN-0.1PT crystals at 10 Hz applied 0.5 MV/m, 1 MV/m, 1.5 MV/m, 2 MV/m respectively (a) and applied 1 MV/m at 1 Hz, 10 Hz, 100 Hz (b). The x-E loops of PMN-0.1PT ceramic with respect to AC electric field at various amplitudes (c) and frequencies (d), which demonstrate the saturation with high electric field and larger electromechanical effect at lower frequencies. The x-P loops PMN-0.1PT crystals at different AC electric field at 10 Hz (e) and different frequencies at 1 MV/m (f).

The effective piezoelectric coefficient, as shown in Eq.(5)

$$d_{33}^{\text{eff}} = \partial x_{33} / \partial E_3 = M_{33} \partial (E_{3(\text{dc})}) + E_{3(\text{ac})})^2$$

$$= 2M_{33} E_{3(\text{dc})} \partial E_{3(\text{ac})} = 2M_{33} E_{3(\text{dc})}$$

This is only valid for a linear dielectric. The saturation of the dielectric response induces a saturation of the strain response (see Fig.3(d)). The corresponding effective piezoelectric coefficient (182 pm/V) is therefore smaller than the value given by Eq.(5) (549 pm/V).

B. The origin of electrostriction

The origin of electrostriction is related to anharmonic effects. In other words, electrostriction coefficients are derived from third derivatives (Eq.8) of the free energy.

Using the elastic Gibbs function $G_2$ and its differential form with respect to the free energy of vacuum:

$$G_2 = U - TS - E_i P_i$$

$$dG_2 = -SdT + X_{ij} dE_{ij} - E_i dE_i$$

where $U$ the internal energy density ($J \cdot m^{-3}$), $T$ the temperature ($K$), $S$ the volume density of entropy ($J \cdot K^{-1} \cdot m^{-3}$). The total differential related to the strain is derived from Eq.(6):

$$dG_2 / dE_j = \partial (dG_2 / dE_k) / \partial E_j T,E dE_k + \partial (dG_2 / dE_j) / \partial T x,E dT + \partial (dG_2 / dE_j) / \partial E_i T,x dE_i$$

$$= \partial X_{kl} / \partial E_j T,E dE_k - \partial S / \partial E_j x,E dT - \partial P_i / \partial E_j T,x dE_i$$

where the first term in Eq.(7) is the isothermal electromechanical coupling introduced in Eq.(2). The second term is the electrocaloric effect, inverse effect of pyroelectricity. The third term is the purely dielectric term. Combining Eqs.(2), (6), and (7), the $m$ electrostrictive tensor can be related to the rate of change of the electric susceptibility.
with respect to strain:

\[
m_{ijkl} = \frac{1}{2} \frac{\partial^3 G_2}{\partial E_i \partial E_j \partial x_{kl}} = \frac{1}{2} \frac{\partial^2 P_i}{\partial E_j \partial x_{kl}}
\]

\[
= \frac{1}{2} \frac{\partial (\epsilon_0 \chi_{ij})}{\partial x_{kl}}
\]

(8)

The three other electrostrictive coefficients can be derived from the electric Gibbs free energy \( G_i \) (\( Q_{ijkl} \)), Gibbs free energy \( G \) (\( M_{ijkl} \)) and Helmholtz free energy \( A \) (\( q_{ijkl} \)).

\[
Q_{ijkl} = -\frac{1}{2} \frac{\partial^3 G_1}{\partial P_i \partial P_j \partial X_{kl}} = -\frac{1}{2} \frac{\partial (\epsilon_0 \chi_{ij})}{\partial X_{kl}}^{-1}
\]

\[
M_{ijkl} = \frac{1}{2} \frac{\partial^3 G}{\partial E_i \partial E_j \partial X_{kl}} = \frac{1}{2} \frac{\partial (\epsilon_0 \chi_{ij})}{2 \partial X_{kl}}
\]

(9)

\[
q_{ijkl} = \frac{1}{2} \frac{\partial^3 A}{\partial P_i \partial P_j \partial x_{kl}} = \frac{1}{2} \frac{\partial (\epsilon_0 \chi_{ij})}{\partial x_{kl}}^{-1}
\]

(10)

Eq. (8) and (9) show that electrostriction coefficients are related to the electric susceptibility (or its inverse) and strain or stress, which enables efficient \( ab \) \( initio \) calculations.\(^8\) In addition, Eq. (8) and Eq. (9) can be rewritten as Eq. (10). This defines the inverse electrostrictive effect, that is the polarization or electric field induced by deforming a sample polarized by an electric field. An equivalent definition is the polarization (or electric field) induced by applying an electric field on (or polarising) a strained material.

\[
\delta P_i = -2m_{ijkl}\delta E_i \delta x_{ij}
\]

\[
\delta E_k = -2q_{ijkl}\delta P_i \delta x_{ij}
\]

(10)

Direct electrostriction (Eq. (8) and (9)) enable applications as actuators, akin to inverse piezoelectricity. The existence of an inverse electrostriction (e.g. Eq. (10)) is often overlooked, probably as it requires the application of a bias (electric or mechanical) field. Similar to direct piezoelectricity, inverse electrostriction enables sensing.

C. General trend of hydrostatic electrostrictive coefficients

To enable the comparison of electrostrictive properties between materials of different symmetry, an hydrostatic electrostriction coefficient \( Q_h \) has been introduced.\(^21\)\(^24\) The hydrostatic electrostrictive coefficient \( Q_h \) is defined as the quadratic relationship between the change of volume \( (\Delta V/V) \) and spontaneous polarization \( P \) (\( P^2 = P_1^2 + P_2^2 + P_3^2 \)), with Eq. (6) of \( Q_{ijkl} \), \( Q_h \) can be expressed as:

\[
Q_h = \frac{\Delta V/V}{P^2} = -\frac{1}{2} \frac{\partial (\epsilon_0 \chi)}{\partial \rho}
\]

where \( \rho \) is hydrostatic pressure.

\[
Q_h \text{ characterizes the electrostrictive effects of isotropic or crystalline materials, for instance } Q_h = Q_{iii} + 2Q_{ijj} \text{ for cubic materials, where } Q_{iii} \text{ and } Q_{ijj} \text{ are respectively the longitudinal and transverse electrostriction coefficients. Hydrostatic electrostriction coefficients can be calculated from the measurements of longitudinal electrostriction in Equation (12), where } \nu \text{ is the Poisson’s ratio in a cubic or isotropic sample.}
\]

\[
Q_{ijj} = -\nu Q_{iii}
\]

\[
Q_h = (1 - 2\nu)Q_{iii}
\]

(12)

\( Q_h \) is a function of the electric susceptibility, but not all materials exhibit the same dependence: perovskite oxides with comparable elastic compliance follow \( Q_h \propto \chi \). Linear dielectrics with a large variation of compliance or low \( \chi \) do not follow such proportionality, whereas soft organic dielectrics with a large elastic compliance range exhibit in general larger \( Q_h \).\(^22\)

To reflect the electro-mechanical nature of electrostriction, a proxy combining the elastic and dielectric properties has been devised to estimate the amplitude of the hydrostatic electrostrictive response.\(^27\) A “universal” linear relationship\(^22\) was found between \( \log(|Q_h|) \) and \( \log(|s/\varepsilon|) \). We therefore propose to define “classical” electrostrictors as materials consistent with Equation (13), illustrated in Fig. 4.

\[
|Q_h| \approx 2.37 \left( \frac{s}{\varepsilon_0 \varepsilon_r} \right)^{0.59}
\]

(13)

\[
\begin{array}{c}
\text{relaxor ferroelectrics} \\
\text{perovskite ferroelectrics} \\
\text{glass ceramics} \\
\text{linear dielectrics} \\
\text{polymer} \\
\text{Y}_{1-x}x\text{FeO}_3\text{O}_{12nh} \\
\text{Ce}_1-x\text{Gd}_x\text{O}_2 \\
\text{Bi}_{1-x}x\text{Ba}_{1-x}\text{LaTiO}_3 \\
\text{La}_{0.5}\text{Sr}_{0.5}\text{TiO}_3 \\
\text{liquid crystalline graphene} \\
\text{MAPbI}_3 \\
\text{Polymer CNT composites}
\end{array}
\]

FIG. 4. Empirical relation of hydrostatic electrostriction coefficient amplitude (\( |Q_h| \)), elastic compliance \( s \), and dielectric permittivity \( (\varepsilon_0 \varepsilon_r) \). The data for classical electrostrictors are from Ref.\(^{20}\) the blue lines corresponds to Eq. (13) and the yellow area corresponds to values within one order of magnitude of the line. Data point for Gd-doped ceria is from Ref.\(^{28}\) bismuth oxide from Ref.\(^{13}\) LAMOX from Ref.\(^{14}\) liquid crystalline graphene from Ref.\(^{27}\) MAPbI\(_3\) from Ref.\(^{15}\) Polymer CNT composites from Ref.\(^{16}\)

The empirical scaling law (Eq. (13)) qualitatively reflects the nature of most dielectrics. However, the units of \( Q_h \) and \( s/\varepsilon \) are both \( m^4/C^2 \). It would therefore follow
that the prefactor (2.37) is not dimensionless but has to be expressed in \((m^2/C^2)^{-0.59}\). What is more this unit probably depends on how many materials are used to fit Eq.(13), as the slope of the fit would probably change. As a consequence, rather than carrying a physical meaning, Eq.(13) is rather useful as a proxy to evaluate qualitatively the electrostrictive properties of a given material, assuming it is a classical electrostrictor.

The outliers in Fig.4 exhibit electrostrictive properties well beyond the general trend defined by Eq.(13) (at least ten times larger). We therefore propose to only qualify as “giant”, electrostrictors that exhibit an electrostrictive \(Q\) coefficient more than one order of magnitude larger than the expected value from Eq.(13). In Figure 4, they lie above the yellow band.

Eqs.(1) or (3) show that the electrostrictive strain is related to the product of the electrostrictive coefficient \(Q\) by the polarization. Therefore, if the permittivity (hence the susceptibility) is modest and/or saturates at low field, a large electric field will not result in a large polarization and the electrostrictive strain will remain of limited amplitude. The field response (hence the \(M\) coefficient) depends on the capabilities of accumulating charges under electric field and of deforming, which correspond respectively to permittivity \(\epsilon_r\) and elasticity \(s\). We therefore put forward that the amplitude of the \(M\) coefficient is correlated to the product of the elastic susceptibility \(s\) by the permittivity \(\epsilon_0\epsilon_r\), both quantities are in \(m^2/V^2\). Fig.5 demonstrates the general trend of the \(M\) coefficient with respect to the product of permittivity and elastic compliance for the same set of materials as in Fig.4. Eq.(14) estimates numerically this relation from a fit excluding 1 “giant” \(Q\) electrostrictors:

\[
|M| \approx 10^4(\epsilon_0\epsilon_r)^{1.14}
\]  

(14)

As for Eq.(13) the power of \(\epsilon_0\epsilon_r\) being different from 1, the pre-factor would have to be in a unit ensuring the homogeneity of the equation and would probably depend on the number of materials used in the fit. Eq.(14) (as Eq.(13)) is therefore not a physical “law” but a proxy to qualitatively evaluate the electrostrictive properties of very different materials.

Nevertheless, as Eq.(13) was used to put forward the definition of giant electrostrictors based on their \(Q\) coefficient, we propose Eq.(14) as the basis for definition of “giant” \(M\) electrostrictors. Materials lying above the yellow band in Fig.6 are more than order of magnitude larger than the value estimated from Eq.(14) and qualify as “giant” \((M)\) electrostrictors.

Experimental data on strain, permittivity, and elasticity may not have been measured on the sample or under the same experimental conditions. As a consequence, there is a dispersion of the data points in Fig.4 or Fig.5. In addition, the measured strain is always the result of the electrostrictive response and Maxwell stress (Eq.15), the latter resulting from the electrostatic attraction bet-

\[
x_M = -\frac{1}{2} s\epsilon E^2
\]  

(15)

Maxwell stress is always negative (compressive), quadratic in electric field, and its coefficient is half of the product of permittivity and elastic compliance. It may be not negligible, especially in the case of materials with large permittivities (e.g. ferroelectric relaxors) or elastic compliances (e.g. polymers). Whether Maxwell strain has been removed or not from the reported strain is not always explicitly mentioned in the reports, potentially contributing to the dispersion of the experimental results.

Figures 4 and 5 reveal that if most of the “giant” \(Q\) electrostrictors also qualify as “giant” \(M\) electrostrictors, there are exceptions (e.g. the two polymer-based composites). A comparison of the polarization (\(|Q_h|\) and field (\(|M|\) electrostrictive coefficients amplitudes is shown in Fig.6. This relation is not monotonic but rather exhibits a “V”-shape where all values fit into a band of ±1 order of magnitude, including “giant” electrostrictors, with the only exception of liquid crystalline graphene nanocomposites.

II. GIANT ELECTROSTRICTORS

Figures 4 and 5 also show that some “classical” electrostrictors exhibit higher responses than “giant” ones. For example, relaxor ferroelectrics (the workhorse of electrostrictive applications) exhibit \(M\) coefficients comparable or higher than “giant” electrostrictors. This is due to their very large permittivity. Increasing the dielectric properties of electrostrictors is an extensively investigated23 approach to improve \(M\). “Giant” electrostrictors therefore exhibit an anomalously large response rather than an absolutely large response.

Classical electrostrictors have been investigated in several reviews (e.g. see Refs.29 and 30). Here we shall
stresses in excess of 500 MPa to exhibit a giant electrostrictive effect. It can generate trostrictors.

The same year, the hydrostatic polarisation coefficient $Q_h$ of (Y, Nb)-stabilized $\delta$-Bi$_2$O$_3$ was reported to have values spanning several orders of magnitude (from $\sim 10^{-3}$ to $\sim 10^3$ m$^3$/C$^2$ for $Q_h$ and from $\sim 10^{-21}$ to $\sim 10^{-12}$ m$^2$/V$^2$ for $|M|$) but correspond within one order of magnitude to the values predicted by Eqs. [13] or [14].

Following our definitions, there are “not so giant” electrostrictors: $\sim 10^3$ m$^3$/C$^2$, or calculated from available experimental values.

However, these materials were far from being the first ones to be reported as “giant” electrostrictors based on their $Q_h$ values. We have classified the reports mentioned above and earlier ones in three general categories: “giant” electrostrictors, “classical” electrostrictors, and “not so giant” electrostrictors:

- “Giant” electrostrictors exhibit coefficients more than ten times larger than expected from Eqs. [13] or [14].
- “Normal” electrostrictors can exhibit $Q_h$ and $M$ values spanning several orders of magnitude for $Q_h$ and from $\sim 10^{-21}$ to $\sim 10^{-12}$ m$^2$/V$^2$ for $|M|$. The upper limit of effective (longitudinal) piezoelectric coefficient ($Q_{33}$) and the figure of merit for energy harvesting applications. These values are either extracted from the publications or calculated from available experimental values.

A more detailed description of “giant” electrostrictors matching our definitions will be presented below.

A. Ce$_{1-x}$Gd$_x$O$_{2-x/2}$

Gadolinium-doped ceria (CGO) has been applied in the field of solid oxide fuel cells, memristors, sensors...
and electrostrictors.\textsuperscript{13,15} Utilizing acceptor dopants like Gd with a 3+ valence state, the fluoride structure of ceria is stabilized with high ionic mobility. Gd-doped ceria exhibits giant electrostriction both in thin films and ceramics.\textsuperscript{46,47} These electromechanical properties are proposed to originate from the local distortion around oxygen vacancies and its change under applied field.\textsuperscript{47} A model of CeO\textsubscript{2} crystal structure with oxygen vacancies has been proposed to maximize the strain response based on the crystalline orientation.\textsuperscript{39}

Since the doping percentage, grain boundary, grain size, crystalline orientation, electrodes, measuring methods, and excitation frequency strongly influence the value of the electrostrictive coefficients, several examples are presented hereafter to give an overview of the giant electrostriction of Gd-doped ceria.

A Ce\textsubscript{0.8}Gd\textsubscript{0.2}O\textsubscript{1.9} thin film sandwiched between Cr electrodes and deposited on a glass cantilever was the first to be reported as showing giant electrostriction.\textsuperscript{11}

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|c|c|}
\hline
\textbf{Giant electrostrictors} & \textbf{Maximum field (MV/m)} & \textbf{Frequency (Hz)} & \textbf{\(|M| (10^{-18} \text{m}^2/\text{V}^2)\)} & \textbf{\(|Q| (\text{m}^2/\text{C}^2)\)} & \textbf{\(\varepsilon_r\)} & \textbf{\(|d_{\text{eff}}| \text{ pm/V}\)} & \textbf{\(d_{\text{eff}} \cdot g_{\text{eff}} \text{ (10}^{-12} \text{m}^2/\text{N})\)} & \textbf{Ref} \\
\hline
Ce\textsubscript{0.8}Gd\textsubscript{0.2}O\textsubscript{1.9} thin film & 6 & 0.01 & 9 & 157 & 28 & 108 & 47.0 & 11 \\
Ce\textsubscript{0.8}Gd\textsubscript{0.2}O\textsubscript{1.9} thin film & 6 & 0.01 & 16 & 282 & 28 & 192 & 148.7 & 20 \\
Ce\textsubscript{0.8}Gd\textsubscript{0.2}O\textsubscript{1.9} ceramics & 0.374 & 0.15–9 & 21.7 & 3797 & 28 & 16.2 & 1.1 & 22 \\
Bi\textsubscript{2}Nb\textsubscript{2}Y\textsubscript{1.6}O\textsubscript{13.9} ceramics & 1.1 & 20–1200 & 12 & 273 & 24.6 & 26.4 & 3.2 & 13 \\
Bi\textsubscript{2}Nb\textsubscript{2}Y\textsubscript{1.6}O\textsubscript{14.3} ceramics & 1.1 & 20–1200 & 6.9 & 142 & 26 & 15.1 & 1.0 & 13 \\
Bi\textsubscript{2}Nb\textsubscript{2}Y\textsubscript{1.6}O\textsubscript{14.7} ceramics & 1.1 & 20–1200 & 5.5 & 83 & 30 & 12.1 & 0.55 & 13 \\
Bi\textsubscript{2}Nb\textsubscript{2}Y\textsubscript{1.6}O\textsubscript{15.1} ceramics & 1.1 & 20–1200 & 4.7 & 118 & 23.5 & 10.3 & 0.51 & 13 \\
La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} ceramics & 11 & 10 & 1.5 & 20 & 28 & 33 & 4.4 & 14 \\
La\textsubscript{2}MoWO\textsubscript{9} ceramics & 11 & 10 & 1.3 & 5.9 & 48 & 28.6 & 1.9 & 14 \\
MAPbI\textsubscript{3} single crystals & 3.7 & 100 & 730 & |Q\textsubscript{33}|=1266 & 50 & 5402 & 65917 & 15 \\
MAPbBr\textsubscript{3} single crystals & 2 & 10 & 900 & |Q\textsubscript{33}|=1417 & 90 & 3600 & 16264 & 13,15 \\
0.5% CNT polymer composites & Not mention & 100 & 9 × 10\textsuperscript{5} & |Q\textsubscript{33}|=1.2 × 10\textsuperscript{5} & 300 & – & – & 16 \\
rGO/PMDS nanocomposites & Not mention & 0.1 & 4 × 10\textsuperscript{4} & |Q\textsubscript{33}|=280 & 1400 & – & – & 14 \\
Soft PZT-5H piezo ceramics & 2.4 & 0.2 & – & – & 3400 & 750 & 18.7 & 13,15 \\
Hard PZT-8 piezo ceramics & 4.2 & 0.2 & – & – & 1000 & 250 & 7.1 & 13,15 \\
0.9PMN-0.1PT ceramics & 2.5 & 500 & 1420 & 0.055 & 18200 & 1040 & 6.7 & 13,15 \\
\hline
\end{tabular}
\caption{Maximum responses of “giant” electrostrictors}
\end{table}

A combination of constant and alternating voltages with two harmonic waves at 10 mHz and 20 mHz were applied on 400 nm-700 nm thin films to ascertain the electrostrictive nature of the response rather than a random vibration. The curvature of the cantilever was detected through the displacement of laser beam received by a CCD camera. From these measurements, we have calculated other coefficients for Ce\textsubscript{0.8}Gd\textsubscript{0.2}O\textsubscript{1.9} thin film. They are reported in Table I. The influence of the oxygen concentration density was then studied in Ref.\textsuperscript{28} as different amounts of Gd in the thin film change the oxygen vacancies concentration. However, a higher concentration of oxygen does not necessarily result in a larger electrostriction.\textsuperscript{25} No correlation has been found between the oxygen vacancy concentration and the electrostrictive properties.

Bulk ceramics of Gd-doped ceria coated with gold-silver mixture as electrodes were studied in Ref.\textsuperscript{32} With the same concentration of oxygen vacancies as in the
films, different grain sizes or grain boundaries caused by different synthesis methods lead to different electromechanical performances. From Ref\[12\] it appears that the grain size, grain boundary resistivity, and electrostrictive performance are not unambiguously correlated. Moreover, lower frequency (10Hz) excitations may not necessarily lead to higher deformations in ceramics. A representative example of 10%mol-doped ceria ceramic measured at low frequency is reported in Table I.

B. Bi\textsubscript{2}Nb\textsubscript{2−x}Y\textsubscript{x}O\textsubscript{15.5−x}

Cubic bismuth oxide (Bi\textsubscript{2}O\textsubscript{3}) in the δ phase adopts a defective fluorite structure with 25% of the oxygen sites left unoccupied. It is one of the best solid-state ionic conductors (with a conductivity around 0.3S/cm at 800°C). Bi\textsubscript{2}O\textsubscript{3} is stable at room temperature in the α (monoclinic) phase, whereas β-Bi\textsubscript{2}O\textsubscript{3} only exists in the range of 729°C up to melting point 830°C. To stabilize δ-Bi\textsubscript{2}O\textsubscript{3} at room temperature, the Bi\textsubscript{2}O\textsubscript{3}-Nb\textsubscript{2}O\textsubscript{5}-Y\textsubscript{2}O\textsubscript{3} ternary oxide system was selected. Isovalent substitution of Bi\textsuperscript{3+} by Y\textsuperscript{3+} produces the disordered fluorite phase with excellent conductivity. By contrast, substitution of Bi\textsuperscript{3+} by Nb\textsuperscript{5+} exhibits ordered fluorite phases with much lower ionic conductivity.

Similar to Gd-doped ceria, (Y, Nb)-stabilized δ-Bi\textsubscript{2}O\textsubscript{3} exhibits giant electrostriction at room temperature, a behavior proposed to be related to the defective fluorite structure and oxygen vacancies. As mentioned earlier, such hypothesis was then reputed by the discovery of giant electrostriction in La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} and related materials that do not adopt the fluorite structure. With the largest concentration of oxygen vacancies in the investigated Bi\textsubscript{2}O\textsubscript{3}-Nb\textsubscript{2}O\textsubscript{5}-Y\textsubscript{2}O\textsubscript{3} ternary oxide system, Bi\textsubscript{7}Nb\textsubscript{0.4}Y\textsubscript{1.6}O\textsubscript{13.9} was reported\[20\] to have the largest $Q_{33} = \frac{-273}{m^1/C^2}$ and $M_{33} = -1.2 \times 10^{-17} m^2/V^2$ (see Table I). Such values qualify (Y, Nb)-stabilized δ-Bi\textsubscript{2}O\textsubscript{3} as a "giant" electrostrictor as they are thousands of times larger than what would be expected from the empirical scaling law (Eq\[13\]).

C. LAMOX

Lacorre’s team demonstrated La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} as a distinct structural group of fast oxide-ion conductors, which is now used in solid oxide fuel cells and oxygen pumping devices. La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} (with a conductivity of around 0.06S/cm at 800°C) can achieve comparable ionic conductivity comparable to commercial 10% of yttria-stabilized zirconia (YSZ) (≈ 0.1S/cm at 1000°C) but 200°C lower.

This lanthanum molybdate shows an abrupt phase transition from a monoclinic phase (α phase) to a cubic phase (β phase) at 580°C and its ionic conductivity increases by two orders of magnitudes. The slightly distorted monoclinic crystal cell exhibits a large (x2x3x4) superstructure relative to the high temperature cell. Since pure La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} (m-LAMOX in the reference contains intrinsic oxygen vacancies without relying on afovental doping. Li’s group\[13\] has observed giant electrostriction at 10Hz and ambient temperature on ceramics.

Several substitutions can be implemented on both the La and Mo sites of pure La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} in order to stabilize the β (cubic) form at the operating temperature of fuel cell. The β phase of tungsten-doped La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} (La\textsubscript{2}MoWO\textsubscript{9}, c-LAMOX in Ref\[14\]) has also been investigated at ambient temperature and also exhibits "giant" electrostriction. Dopants such as tungsten are believed to change the activation energy barriers in O-vacancy hopping mechanisms. Accordingly, the behavior of giant electrostriction would be decided by the element and concentration of dopants.

A way to consider the high-temperature form of the β-La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} is through its structural relationship with β-SnWO\textsubscript{9}. In Ref\[14\] polarization-electric field (P-E) and strain-electric field (x-E) curves have been measured. Electrostrictive coefficients are deduced by quadratic fits of the polarization vs electric field and strain vs polarization/E-field curves respectively. The magnitude of the electrostrictive coefficients in m- and c-LAMOX are $M_{33} \sim 10^{-18} m^2/V^2$ and $Q_{33} \sim 10 m^2/C^2$ respectively, as shown in Table I. With less symmetry distortion, c-LAMOX is slightly less electrostrictive than m-LAMOX despite its higher permittivity. The occurrence of giant electrostriction in LAMOX demonstrates that this phenomenon can exist in non-fluorite structures.

As a conclusion, giant electrostriction has been measured in ceramics and thin films of ionic conductors. Contrary to what was initially believed, the phenomenon is not confined to the fluorite structure. All of them, though, have intrinsic oxygen vacancies and the current hypothesis is that the global strain developed under electric field is related to the field-response of the large local strain induced by the presence of oxygen vacancies. This would, however, indicate that a larger concentration of oxygen vacancies lead to a larger electrostrictive response (as long as they do not interact), which is not what reports on Gd-doped ceria\[20\] indicate. The relationship between the oxygen vacancy concentration and the overall deformation remains to be unravelled. Another common feature of the electrostrictive response of these materials is that it occurs at lower frequency than “classical” electrostriction (around 10Hz), with a sharp decrease when the frequency of the applied field increases. Such low threshold frequency is consistent with electroactive species heavier than electrons, such as oxygen vacancies. In addition, Gd-doped ceria\[20\] as well as LAMOX\[62\] exhibit anelastic behavior. The nature of the interaction between elastic and electric dipoles therefore appears necessary to the understanding of this phenomenon.
D. Lead halide perovskites

The initial interest in lead halide perovskites laid in photovoltaics, photodetectors, radiation detectors, and light-emitting diodes (LED) due to their high power conversion efficiency (>20%), tunable bandgap, large mobilities of electrons and hole, and low fabrication cost. They have since then also attracted attention for their electromechanical properties.

In Ref.15, three different samples are reported to exhibit electrostrictive responses: inorganic lead perovskite CsPbI$_3$ single crystal (500nm) exhibits a +0.2% strain under 5 MV/m ac bias at 10 Hz ($M = -8 \times 10^{-17} \text{m}^2/\text{V}^2$); organic–inorganic hybrid perovskites (OIHPs) MAPbBr$_3$ single crystal (95μm) exhibits a -0.36% strain under 2 MV/m at 10 Hz ($M = -9 \times 10^{-16} \text{m}^2/\text{V}^2$); organic–inorganic hybrid perovskites MAPbI$_3$ single crystal (40μm) exhibits under 3.7 MV/m at 100 Hz a maximum negative strain reaching -1% with $M = -7.3 \times 10^{-16} \text{m}^2/\text{V}^2$. Their electrostrictive performances are stable either under 1200 ac loops actuation or 60s of square waves. Unlike the previous oxides, lead halide perovskites do not contain oxygen vacancies. It is speculated that lattice deformation is possibly caused by the formation of additional defects.

Several alternative mechanisms to electrostriction were discarded such as Maxwell strain, organic dipoles, local polar fluctuations, or ferroelectricity: the depolarization field to yield Maxwell strain caused by ion accumulation would 527 MV/m, which is two orders magnitude larger than the applied field 3.7 MV/m. The existence of organic dipoles is excluded due to the electrostrictive (rather than piezoelectric) response of CsPbI$_3$. Local polar fluctuations occur at the picosecond or a few hundred femtosecond timescale, which is much faster than the electromechanical response of MAPbI$_3$. The electromechanical performance of MAPbBr$_3$ rules out the contribution from ferroelectricity since MAPbBr$_3$ single crystal has centrosymmetric structure and thus cannot be ferroelectric.

Lead halide perovskite may therefore constitute a distinct group of “giant” electrostrictors as they do not contain (large number of) oxygen vacancies. Their electrostrictive response nevertheless shares with the ones of ionic conductors that are “giant” electrostrictors to occur at relatively low frequency. This suggests that the electro-active species differ from the ones of classical electrostriction. Similarly to ionic conductor-based electrostrictors, anelastic relaxation was reported at 0.48, 2.5 and 6.1 kHz in MAPbI$_3$.

In addition to ceramics and thin films of inorganic or organic-inorganic materials, giant electrostrictive responses have also been reported in polymers and polymer-based composites. Such electrostrictors are reviewed below.

E. Polymer nanocomposites

Flexible electrostrictive materials have numerous applications, e.g. in wearable sensors and MEMS. Inspired by the introduction of dielectric mismatched nanodomains in a dielectric elastomer and electron-irradiated poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] copolymer, a large electrostriction has been observed in a composite of reduced graphene oxide (rGO) embedded in a polydimethylsiloxane (PDMS) matrix. These nanocomposites (rGO/PDMS) combine a “giant” polarization electrostriction coefficient $Q$, the large elastic compliance $s$ from polymers, and a large dielectric contribution due to the surface defects from rGO, which bring about ultra-large field electrostriction coefficient $M$ (see Table I) despite not qualifying them as “giant” $M$ electrostrictors (see Fig.9).

Locally aligned structure of PDMS contains 4.7wt% of rGO flakes with large anisotropy. The nanocomposites have large permittivity (around 1500 at 0.1 Hz), which is attributed to liquid crystals with long-range orientational order. The ordered structure is expected to promote the formation of extended microcapacitors.

The permittivity of pure PDMS slightly decreases as a function of strain. The field electrostrictive coefficient ($M$) and relative permittivity of this pure polymer are around $-1.5 \times 10^{-18} \text{m}^2/\text{V}^2$ and 2.8 at 100 Hz, respectively. In contrast, rGO/PDMS composites exhibit a steep decrease in the permittivity-strain curve. The rGO doping therefore improves the electrostrictive responses based on the Equation 8 and Equation 9. The reported field electrostrictive coefficient is around $-1.2 \times 10^{-14} \text{m}^2/\text{V}^2$ at 0.1 Hz in the permittivity-strain curve. The relative permittivity reaches 1500 without applied strain. The dielectric spectroscopy used in the experiment enables to avoid the contribution of otherwise non-negligible Maxwell stress effects. Indeed, for this nanocomposite, the Maxwell strain coefficient ($\epsilon_\text{max} \epsilon_r / 2$) is $-3.1 \times 10^{-15} \text{m}^2/\text{V}^2$ is only one order of magnitude lower than the electrostrictive field coefficient $M \sim -1.2 \times 10^{-14} \text{m}^2/\text{V}^2$. In contrast, e.g. in bismuth oxide, the Maxwell strain coefficient is $-2.5 \times 10^{-21} \text{m}^2/\text{V}^2$ and negligible compared to the electrostriction coefficient $M \sim -1.2 \times 10^{-17} \text{m}^2/\text{V}^2$. Therefore, applying high voltages to composites will always lead to a strain combining the electrostrictive and electrostatic responses.

Another type of composites investigated for their electrostrictive properties is the carbon nanotubes (CNT)-polymer composites.CNT concentration varies from 0.05wt% to 2wt%, covering the percolation threshold around 1wt%. At 100 Hz, the permittivity of CNT composites rises with increasing concentration of CNT. $M_{33}$ demonstrates the same increasing trend except that the deformation of 2wt% CNT composites switches sign (from negative to positive). The maximum negative $M_{33}$ is obtained at 1wt%, i.e. at the percolation threshold. $Q_{33}$ is in the range of $-6 \times 10^3 \text{m}^4/\text{C}^2$ to $-1.2 \times 10^5 \text{m}^4/\text{C}^2$. In contrast to $M$, the maximum negative
$Q_{33}$ is attained at 0.5 wt%, below the percolation.

As a conclusion, “giant” electrostrictors based on polymers and composites exhibit maximum strains that are much larger than inorganic ones (several percents versus a few 0.1%), field electrostriction coefficient ($M$) are several orders of magnitude larger than their inorganic counterparts but do not qualify them as “giant” electrostrictors as their performance are comparable with the expectation from Eq. [14].

III. INTERESTS OF GIANT ELECTROSTRICTORS

The discovery of functional materials exhibiting responses much larger than anticipated is always of interest. From a fundamental point of view, understanding the origin of this response would enable to engineer dedicated structures to further enhance this property. From an applied point of view, such a large electromechanical response offers the perspective of an alternative to piezoelectric materials that are in majority lead-based. However, before these materials revolutionize the market of electro-mechanical devices, several major advancement needs to be made. We review the most pressing ones below.

A. Fundamental interest

According the prevailing hypothesis, “giant” electrostriction is related to the field-dependence of the local strain induced by oxygen vacancies in inorganic materials. The “active ingredient” would then differ from the electrons of classical electrostriction. The exact origin of electrostriction in inorganic materials remains to be determined before structures can be tailored to harness its full potential.

The relation between oxygen vacancy concentration and electrostrictive properties also needs to be clarified. The observed increase of the response amplitude with oxygen vacancies concentration in (Nb,Y)-doped Bi$_2$O$_3$ [13] is in contradiction with the results on Gd-doped ceria [11]. Such increase may only occur as long as the vacancies do not interact with each other. The exact role of oxygen vacancies in “giant” electrostriction remains to be understood.

The fact that the few materials discovered so far exhibit such a response already vastly surpassing their classical counterparts suggests that more improvement can still be made. Such an improvement lead the piezoelectric perovskites from BaTiO$_3$ ($d_{33} = 85.6$ pm/V single crystal [79]) to PZT ($d_{33} = 750$ pm/V [35]) with a tenfold improvement. If “giant” electrostriction is indeed related to local strain relaxation in the vicinity of an oxygen vacancy induced by an aliovalent dopant, then the height of the energy barrier preventing the oxygen vacancy to jump from one oxygen site to another should be as high as possible. This suggests that even larger electrostrictive properties may be found in oxygen-deficient materials with low ionic conductivity.

As a consequence, the study of dopants having a strong affinity for the oxygen vacancies they induce represents a promising path to increase the electromechanical response.

Another potentially promising route would be to increase the permittivity of these materials. In this way, the large polarization coefficients ($Q$) would translate into large field coefficients ($M$).

It also remains to be clarified whether the other family of “giant” electrostrictors (lead halide perovskites and polymer nanocomposites) present a similar mechanism (local, field-dependent strain) or not. In order to describe and predict such behavior, an appropriate effective medium theory would be of great help.

From this understanding of the material properties that give rise to “giant” electrostrictive responses, the possibility arises to tune structures to exacerbate this response further. Besides, literature survey or databases exploration could point out to materials already available that have not been considered so far for their electrostrictive properties.

B. Interest for applications

Electrostriction is of interest for both actuation and sensing applications [36]. The strain response usually exhibits low hysteresis, is weakly temperature dependent and exists in all dielectrics. In addition, electrostrictive materials do not require poling and therefore exhibit superior fatigue behavior. As such, it offers a potential alternative to piezoelectrics. The modest performances of classical inorganic electrostrictors has so far restricted electrostrictors to niche markets. The discovery of “giant” electrostrictors may trigger a renewed interest in the application potential of these materials. Apart from the amplitude of the response, a major difference between classical inorganic electrostrictors and their “giant” counterparts lies in the cutoff frequency of “giant” electrostriction. Giant electrostrictors exhibit their highest responses in the $10^{-2}$ Hz to $10^3$ Hz range. Their application to high frequency devices is therefore not possible at the present time.

All the “giant” electrostrictors discovered so far contract under electric field, whereas classical relaxors or ferroelectrics usually expand. This means that Maxwell strain will actually be beneficial to these materials, contributing to larger negative strain instead of decreasing positive strains. “Giant” electrostrictors may therefore have larger deformation under the field thanks to this electrostatic attraction.

Transduction coefficient ($d_{33} \cdot g_{33}$) of the piezoelectric energy harvesting materials is another potential field of interest for “giant” electrostrictors. It is difficult to obtain simultaneously high piezoelectric coefficient $d_{33}$ and
low permittivity $\varepsilon$ using doping or composites. “Giant” electrostrictors usually present low permittivities and high effective piezoelectric coefficients, as shown in the Table I. This may provide an avenue to improve the performance of energy harvesting devices.

There are, however, several hurdles to overcome. The first one is not related to the materials themselves but to the electronic environment of these sensors and actuators. Indeed, such systems have been carefully optimised for linear electromechanical responses. The quadratic nature of the electrostrictive response may therefore pose a challenge, unless electrostrictors are used as effective piezoelectrics. This would impose to control their dielectric losses as bias voltages would have to be imposed. Measuring and enhancing their breakdown voltage is another crucial aspect of the applicability of these materials. This is not restricted to “giant” electrostrictors, though, as the effective piezoelectric coefficient of PMN-0.1PT (a “classical” electrostrictor) can reach 1500 pm/V under a bias field of 3.7 kV/cm. This value is 2-3 times larger than the one of PZT ceramics (∼3.7 kV/cm). This value is large rather than necessarily limited to “giant” electrostrictors, even for the same material. For example in Ref.32 electrostrictors are presented by samples obtained with different sintering procedures. The low frequency longitudinal electrostrictive strain coefficient $M_{13}^0$ of these samples vary from $-2.38 \times 10^{-17} \text{m}^2/\text{V}^2$ to $-21.7 \times 10^{-17} \text{m}^2/\text{V}^2$. Such variations suggest that the microstructure of the samples play a role in the response (as is the case for piezoelectrics). A better understanding of the role of the microstructure on the final properties would help designing synthesis methods ensuring more consistent performances.

IV. CONCLUSION

Electrostriction is an electromechanical phenomenon that exists in all dielectrics. In the direct effect, the induced strain or stress is proportional to the square of the polarisation or the electric field (linear dielectrics). Reverse effects exists as well, provided the sample is pre-strained or polarised. This enables both actuation and sensing. A pseudo-piezoelectric effect (linear response at the same frequency as the excitation) can be induced under a bias field much larger than the excitation field.

The applications of electrostriction have so far been restricted to niche markets due to the superior performances of piezoelectrics. The recent discovery of “giant” electrostrictors has the potential to challenge this state.

We propose to define as “giant”, electrostrictors that exhibit electrostriction coefficients at least ten times larger than the value expected either from the empirical relation put forward by Newnham for the polarisation ($Q$) coefficient or the one we propose for the field ($M$) coefficient. The latter is of most interest for applications as it relates the induced strain to the electric field. We also demonstrate that a “giant” $Q$ coefficient does not necessarily ensure a “giant” $M$ coefficient.

From these definitions, we review the existing literature on electrostrictors and classify them as indeed “giant”, “normal”, or “not so giant”. We also underline the fact that the electrostriction coefficients of “giant” electrostrictors are anomalously large rather than necessarily very large in absolute values.

An extensive review of the “giant” electrostrictors enables to underline their common characteristics: the existence of point defects (e.g. oxygen vacancies) and the restriction of the “giant” response to low frequencies. Such a response is not limited to a particular structure nor to the organic, inorganic, or mixed nature of the material. It even extends to composites.

Much work remains to be done to understand the fundamental mechanism or mechanisms at play in “giant” electrostriction. For example, the exact role of point defects, the relationship between their density and the amplitude of the electrostrictive response, the role of elastic dipoles and microstructure etc. need to be clarified to understand how to engineer materials able to exhibit even larger responses.

Such an understanding is a pre-requisite to replacement of too-often lead based piezoelectric materials by environmentally friendlier electrostrictors with superior, reliable, and optimised properties.

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