Field-dependent nonlinear piezoelectricity: a focused review

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ABSTRACT
This contribution presents a multidisciplinary review of the so-called field-dependent nonlinear piezoelectricity. It starts with an introduction that poses the literature analysis framework, through defining this operational (that is often met in practice) piezoelectric field-dependent nonlinearity. Indeed, the latter is a less known phenomenon although it is inherent to stress-free actuation responses of corresponding smart materials, actuators and structures. Then, related experimental observations from piezoelectric materials, actuator devices and smart structures tests are multidisciplinary surveyed for understanding the underlying mechanisms of the encountered field-dependent nonlinearity. Next, empirical material and numerical structural modelling and simulation approaches are critically reviewed from, respectively, the constitutive and finite element analysis points of view. Summary conclusions and few future directions for research are finally provided as a closure. It is worth mentioning that, although it is concise (retains only experiments and experimentally-correlated models and simulations), this critical review covers the last three decades period which is almost the whole age of the piezoelectric materials, actuators and smart structures research field.

1. Introduction

Lead Zirconate Titanate (PZT) piezoelectric ceramics (shortly piezoceramics) are technologically the most important ferroelectric materials [1]. Indeed, they are nowadays the most popularly used materials for sensing, actuation and transduction (energy conversion) of smart metallic, composite or metallic-composite hybrid structures for their shape, vibration, noise and health active control. Piezoceramics can be divided into two types: hard and soft. Within the coercive (depoling) field (E_c) both of them, particularly the latter (soft), and from a threshold electric field (E_t) value they have a nonlinear response (induced strain, displacement and blocking force) with increasing the applied (actuation) electric field (E_a). This phenomenon is known as the field-dependent nonlinearity of piezoelectric materials [2], actuators [3] and structures [4]. It is not an electrostrictive behaviour, as it was experimentally measured at the same frequency for the tested sample and applied field, but a kind of additive response [2]. In general, the occurring onset of this phenomenon (i.e., the threshold field) is material composition, aging time, direct current (DC) bias field, operating temperature and frequency.
(of the applied field) dependent [5]. Therefore, $E_t$ value may vary with the measured output (strain, displacement, blocking force). Although above influences are out of the present scope, they will be given, for information, as test environment conditions. Here, the interest is on piezoelectric materials, actuators and structures responses to increasing static, quasi-static (at very low frequency – LF) or LF harmonic field (or voltage) positive increments. This represents the ascending branch of the so-called minor hysteresis loop around the initial spontaneous polarization state. Indeed, it is the latter that is met under operational electric field that lies generally well below the coercive one (i.e., far below saturation) which is the onset for the major hysteresis loop occurring [6]. In summary, the field-dependent nonlinearity covers here the piezoelectric actuation response under the field range between the threshold value (onset of field-dependence) and a high fraction (up to 90%) of the coercive one (onset of saturation and depolarization nonlinearity).

In most practical smart structures applications, piezoceramic actuators are looked for generating high stroke (displacement) and blocking force that require relatively high electric field actuation (up to 200V/mm, 500V/mm or 1kV/mm for shear, transverse or longitudinal response modes, respectively). Thus, using the standard linear constitutive equations, which consider the electromechanical properties provided by the manufacturers and measured under weak signals (applied field below 10V/mm), becomes questionable and even no longer applicable [7] from the threshold field on. Indeed, it is well recognized from experimental data, particularly for soft piezoceramics, that the electromechanical properties increase considerably with increasing the applied electric field even at sub-switching levels [8]; i.e., they are highly field-dependent [9]. While this piezoceramics nonlinearity can be considered as a limitation for some applications, like high precision positioning, it is also a chance for increasing the performance of other ones, like smart actuator devices [3]. Therefore, it is essential to understand the mechanisms and origin of this nonlinearity, not only from the interesting scientific point of view but also for enhancing and contributing to widen piezoceramics-based smart structures technology [5]. These are the main objectives of the present critical review.

Therefore, hereafter, experimental observations from smart piezoelectric (bulk or composite) materials, actuator (layered) devices and structures (beams, plates, shells) tests are first multidisciplinary surveyed regarding the origin of their field-dependent nonlinear properties and free-stress actuation response mechanisms. Then, available empirical material (constitutive) and numerical (finite element-FE) structural models are critically reviewed with regards to their outputs correlations with experimental measurements. Finally, summary conclusions and few future directions for research are provided. The objective is to direct future investigations toward practical significance of this stress-free piezoelectric actuation field-dependent nonlinearity, and toward developing enhanced or new numerical (FE in particular) handling models that are well correlated with experimental measurements of smart piezoceramics-based actuators and structures usual performance indicators (induced strain, displacement and blocking force).

It is worth mentioning that this focused critical review, although it covers the last three decades (almost the age of piezoelectric materials, actuators and smart structures research field), is intentionally made concise; therefore, only a selection of (the thought key) references, primarily those containing experimental measurements or/and test-correlated models, are considered. Apologize is then made for the non-cited numerous other publications related to this highly important research and engineering topic.
2. Experimental observations

The experimentally visible (even when unnoticed or uncommented) piezoelectric field-dependent nonlinearity in the open research literature is reviewed hereafter separately for piezoelectric materials (bulk or composite), actuator (layered) devices and smart structures (beams, plates, shells). Indeed, beside material issues, actuators packaging or structural integration ones, like surface bonding, embodying techniques or mechanically constraining conditions, may affect the field-dependent nonlinear response key indicators (induced strain, displacement, blocking force).

2.1. Piezoelectric materials

It is well recognized in material science that the piezoelectric effect is composed of two contributions: the intrinsic effect, resulting from the unit cell (single domain, single crystal) homogenous deformation caused by the applied electric field, and the extrinsic effect, due to the elastic deformation resulting from non-180° domain walls [5]. The latter can be further sub-divided into domain wall vibration, usually assumed to contribute solely for electric field levels below around 10V/mm (weak signal), domain wall translation and domain switching [8]. It is also well accepted that the domain wall motion excitation is reversible under a weak signal and irreversible from a threshold electric field smaller than the coercive one [2]. In the latter reference, these experimentally observed phenomena are explained by thermally activated domain wall fluctuations and nucleation. While in [2] it is considered that the reversible domain wall motion contributes to a large part of the weak signal, others [9] consider that remnant polarization irreversible change is caused by both spontaneous polarization reorientation (switching) and hysteresis in the mechanical boundary conditions (BC) for ferroelectric domains under applied electric field. The nonlinearity from the intrinsic contribution is very small and that from the extrinsic one is the main source for relatively low-to-moderate driving level [5].

The above nonlinear response of piezoelectric materials to the applied electric field has been the subject of several experimental researches during the previous two decades. Indeed, in [2], hard (PZT-8) and soft (PZT-5) piezoceramics piezoelectric and dielectric coefficients have been shown to increase with increasing the applied electric field above a certain level (threshold field), defining the nonlinearity onset, of 20 V/mm for soft PZT and 80V/mm for hard PZT. These values were found to be moderately varying in the tested range of [2 Hz – 2 kHz]; for example, above first value was at 400Hz, while it is 10 V/mm at low frequency and without substantial increase at higher frequency [2]. The threshold electric field value is determined graphically or empirically as a few percent (2.5% in [10]) of the relative deviation of the varying property from the weak-signal constant value. The onset of measurable nonlinearity was found to be accompanied by the appearance of hysteresis loops (at 200 Hz and room temperature) [5], a lossy nature that suggests that the field-dependent nonlinearity is generated by the domain wall motion; hence, it comes mainly from extrinsic contributions at relatively low-to-moderate driving field level.

In analogy to the stress-strain elastoplastic nonlinear response, the hard (PZT-4) and soft (PZT-5A) piezoceramics field-dependent one was divided into three regions: a first (I) linear region, a second (II) region with increased response sensitivity, nonlinearity and
hysteresis, and a third (III) region of saturation and depoling [11]. For example, region II spans from 15 V/mm to 500 V/mm for PZT-5A and from 100 V/mm to 1300 V/mm for PZT-4, representing around $0.37E_c$ ($E_c = 1350V/mm$) and $0.93E_c$ ($E_c = 1400V/mm$) respectively. It is also claimed in [11] that the changes in the response and loss with the driving field amplitude are mainly due to the irreversible process and hysteresis effect in the materials, which are different from a nonlinear process, and to the nonlinear effect. The latter refers to the nonlinear single value relationship between a material property and a driving field. Besides, the contributions to the piezoelectric materials properties are here grouped into three sources: the *intrinsic* contribution (*always reversible*), the contribution from the *domain* boundary motion and that from the *phase* boundary motion; the latter two ones were judged not well understood while they are the cause of most observed nonlinearities and hysteresis [11].

As summarized in Table 1, the field-dependent piezoelectric nonlinear response was experimented for both hard and soft piezoceramics, as discussed above, and separately for their three response modes: longitudinal, transverse and shear. Other ferroelectric materials have been also investigated such as lead-free [2], single crystals [10], electrostrictive [10,11], thin films [1] and copolymers [11]. It can be noticed that the most studied response mode is the longitudinal one; also, the temperature influence has not been well investigated.

### 2.2. Actuator devices

Piezoceramic actuators show a nonlinear relationship between the applied electric field and resulting actuation strain. Within the depoling limits, this field-dependent nonlinearity was considered the most evident among the identified four nonlinearities: piezoceramic material depoling, *transverse* ($d_{31}$) coupling constant dependence on strain (*strain-dependence*), field-strain hysteresis and frequency dependence and creep [13]. Later, the *transverse shear* ($d_{15}$) coupling constant dependence on the applied electric field (*field-dependence*) was shown experimentally for a shear-mode piezoceramic actuator within a micro droplet ejecting system [14]. Recently, the nonlinear relation between the statically measured rate of twist and applied voltage was observed experimentally for both *in-plane shear* ($d_{36}$) [15] and transverse shear-induced *torsion* [16] lead-free single crystal actuators. This nonlinearity, attributed to partial depoling and domain wall motion, was taken into account by an updating of the FE model. The simulation approach was not explained, but nonlinear curves of $e_{36}$ [15] and $e_{15}$ [16] versus applied electric field were embedded (without explanations) in the rate of twist versus applied

| Reference | Response | Material(s) | Coefficients$^a$ | Frequency | Temperature |
|-----------|----------|-------------|------------------|-----------|-------------|
| [1]       | Longitudinal | PZT (home composition) | $\varepsilon_{33}, d_{33}$ | 20Hz-100 kHz | Not indicated |
| [2]       | Longitudinal | PZT-8, PZT-5, PLZT | $\varepsilon_{33}, d_{33}$ | 2Hz-2kHz | Not indicated |
| [3]       | Shear | PZT-5A | $\varepsilon_{11}, d_{15}$ | 10Hz | 77 < T(K) < 303 |
| [5]       | Longitudinal | PZT-5A | $\varepsilon_{33}, d_{33}$ | 200Hz | Room |
| [9]       | Transverse | PZT-5A | $s_{11}, \varepsilon_{33}, d_{31}$ | 10Hz-10kHz | Room |
| [12]      | Longitudinal | PZT (home composition) | $Y_y, \varepsilon_{33}, d_{33}$ | 1Hz, 100Hz | Not indicated |

$^a$ $\varepsilon$: dielectric constant, $d$: strain piezoelectric constant, $s$: elastic compliance, $Y$: Young’s modulus
voltage curves. Shear ($d_{15}$) MFC actuator experimental response was also shown to be nonlinear, as illustrated in Figure 1 [17].

Table 2 summarizes the characteristics of reviewed actuator devices field-dependent nonlinearity experiments. Noticeably, the longitudinal response ($d_{33}$) has not been investigated for the field-dependent piezoelectric nonlinearity.

| Reference | Actuator | Material | Response | Coefficient | Nonlinearity |
|-----------|----------|----------|----------|-------------|--------------|
| [13]      | Bender   | PZT G-1195 | Transverse | $d_{11}$ | $d_{11}$ vs. strain |
| [14]      | Shear    | Soft PZT (home-made) | Shear | $d_{15}$ | $d_{15}$ vs. $E_a$ |
| [15]      | Torsion  | BZT-BCT single crystal (Lead-free) | In-plane shear | $d_{36}$ | $e_{36}$ vs. $E_a$ |
| [16]      | Torsion  | NBT-BT-Mn piezoceramic (Lead-free) | Shear | $d_{15}$ | $e_{15}$ vs. $E_a$ |
| [17]      | Shear    | PZT-5A1   | Shear | $d_{15}$ | Displ. vs. $E_a$ |

Table 2. Characteristics of reviewed actuator devices field-dependent nonlinearity experiments.

2.3. Smart structures

The feasibility of integrating transverse ($d_{31}$)-mode piezoceramic (PZT G-1195) co-localized actuator pairs into metallic (aluminium) and laminated composite (glass/epoxy, graphite/epoxy) beam coupons has been investigated three decades ago analytically, numerically (Rayleigh-Ritz) and experimentally (manufacturing, modal analysis and static traction testing). Both surface-bonded and embedded configurations were considered [4]. From the three smart structure types, only the piezoceramics-embedded graphite/epoxy composite smart beam has provided a linear response under increasing applied voltages. The other two smart beams have provided rather nonlinear deflection-voltage curves, showing a reasonable model-test correlation for the aluminium beam with a symmetrically surface-bonded actuators pair, but a strong deviation between the linear model and experimental data of the piezoceramics-embedded glass/epoxy composite smart beam. Glass/epoxy composite plates with surface bonded transverse mode piezoceramic patches, showed also a 30–35%
maximum discrepancy between the nonlinear deflection-voltage experimental and numerical (ANSYS® 2D plane strain FE) curves [18]. The integration of longitudinal (d33)-mode active fibre composite (AFC) piezoflex® patches into plain weaved carbon/epoxy cross ply laminated composite beams was investigated experimentally and numerically (ANSYS® 3D Solid5 FE) [19]. The displacement-voltage test-model correlation was also bad.

The integration of transverse shear (d15)-mode piezoceramic (PZT PIC255) patches segmented layer between glass/epoxy composite faces in plate [20,21] and beam [22] smart structures has been investigated for shear [20] and torsion [21,22] actuation purposes. For each configuration, the experimental displacement – voltage relationship was found strongly nonlinear and non-catchable by linear FE models (AB AQUS® C3D20E FE), as illustrated in Figures 2, 3 and 4. The latter figure shows, in particular, a partial (due to applied positive actuation voltages only) minor hysteresis under static loading but not under quasi-static one. This proves that the hysteresis is mainly a static phenomenon.

3. Modelling and simulations

The physical, phenomenological or empirical modelling of piezoelectric materials field-dependent nonlinear behaviour and the FE numerical simulation handling of such nonlinearity for piezoelectric materials (bulk or composite) actuator (layered) devices and smart structures (beams, plates, shells) are hereafter reviewed separately. Indeed, the models are often developed for specific materials or applications, like for simulating the polarization process, specific actuators or smart structures applications.

3.1. Piezoelectric materials

Usual (standard, manufacturer) weak signal measured electromechanical coefficients of piezoelectric materials are no longer descriptive of their behaviour beyond the nonlinearity threshold field [2]. Indeed, it is well proved experimentally that the piezoelectric and dielectric coefficients increase with increasing the applied electric field [9]. This field-dependence has

Figure 2. Experimental vs. simulated transverse deflection of a cantilever sandwich plate made of two same-axially poled shear (d15) actuation core and glass/epoxy composite faces [20].
been modelled according to the thought mechanism behind such nonlinearity and the investigated piezoelectric response mode.

One of the field-dependent piezoelectric nonlinearity models uses a Rayleigh law, derived from thermodynamic theory, for the piezoelectric longitudinal response, as [8] (see for example [23] for the dielectric response):

\[ d(E) = d_i + \alpha_p E \]  

(1)
Where $d_i$ is the initial strain (charge) piezoelectric coupling coefficient, $a_{cp}$ is the Rayleigh coefficient associated to the converse piezoelectric ($cp$) effect. Nevertheless, this linear relationship between the piezoelectric coupling coefficient, $d$, and applied AC electric field amplitude, $E$, gave poor fit to experimental data of various transverse response mode soft piezoceramics [9]. Thus, in the latter reference, a new mathematical model, in which the ceramics behave like a hysteretic transducer, has been proposed; it gives the Rayleigh law as a particular case. In [9], it is assumed that the electromechanical properties observed hysteretic changes are due to the field-dependency of the mechanical stress acting at the inter-domain boundaries in the partially constrained crystallites of the ceramics. It was found [9], using an electric pulse technique, that irreversible changes in the remnant polarization take place even at electric fields much smaller than the coercive one. This approach was presented as an alternative to that based on the 90° reorientation (see for example [5]) and tetragonal/rhombohedral phase boundary motion. It defends the hypothesis that the high field behaviour of soft piezoceramics is caused by hysteresis, resulting from the polarization reorientation (switching) [9].

Higher-order polynomial piezoelectric coupling coefficients relationships with the applied AC electric field amplitude were proposed for the transverse and shear response modes of hard and soft piezoceramics by fitting their experimental data, for example for soft PZT EC65 [24]:

$$d_{15}(E) = 560 + 1170E - 205E^2 + 142E^3$$  \hspace{1cm} (2a)

$$d_{33}(E) = 455 + 529E - 153E^2 + 43E^3$$  \hspace{1cm} (2b)

$$d_{31}(E) = 192 + 312E - 126E^2 + 12E^3$$  \hspace{1cm} (2c)

It is thought [24] that the experimentally observed increase in the strain piezoelectric coefficients is due to the larger extrinsic contribution caused by the increased domain switching under larger fields. It is then considered that the observed nonlinearities are stronger than what can be predicted by the electric field-linear Rayleigh law.

As alternative to above polynomial representation of the field dependent nonlinearity, a general power law, having the Rayleigh one as a particular case, was found to be more suitable for describing the soft piezoceramics shear response mode field-dependent non-linear behaviour above $E_t$ [25]:

$$d_{15}(E) = d_i[1 + (\gamma E)^a]; \ a \approx 1.2$$  \hspace{1cm} (3)

Where $\gamma$ is a soft PZT dependent nonlinear parameter. Here ([25]), it is thought that the experimental results indicate that the irreversible motion of the non-180° walls causes the piezoceramics nonlinearity, while the contribution of the 180° walls to the linear and nonlinear coefficients is negligible. Thus, it is considered that the nonlinear contribution to the soft piezoceramics total strain response plays a major role at applied fields well below the depoling (coercive) one [25]. Further experiments, including also the transverse response mode [26], showed that above power law remains valid for the shear response mode within tested temperature range of $[77K - 303K]$. However, for the transverse response mode [3], the power parameter was found unity ($\alpha = 1$),
corresponding to the particular case of Rayleigh law (linear in $E$). It is worthy to mention that the above multiplicative power law has been rather used in the last two references in this additive form [3]:

$$d_{15}(E) = d_i + (\gamma E)^\alpha; \quad \alpha \approx 1.2 \tag{4}$$

Nevertheless, recent unpublished comparisons by the present author of the multiplicative (3) and additive (4) forms for modelling the measured nonlinear shear strain piezoelectric coupling coefficients of $x$-polarized PZT PIC255 (soft) and Macro-Fibre Composite (MFC) patches have shown that the additive form is not performant, while the multiplicative one was satisfactory.

Field-dependent and strain-dependent quadratic polynomial laws have been proposed [27] for length shear response mode of hard and soft length poled piezoceramics piezoelectric and dielectric, and elastic coefficients, respectively. Hence, for the effective (eff) shear strain piezoelectric coupling coefficient, this law was:

$$d_{15}(\text{eff}) = d_{15} + d_{1115}E^2 \tag{5}$$

As summarized in Table 3, the field-dependent piezoelectric nonlinear response was modelled using phenomenological, empirical or thermodynamic approaches; both hard and soft piezoceramics were considered. However, the transverse response mode was less investigated.

It is worthy to mention that the power law (3) appears as a universal model as it reduces to the Rayleigh law (1) and phenomenological one (5) for particular integer powers (1 and 2, respectively). However, the fitting polynomial laws (2) can be used only for the specific material (PZT EC65) for which they were developed.

### 3.2. Actuator devices

Linear piezoelectric coefficients, determined at a low measuring voltage, are considered insufficient for describing the piezoceramics-based actuator devices performance in

| Reference | Response  | Material(s)                | Coefficient(s) | Model                                      |
|-----------|-----------|----------------------------|----------------|--------------------------------------------|
| [1]       | Longitudinal | PZT (home composition)     | $d_{33}$       | Phenomenological approach, Rayleigh law (1) |
| [3]       | Shear     | PZT-5A                     | $d_{15}$       | Additive power law (4)                     |
| [5]       | Longitudinal | PZT-5A                     | $d_{33}$, $d_{31}$ | Phenomenological theory based on 90° wall motion, Physical model |
| [6]       | Transverse | PZT-5A                     | $d_{31}$       | Mathematical model of a hysteretic transducer |
| [9]       | Transverse | PZT-5A                     | $d_{31}$       |                            |
| [24]      | Longitudinal | PZT EC65                  | $d_{33}$       | Empirical cubic polynomial (2)             |
| [25]      | Transverse | Soft: N-10, N-21, 3203HD, PZT-5A, PZT-SA, Hard: PZT-4, PZT-8 | $d_{31}$, $d_{15}$ | Empirical multiplicative power law (3)   |
| [26]      | Transverse | N-21, 3203HD, PZT-5H       | $d_{33}$       | Empirical multiplicative power law (3)   |
| [27]      | Shear     | N-10, N-82                 | $d_{31}$       | Thermodynamic quadratic polynomial law (5) |

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many applications [25]. This is the case, in particular, for piezoelectric stack actuators that show a pronounced nonlinear displacement–voltage relationship. To accommodate nonlinearities present at high regime, a phenomenological modelling, based on experiments curve fitting (cubic regression) approach was proposed in [28]. Later, a combination of the theory of electro-elasticity [29] and an experimentally identified Preisach model that takes into account the nonlocal (macroscopic) memory of the piezoceramic was suggested [6]. The resulting piezo-ferro-elasticity extends [29] to arbitrary polarization in the ferroelectric domain. Here ([6]), the only source of nonlinearity was considered the hysteresis between the electric field and polarization. Besides, the focus was made on the minor hysteresis loops around the initial spontaneous state of polarization. Preisach model, representing the ferroelectric hysteresis only and restricted to 180° domain-wall effect, was also used within a geometrically and materially nonlinear piezoelectric three-dimensional beam FE including warping effect [30]. However, it was not successful for representing the experimental nonlinear response of the helical spring bimorph actuator in [31]. It was argued that the used Preisach function does not exactly correspond to the physical hysteresis of the PZT-5H material; an experimentally determined one might be better. It is worth mentioning that the Preisach hysteresis operator is purely phenomenological model that is not thermodynamically consistent [32]. It is considered superior to other models due to its fast evaluation, capability to fully describe minor hysteresis loop and well-established procedures for its updating to measured data [33].

The static nonlinear response of MFC longitudinal-response actuator to monotonically increasing voltage was represented [34] through an experimentally identified nonlinear piezoelectric constitutive model from the Gibbs thermodynamic potential following [35]. Therefore, a quadratic representation of axial and transverse strains in term of the electric field was obtained which coefficients were experimentally fitted from quadratic regressions:

\[ \varepsilon_1 = d_{31}E_3 + \frac{1}{2}d_{133}E_3^2 \]  

\[ \varepsilon_3 = d_{33}E_3 + \frac{1}{2}d_{333}E_3^2 \]  

(6a)

Shear \((d_{15})\) MFC experimental nonlinear actuation response [17] was modelled via a nonlinear FE homogenization that considers the power law (3), with PZT-5A assumed parameters, for the shear coupling coefficient of the PZT fiber [36]. The correlation with experiments [17] was only qualitatively good, as illustrated in Figure 5. However, when the power law (3) parameters were identified from the tests, using either parametric analysis [37] or Levenberg-Marquardt-Fletcher algorithm [38]-based optimization [39], the correlation was quantitatively very good, as shown in Figure 6.

A quasi-static model based on polarization hysteresis loop and butterfly curve of its piezoelectric material was proposed for the analysis of a double clamped three-layer bimorph actuator [40]. The nonlinear behaviour was considered through a Preisach operator and Landau free energy was incorporated in the total energy density of the polarization. However, while the model seems to fit relatively well the hysteresis loop, it does not represent well the butterfly one. In addition, the displacement versus voltage
nonlinear relationship correlation with experiment was qualitatively, but not quantitatively, good. It is worthy to mention that unrealistic assumptions were retained for the materials, like isotropic behaviour for a piezoceramic material and nil Poisson’s ratio for all materials. The present author thinks that this may explain, at least partially, the bad correlations with experiments from [41].

Late in the previous decade, a simple phenomenological model of vibrating domain wall has been implemented successfully, within the commercial FE code ANSYS®, using the latter’s linear constitutive equations in order to consider the contribution of domain wall motion to the electromechanical properties of functionally graded cantilever piezoceramic bimorph actuators [42].
Table 4. Characteristics of reviewed actuator devices field-dependent nonlinearity models.

| Reference | Actuator | Material | Response | Coefficient | Model |
|-----------|----------|----------|----------|-------------|-------|
| [6]       | Stack    | PZT-SH (BMS32) | Longitudinal | $d_{33}$ | [29]+ Preisach |
| [28]      | Stack    | Soft PZT | Longitudinal | $d_{33}$ | Curve fitting |
| [30]      | Bimorph spring | PZT-SH | Transverse | $d_{31}$ | Preisach |
| [34]      | Bender   | MFC      | Longitudinal | $d_{33}$ | Electro-elasticity [35] |
| [36,37,39]| Shear    | MFC      | Shear     | $d_{15}$ | Power law (3) |
| [40]      | Tri-morph | PZT      | Transverse | $d_{31}$ | Polarization hysteresis & butterfly [41] |
| [42]      | Bimorph  | PZT C-91 | Transverse | $d_{31}$ | Phenomenological (vibrating domain-wall) |

Table 4 gathers the proposed models for simulating actuator devices nonlinear responses. Most of them are based on the domains switching mechanism.

3.3. Smart structures

The first approach for modelling the nonlinear bending/twisting strain/displacement responses under increasing applied electric field of transverse mode piezoceramic (PZT G-1195) patches between aluminium and graphite/epoxy unidirectional (UD) laminated plates was based on the induced/actuation strain concepts [43]. The nonlinearity was attributed not to the electric field-dependence but to the strain-dependence of the transverse ($d_{31}$) coupling coefficient of the actuator piezoceramic material:

$$\begin{bmatrix} A_x \\ A_y \end{bmatrix} = \begin{bmatrix} d_{31}(\varepsilon_x) \\ d_{32}(\varepsilon_y) \end{bmatrix} E_3$$

(7)

Where the induced strain-dependent piezoelectric coupling coefficients were determined from quadratic curve fitting of strain versus field data from unconstrained piezoceramics. The nonlinearity modelling procedure is iterative. For the free BC, the proposed Rayleigh-Ritz exact solution matched well the longitudinal and transverse strain measurements under varying applied electric field using the strain-dependent coupling coefficient but not using the field-dependent one. However, the present author thinks that this is expected as the field-dependence has to be considered only for modelling the nonlinear responses, which is not the case here as the shown response of the aluminium substrate was linear. Noticeable is the fact that transverse strain vs. applied field was nonlinear while the longitudinal one was linear. On the other hand, for cantilever BC, bad correlation between the model and tests for all test articles was obtained for the longitudinal bending. The present author thinks then that it would be interesting to try the field-dependence for these composite substrates nonlinear responses. This strain-dependence of the transverse coupling coefficient, identified from unconstrained piezoceramics free actuation tests, was used since this work [43] as a reference by many other researchers [44–49]. In particular, the type of nonlinearity was considered weak in [44] as the nonlinear actuation effect was found to be closely related to the substrate stiffness; the lower it is, the higher is the nonlinear induced strain effect. However, this approach was later considered non-consistent by [50]. Instead, a rotationally invariant linearized then specified to polarized piezoceramic plates recoverable (cannot account for hysteresis effects) electro-elasticity theory was presented, then used for satisfactory simulating...
data in [13] for a stress-free thin plate of thickness polarized and electroded PZT G-1195 via this cubic strains–field relation:

\[ S_1 = S_2 = d_{31} E_3 + \frac{1}{2} \beta_{31} E_3^2 + \frac{1}{6} \gamma_{331} E_3^3 \]  

(8)

With the known \( d_{31} \) (nm/V) and measured data (Figure 6 of [13]) least squares fitted higher order parameters \( \beta_{31} = 0.8054 \text{ pm}^2/\text{V}^2 \) and \( \gamma_{331} = -0.7754 \text{ fm}^3/\text{V}^3 \). As the test-model correlation was good, although not considering the hysteresis effect, it can be here concluded that this field-dependence nonlinearity is not due to the piezoceramic material hysteresis. Nevertheless, this result can be expected as it used PZT G-1195 hard piezoceramics which hysteresis is known to be low and, thus, can be neglected.

4. Summary conclusions and future directions

Experimental observations and modelling for the simulation of piezoelectric materials, actuator devices and smart structures nonlinear strain/displacement versus increasing electric field/voltage responses were multidisciplinary reviewed. It appears that, according to, respectively, physics or structure scientists, this type of nonlinearity is seen as an electric field- or induced strain-dependence of the piezoelectric coupling coefficients. The universal power law (3) can model the piezoelectric coefficients field-dependence as it was already proved experimentally for shear monolithic piezoceramics and MFC, although its use for transverse [51] and longitudinal [34] MFC remains to be correlated with experimental measurements or implemented, respectively. On the other hand, the piezoelectric coefficients induced strain-dependence handling for smart structures relies on the strain-field curve fitting-based relationships (7) for unconstrained piezoceramics. However, this approach performance is qualitative only.

For piezoelectric actuator devices, material scientists rely more on hysteresis operators [32,33], and polarization switching [52–54] for handling this type of nonlinearity. For this purpose, in analogy to plasticity, the constitutive equations use the decomposition (additive split) of the strains and electric displacements into reversible and irreversible parts and the piezoelectric coefficients are considered remnant polarization magnitude dependent [55].

It is also clear from the present critical review that neither of the above mentioned approaches is successful for all scales (material, actuator device, structure). It is then believed here that a multidisciplinary approach that combines different modelling techniques from different research communities shall be the key issue for successful modelling of this type of nonlinearity. For example, within a linear FE simulation approach, the so-called third-order material properties tensors [56], here the piezoelectric, often used in electro-acoustic community can be investigated:

\[ e^1_{mpl} = e_{mpl} + l_{mpl}E_q^1 + e_{mplq}S^1_{qr} \]  

(9)

These effective (or changed) coefficients are field- and strain-dependent. They generalize above separately considered field or strain-dependences. However, they are incomplete and inconsistently derived (rotationally invariance not respected) [57]. Nevertheless, despite this theoretical shortcoming, recent use of third-order elastic constants for designing implantable SAW (Surface Acoustic Wave) sensors was found to be very beneficial for good test-model (FE) correlation [58], confirming the good potential of this earlier mentioned approach (9) for handling field-dependent nonlinear piezoelectricity.
As a closure, it is worth mentioning that this focused multidisciplinary critical review was intentionally limited to stress-free actuation nonlinear response. Mechanically loaded or pre-stressed actuators exhibit also field-dependent elastic coefficients \([12,59]\). Besides, the resonant frequency of bending vibrations can be electric field-dependent \([60]\). This issue was also not considered in this review. Together with the environmental temperature effect \([11,58]\), these loading parameters complexities play an important role in medium to large driving field.

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No potential conflict of interest was reported by the author.

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