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Explicit nonlinear homogenization for magneto-electro-elastic laminated materials

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ABSTRACT

In this work we propose an explicit procedure for the homogenization of laminated magneto-electro-elastic nonlinear materials. It means that we determine the effective response of a multilayered structure composed of materials with an arbitrarily nonlinear and anisotropic coupled behavior. In order to obtain a general theory, we take into consideration an arbitrary lamination direction, which is useful to exploit the anisotropic character of components. This technique is characterized by closed form expressions, which can be simply implemented through the basic operations of tensor calculus. To conclude, we discuss some particular cases and various applications.

1. Introduction

The magneto-electro-elastic coupling has been observed in a large class of pure substances and composites with several promising applications in material science and nanotechnology (Spaldin and Fiebig, 2005; Eerenstein et al., 2006; Ramesh and Spaldin, 2007; Nan et al., 2008). As a matter of fact, they exhibit a remarkable cross-coupling between the electric and magnetic ferroic orders (Kimura et al., 2003; Garcia et al., 2010). However, while single phase magneto-electric materials offer a weak coupling (at very low temperature), heterogeneous systems show a stronger interaction at room temperature (Lawes and Srinivasan, 2011). In these composite materials, the coupling between polarization and magnetization is mechanically mediated through the magnetostrictive and piezoelectric behavior of components. Different microstructures have been proposed and thoroughly analysed in recent literature (Ramirez et al., 2006; Kuo and Pan, 2011). Presently, magneto-electro-elastic heterostructures are strongly indicated for achieving low-power systems (Wang et al., 2010). For instance, the electrical/mechanical reorientation of the magnetization dissipates a very small amount of energy and it is appropriate for memories, spintronics and new paradigms of computation (D'Souza et al., 2011; Roy et al., 2011; Tiercelin et al., 2011; Giordano et al., 2012, 2013; Dusch et al., 2013).

To determine the effective properties of heterogeneous materials and structures several nanomechanical techniques and homogenization schemes have been developed and discussed in the literature (Milton, 2004; Torquato, 2002). The first procedures have been worked out for linear systems and, at a subsequent time, many others have been elaborated for the nonlinear case. The standard approach for analysing dispersion of particles is based on the Eshelby theory (Eshelby, 1957) and it leads to a series of linear homogenization schemes largely utilized in the scientific community (Asaro and Lubarda, 2006; Mura, 1987; Giordano, 2003, 2007; Giordano and Palla, 2008). These linear techniques have been generalized in order to take into account the coupling of different physical quantities: the Eshelby theory has been updated for considering the magneto-electro-elastic behavior (Huang and Kuo, 1997; Huang et al., 1998) and, subsequently, the effective properties of multiphysical composites have been determined as well (Shi et al., 2004; Corcolle et al., 2008; Huang et al., 2009; Koutsawa et al., 2011). A methodology based on the Eshelby heritage has been introduced for analysing the effective nonlinear properties of dispersions of dielectric (Giordano and Rocchia, 2005, 2006) and elastic (Giordano et al., 2008, 2009; Palla et al., 2010; Colombo and Giordano, 2011; Giordano, 2013) nonlinear particles embedded in a linear matrix. More general results, based on variational principles for nonlinear materials, have been obtained by Talbot and Willis (1985). Variational methods for deriving improved bounds and estimates for nonlinear media can be found in a complete review by Ponte Casta neda and Suquet (1998). These theoretical approaches play a central role for evaluating the effective coupled response and, especially, the equivalent magnetoelectric coefficients of

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heterostructures of technological relevance. An exhaustive description of the theoretical modelling for magnetostrictive-piezoelectric systems can be found in the review by Bichurin et al. (2010, 2010).

In this paper, we elaborate a theoretical methodology for determining the linear and nonlinear effective properties of multilayered magneto-electro-elastic heterostructures. From the historical point of view, the first pioneering results concerning the homogenization of laminates were obtained by Backus for the pure elastic case (Backus, 1962) and by Tartar for the pure dielectric one (Tartar, 1979). They developed a general method for determining the linear effective tensors in the absence of physical couplings and for a fixed lamination direction. See, for instance, Eqs.(9.7) (Tartar) and (9.9) (Backus) in the classical textbook by Milton (2004). More recently, some approaches have been proposed to deal with fully coupled linear magneto-electro-elastic laminates (Kim, 2011; Challagulla and Georgiades, 2011; Bravo-Castillero et al., 2008). Here we generalize the Backus–Tartar idea in order to obtain the overall properties of nonlinear and fully coupled laminated materials. We develop a self-consistent fully algebraic technique based on the definition of some *ad hoc* operators, which allow to explore the effects of the orientation of the interfaces on the effective linear and nonlinear response of the system. We remark that all the achievements of the present paper can be also used in dynamic regime if we consider the wavelength of a propagating wave (perpendicularly to the layers) much larger than the thickness of the layers. In this case we are working in the so-called quasi-static regime and any layer feels a nearly static applied field.

The structure of the paper is the following. In Section 2, we introduce the formalism used to describe nonlinear materials with a coupled behaviour. In Section 3, we obtain the linear and nonlinear response of the laminated geometry with a fixed lamination direction. Next, we perform the generalization to an arbitrary orientation of interfaces in Section 4. Finally, we present some particular cases and specific applications in Section 5. We discuss the agreement with known results for the linear response and new extensions for the nonlinear one.

2. Multiphysics formalism

In this section we give a brief outline of the generalized variables used to describe the behavior of coupled nonlinear materials. For the linear thermo-magneto-electro-elasticity all possible combinations of variables have been thoroughly discussed by Pérez-Fernández et al. (2009).

Here, in order to take into account all possible nonlinear couplings among electric, magnetic and elastic quantities we consider the time variation of the total energy density (Landau et al., 1984; Landau and Lifshitz, 1986)

$$\frac{\mathrm{d}u}{\mathrm{d}t}=T_{ij}\frac{\mathrm{d}\epsilon_{ij}}{\mathrm{d}t}+E_i\frac{\mathrm{d}D_i}{\mathrm{d}t}+H_i\frac{\mathrm{d}B_i}{\mathrm{d}t},\tag{1}$$

where we have introduced a nonlinear energy function $u=u(\hat{\epsilon},\vec{D},\vec{B})$. Here T_{ij} represents the Cauchy stress tensor, ϵ_{ij} the infinitesimal strain tensor, E_i and H_i the electric and magnetic fields and, finally, D_i and B_i the electric and magnetic inductions. From Eq. (1) we immediately obtain the constitutive equations in terms of the energy function u (Type 2 formulation in Pérez-Fernández et al. (2009))

$$T_{ij}=\frac{\partial u}{\partial\epsilon_{ij}},\quad E_i=\frac{\partial u}{\partial D_i}\quad\text{and}\quad H_i=\frac{\partial u}{\partial B_i}.\tag{2}$$

Alternatively, we can define the free energy density $w(\hat{\epsilon},\vec{E},\vec{H})$ through a Legendre transform of the electromagnetic field (Landau et al., 1984; Landau and Lifshitz, 1986)

$$w(\hat{\epsilon},\vec{E},\vec{H})=u(\hat{\epsilon},\vec{D}(\hat{\epsilon},\vec{E},\vec{H}),\vec{B}(\hat{\epsilon},\vec{E},\vec{H}))-\vec{D}(\hat{\epsilon},\vec{E},\vec{H})\cdot\vec{E}$$
$$-\vec{B}(\hat{\epsilon},\vec{E},\vec{H})\cdot\vec{H}.\tag{3}$$

From Eq. (3) we simply obtain the constitutive equations in terms of the free energy function w (Type 10 formulation in Pérez-Fernández et al. (2009))

$$T_{ij}=\frac{\partial w}{\partial\epsilon_{ij}},\quad D_i=-\frac{\partial w}{\partial E_i}\quad\text{and}\quad B_i=-\frac{\partial w}{\partial H_i},\tag{4}$$

which are nonlinear relations between $(\hat{\epsilon},\vec{E},\vec{H})$ and $(\hat{T},\vec{D},\vec{B})$. Now, in order to obtain a very compact formalism we introduce the modified Voigt notation (sometimes called Mandel or Kelvin mapping) for the elastic quantities

$$\vec{\tau}^T=\left(T_{11},T_{22},T_{33},\sqrt{2}T_{23},\sqrt{2}T_{13},\sqrt{2}T_{12}\right),\tag{5}$$

$$\vec{\varepsilon}^T=\left(\epsilon_{11},\epsilon_{22},\epsilon_{33},\sqrt{2}\epsilon_{23},\sqrt{2}\epsilon_{13},\sqrt{2}\epsilon_{12}\right),\tag{6}$$

which is very efficient since norms and scalar products are preserved. Moreover, contrarily to the standard Voigt notation, stress and strain are treated identically. The Mandel or Kelvin mapping can be also written as $\tau_p=(\delta_{ij}+\sqrt{2}(1-\delta_{ij}))T_{ij}$ and $\varepsilon_q=(\delta_{kl}+\sqrt{2}(1-\delta_{kl}))T_{kl}$, where $p=i\delta_{ij}+(1-\delta_{ij})(9-i-j)$ and $q=k\delta_{kl}+(1-\delta_{kl})(9-k-l)$. So, we can define the generalized stress and the generalized strain as follows

$$\mathcal{K}^T=\left(\vec{\tau}^T,\vec{D}^T,\vec{B}^T\right)\quad\textit{and}\quad\mathcal{Z}^T=\left(\vec{\varepsilon}^T,-\vec{E}^T,-\vec{H}^T\right),\tag{7}$$

In this work we take into consideration a nonlinear behavior between $\mathcal{K}$ and $\mathcal{Z}$ described by the following three terms expansion

$$\mathcal{K}_i=\mathcal{L}_{ij}\mathcal{Z}_j+\mathcal{R}_{ijk}\mathcal{Z}_j\mathcal{Z}_k+\mathcal{S}_{ijkh}\mathcal{Z}_j\mathcal{Z}_k\mathcal{Z}_h,\tag{8}$$

where the generalized linear stiffness $\mathcal{L}_{ij}$ and the nonlinear susceptibilities $\mathcal{R}_{ijk}$ and $\mathcal{S}_{ijkh}$ have been introduced. It is possible to prove that $\mathcal{L}_{ij}$, $\mathcal{R}_{ijk}$ and $\mathcal{S}_{ijkh}$ are invariant to any permutation of the indexes. We remark that $\mathcal{L}$ contains the elastic, magnetic, dielectric, piezoelectric, piezomagnetic and magnetoelectric linear responses of the material. In addition, their nonlinear counterparts are described by tensors $\mathcal{R}$ and $\mathcal{S}$ (for example, they represent nonlinear electromagnetism, nonlinear elasticity, magnetostrictive or electrostrictive behaviors, Kerr effect and so on) (Newnham, 2005). In the following developments we use these notations for the tensor contractions

$$\begin{aligned}
&\mathcal{V}_j\mathcal{W}_j &&=\mathcal{V}\cdot\mathcal{W}\\
&\mathcal{A}_{ij}\mathcal{P}_{jst\ldots} &&=(\mathcal{A}:\mathcal{P})_{ist\ldots}\\
&\mathcal{B}_{ijk}\mathcal{P}_{jst\ldots}\mathcal{Q}_{krq\ldots} &&=(\mathcal{B}\,\dot{:}\,\mathcal{P}\otimes\mathcal{Q})_{ist\ldots rq\ldots}\\
&\mathcal{C}_{ijkl}\mathcal{P}_{jst\ldots}\mathcal{Q}_{krq\ldots}\mathcal{G}_{lnm\ldots} &&=(\mathcal{C}::\mathcal{P}\otimes\mathcal{Q}\otimes\mathcal{G})_{ist\ldots rq\ldots nm\ldots}.
\end{aligned}\tag{9}$$

For instance, this formalism can be used to rewrite the constitutive equation in the form

$$\mathcal{K}=\mathcal{L}:\mathcal{Z}+\mathcal{R}\,\dot{:}\,\mathcal{Z}\otimes\mathcal{Z}+\mathcal{S}::\mathcal{Z}\otimes\mathcal{Z}\otimes\mathcal{Z},\tag{10}$$

where of course, $\mathcal{L}$, $\mathcal{R}$ and $\mathcal{S}$ may represent any form of anisotropy, i.e. any kind of crystal symmetry (Nye, 1985; Sirotine and Chaskolskaia, 1984).

3. Nonlinear homogenization

We consider a laminated structure composed of N different layers, each described by tensors $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$ (defined in Eq. (10)) and having thickness d_i, $i=1,\ldots,N$ (see Fig. 1 for details). We can define the volume fraction of each component as $\phi_i=d_i/D$ where $D=\sum_{i=1}^N d_i$ is the total length of the structure. The system is loaded

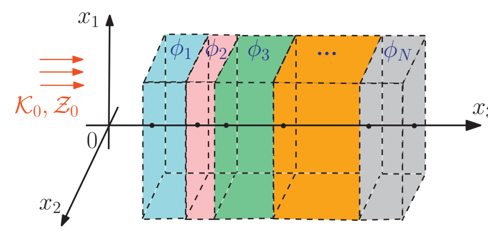

Fig. 1. Schematic representation of a multilayered structure composed of N different components. Each layer is described by tensors $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$ (i = 1, . . ., N). The whole structure is loaded by the generalized fields $\mathcal{K}_0$ and $\mathcal{Z}_0$.

by the remotely applied uniform fields $\mathcal{Z}_0$ and $\mathcal{K}_0$. We suppose that the interfaces are perpendicular to the axis x_3. In a following Section we will generalize this formalism to consider an arbitrary direction of lamination. We assume to have no free electric charge and no electric current distributed on the interfaces and we study the continuity of the physical fields across them. As well known, the continuous components of $\mathcal{K}$ across the interface are T_{13}, T_{23}, T_{33}, D_3 and B_3. Similarly, the continuous components of $\mathcal{Z}$ are ε_{11}, ε_{12}, ε_{22}, E_1, E_2, H_1 and H_2. We observe that these two sets of continuous components are complementary in the structure of vectors $\mathcal{Z}$ and $\mathcal{K}$. It means that the ith component of $\mathcal{Z}$ is continuous if and only if the ith component of $\mathcal{K}$ suffers a discontinuity. We can define $\mathbf{C}_{\mathcal{K}} = \{3, 4, 5, 9, 12\}$ as the subset containing the positions of continuous components of $\mathcal{K}$. We have $\text{card}(\mathbf{C}_{\mathcal{K}}) = n$ having defined $n = 5$. Similarly, we can define $\mathbf{C}_{\mathcal{Z}} = \{1, 2, 6, 7, 8, 10, 11\}$ as the subset containing the positions of continuous components of $\mathcal{Z}$. Now, we have $\text{card}(\mathbf{C}_{\mathcal{Z}}) = m$ having defined $m = 7$. We introduce the following operators

$$\begin{aligned}
\mathcal{P} &= \sum_{i=1}^{n}\sum_{j\in\mathbf{C}_{\mathcal{K}}}\mathcal{F}_{i,j}^{n,n+m},\\
\mathcal{Q} &= \sum_{i=1}^{m}\sum_{j\in\mathbf{C}_{\mathcal{Z}}}\mathcal{F}_{i,j}^{m,n+m},\\
\mathcal{N} &= \mathcal{P}^T:\mathcal{P} = \sum_{j\in\mathbf{C}_{\mathcal{K}}}\mathcal{F}_{j,j}^{n+m,n+m},\\
\mathcal{M} &= \mathcal{Q}^T:\mathcal{Q} = \sum_{j\in\mathbf{C}_{\mathcal{Z}}}\mathcal{F}_{j,j}^{n+m,n+m},
\end{aligned} \tag{11}$$

where $\mathcal{F}_{i,j}^{a,b}$ is an elementary matrix with a rows and b columns with the element (i, j) equal to one and all the others equal to zero. In other terms, we can affirm that $\mathcal{N}$ is a diagonal matrix with $\mathcal{N}_{ii} = 1$ for $i \in \mathbf{C}_{\mathcal{K}}$ and $\mathcal{N}_{ii} = 0$ if $i \notin \mathbf{C}_{\mathcal{K}}$. Similarly, $\mathcal{M}$ is a diagonal matrix with $\mathcal{M}_{ii} = 1$ for $i \in \mathbf{C}_{\mathcal{Z}}$ and $\mathcal{M}_{ii} = 0$ if $i \notin \mathbf{C}_{\mathcal{Z}}$. Moreover, the operators $\mathcal{N}$ and $\mathcal{M}$ are idempotent linear transformations ($\mathcal{N}^2 = \mathcal{N}$ and $\mathcal{M}^2 = \mathcal{M}$) and they decompose $\mathfrak{R}^{n+m}$ in the direct sum of two subspaces, i.e. $\mathcal{N} + \mathcal{M} = \mathcal{I}_{n+m}$ (the symbol $\mathcal{I}_r$ represents the identity matrix of order r). On the other hand, $\mathcal{P}$ can by simply obtained from $\mathcal{N}$ by removing the null rows; in the same way, $\mathcal{Q}$ can by simply obtained from $\mathcal{M}$ by removing the null rows. We remark that operators $\mathcal{P}$ and $\mathcal{Q}$ are useful to obtain the factorization of $\mathcal{N}$ and $\mathcal{M}$ given in Eq. (11). This factorization will be particularly useful in the next Section for introducing an arbitrary lamination direction.

The physical meaning of the introduced operators follows: the vector $\mathcal{N}:\mathcal{K} = \mathcal{P}^T:\mathcal{P}:\mathcal{K}$ has the same structure of $\mathcal{K}$ but all the discontinuous components are imposed to zero. In other words, we have $(\mathcal{N}:\mathcal{K})_j = \mathcal{K}_j$ if $j \in \mathbf{C}_{\mathcal{K}}$ and $(\mathcal{N}:\mathcal{K})_j = 0$ if $j \in \mathbf{C}_{\mathcal{Z}}$. On the other hand, $\mathcal{M}:\mathcal{Z} = \mathcal{Q}^T:\mathcal{Q}:\mathcal{Z}$ has the same structure of $\mathcal{Z}$ but all the discontinuous components are imposed to zero. It means that $(\mathcal{M}:\mathcal{Z})_j = \mathcal{Z}_j$ if $j \in \mathbf{C}_{\mathcal{Z}}$ and $(\mathcal{M}:\mathcal{K})_j = 0$ if $j \in \mathbf{C}_{\mathcal{K}}$.

Interestingly enough, this formalism can be used for any linear physical coupling described by two dual sets of variables (here $\mathcal{K}$ and $\mathcal{Z} \in \mathfrak{R}^{n+m}$) exhibiting the continuity of the complementary components (n components of $\mathcal{K}$ and m complementary components of $\mathcal{Z}$) across a given interface. We present here the analysis of the magneto-electro-elastic case, but it can be easily generalized to more complex situations, e.g. with thermal and/or other transport properties.

As anticipated, we generalize the so-called Backus–Tartar method in order to consider the fully coupled and nonlinear magneto-electro-elastic case. This approach is based on the following idea: instead of using Eq. (10), we try to rewrite the constitutive law of each layer in a particular form where the continuous components of the physical fields appear on the right-hand side of the equation. Therefore, as we will describe below, the effective magneto-electro-elastic tensors can be simply found by just averaging both sides of this constitutive equation. We define in the ith layer the coupled fields $\mathcal{K}_i$ and $\mathcal{Z}_i$. So, the set of continuous components can be inserted in a new state vector $\mathcal{X}_i$ and, similarly, the discontinuous components can be collected in a dual state vector $\mathcal{Y}_i$

$$\mathcal{X}_i = \mathcal{N}:\mathcal{K}_i + \mathcal{M}:\mathcal{Z}_i, \tag{12}$$

$$\mathcal{Y}_i = \mathcal{M}:\mathcal{K}_i - \mathcal{N}:\mathcal{Z}_i. \tag{13}$$

For the following purposes we invert Eqs. (12) and (13), eventually obtaining

$$\mathcal{K}_i = \mathcal{N}:\mathcal{X}_i + \mathcal{M}:\mathcal{Y}_i, \tag{14}$$

$$\mathcal{Z}_i = \mathcal{M}:\mathcal{X}_i - \mathcal{N}:\mathcal{Y}_i. \tag{15}$$

To prove these results we can simply substitute Eqs. (14) and (15) in Eqs. (12) and (13) and apply the idempotency properties $\mathcal{N}^2 = \mathcal{N}$ and $\mathcal{M}^2 = \mathcal{M}$. Starting form the general constitutive equation given in Eq. (10), we have in each layer (i = 1, . . ., N)

$$\mathcal{K}_i = \mathcal{L}_i:\mathcal{Z}_i + \mathcal{R}_i \therefore \mathcal{Z}_i\otimes\mathcal{Z}_i + \mathcal{S}_i :: \mathcal{Z}_i\otimes\mathcal{Z}_i\otimes\mathcal{Z}_i, \tag{16}$$

and we try to determine the quantities $\mathcal{A}_i$, $\mathcal{B}_i$ and $\mathcal{C}_i$ such that the constitutive equation assumes the form (i = 1, . . ., N)

$$\mathcal{Y}_i = \mathcal{A}_i:\mathcal{X}_i + \mathcal{B}_i \because \mathcal{X}_i\otimes\mathcal{X}_i + \mathcal{C}_i :: \mathcal{X}_i\otimes\mathcal{X}_i\otimes\mathcal{X}_i, \tag{17}$$

where $\mathcal{X}_i$ and $\mathcal{Y}_i$ are defined in Eqs. (12) and (13). To explicitly obtain the structure of $\mathcal{A}_i$, $\mathcal{B}_i$ and $\mathcal{C}_i$ we must compare Eqs. (16) and Eq. (17). Essentially, by substituting Eqs. (14) and (15) in Eq. (16), by considering Eq. (17) and by separating the terms of the same order in $\mathcal{X}_i$, we easily obtain the results

$$\mathcal{A}_i = (\mathcal{M} + \mathcal{L}_i:\mathcal{N})^{-1}:(\mathcal{L}_i:\mathcal{M} - \mathcal{N}), \tag{18}$$

$$\begin{aligned}
\mathcal{B}_i &= (\mathcal{M} + \mathcal{L}_i:\mathcal{N})^{-1}:\mathcal{R}_i \therefore \mathcal{D}_i\otimes\mathcal{D}_i,\\
\mathcal{C}_i &= (\mathcal{M} + \mathcal{L}_i:\mathcal{N})^{-1}:\mathcal{S}_i :: \mathcal{D}_i\otimes\mathcal{D}_i\otimes\mathcal{D}_i\\
&\quad -(\mathcal{M} + \mathcal{L}_i:\mathcal{N})^{-1}:\mathcal{R}_i \therefore \mathcal{D}_i\otimes(\mathcal{N}:\mathcal{B}_i)\\
&\quad -(\mathcal{M} + \mathcal{L}_i:\mathcal{N})^{-1}:\mathcal{R}_i \because (\mathcal{N}:\mathcal{B}_i)\otimes\mathcal{D}_i,
\end{aligned}$$

where to compact the notation, we defined

$$\mathcal{D}_i = \mathcal{M} - \mathcal{N}:\mathcal{A}_i. \tag{19}$$

It is important to observe that $\mathcal{A}_i$, $\mathcal{B}_i$ and $\mathcal{C}_i$ exhibit the same symmetries of $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$: all quantities are invariant to any permutation of their indexes. We remark that these symmetries are related to the minus signs introduced in Eq. (7) for $\mathcal{Z}$ and in Eq. (13) for $\mathcal{Y}_i$. Anyway, each material is now described by Eq. (17). We embed our heterogeneous system in an environment with uniform fields $\mathcal{Z}_0$ and $\mathcal{K}_0$. So, we can determine $\mathcal{X}_0$ and $\mathcal{Y}_0$ with transformations given in Eqs. (12) and (13) (with i = 0). The vector $\mathcal{X}_0$ contains all the continuous physical fields and, therefore, it remains unchanged in each layer (it means that $\mathcal{X}_0=\mathcal{X}_i=\langle\mathcal{X}\rangle\;\forall\, i=1,\ldots,N$). The average value of $\mathcal{Y}$ in the system is given by

$$\langle\mathcal{Y}\rangle=\sum_{i=1}^{N}\phi_i\mathcal{Y}_i=\sum_{i=1}^{N}\phi_i\mathcal{A}_i:\langle\mathcal{X}\rangle+\sum_{i=1}^{N}\phi_i\mathcal{B}_i\,\dot{:}\,\langle\mathcal{X}\rangle\otimes\langle\mathcal{X}\rangle$$
$$+\sum_{i=1}^{N}\phi_i\mathcal{C}_i::\langle\mathcal{X}\rangle\otimes\langle\mathcal{X}\rangle\otimes\langle\mathcal{X}\rangle. \tag{20}$$

Hence, we can define the effective tensors $\mathcal{A}_{\mathit{eff}}$, $\mathcal{B}_{\mathit{eff}}$ and $\mathcal{C}_{\mathit{eff}}$ as follows

$$\mathcal{A}_{\mathit{eff}}=\sum_{i=1}^{N}\phi_i\mathcal{A}_i,\quad \mathcal{B}_{\mathit{eff}}=\sum_{i=1}^{N}\phi_i\mathcal{B}_i,\quad \mathcal{C}_{\mathit{eff}}=\sum_{i=1}^{N}\phi_i\mathcal{C}_i. \tag{21}$$

It means that the quantities $\mathcal{A}_i$, $\mathcal{B}_i$ and $\mathcal{C}_i$ are natural parameters for the nonlinear laminated structure since they can be directly averaged (in the sense of the weighted arithmetic mean) in order to obtain the homogenized response of the overall system. To conclude, we can invert the transformation in Eq. (18) by obtaining the final effective properties in the form

$$\mathcal{L}_{\mathit{eff}}=(\mathcal{M}:\mathcal{A}_{\mathit{eff}}+\mathcal{N}):\mathcal{D}_{\mathit{eff}}^{-1}, \tag{22}$$

$$\begin{aligned}
\mathcal{R}_{\mathit{eff}} &= (\mathcal{M}+\mathcal{L}_{\mathit{eff}}:\mathcal{N}):\mathcal{B}_{\mathit{eff}}\,\dot{:}\,\mathcal{D}_{\mathit{eff}}^{-1}\otimes\mathcal{D}_{\mathit{eff}}^{-1},\\
\mathcal{S}_{\mathit{eff}} &= (\mathcal{M}+\mathcal{L}_{\mathit{eff}}:\mathcal{N}):\mathcal{C}_{\mathit{eff}}::\mathcal{D}_{\mathit{eff}}^{-1}\otimes\mathcal{D}_{\mathit{eff}}^{-1}\otimes\mathcal{D}_{\mathit{eff}}^{-1}\\
&\quad+\mathcal{R}_{\mathit{eff}}\,\dot{:}\,\mathcal{I}_{n+m}\otimes(\mathcal{N}:\mathcal{B}_{\mathit{eff}}\,\dot{:}\,\mathcal{D}_{\mathit{eff}}^{-1}\otimes\mathcal{D}_{\mathit{eff}}^{-1})\\
&\quad+\mathcal{R}_{\mathit{eff}}\,\dot{:}\,(\mathcal{N}:\mathcal{B}_{\mathit{eff}}\quad\mathcal{D}_{\mathit{eff}}^{-1}\otimes\mathcal{D}_{\mathit{eff}}^{-1})\otimes\mathcal{I}_{n+m},
\end{aligned}$$

where

$$\mathcal{D}_{\mathit{eff}}=\mathcal{M}-\mathcal{N}:\mathcal{A}_{\mathit{eff}}. \tag{23}$$

Eqs. (18), (21) and (22) represent the main result of this work. They allow for the exact homogenization of the linear and nonlinear tensor properties of the laminated structure. We underline that these final expressions can be easily implemented in a software code by means of the standard procedures of tensor calculus. For instance, for the linear homogenization we obtain the explicit and compact expression

$$\begin{aligned}
\mathcal{L}_{\mathit{eff}} &= \Big[\mathcal{M}:\sum_{i=1}^{N}\phi_i(\mathcal{M}+\mathcal{L}_i:\mathcal{N})^{-1}:(\mathcal{L}_i:\mathcal{M}-\mathcal{N})+\mathcal{N}\Big]\\
&\quad:\Big[\mathcal{M}-\mathcal{N}:\sum_{i=1}^{N}\phi_i(\mathcal{M}+\mathcal{L}_i:\mathcal{N})^{-1}:(\mathcal{L}_i:\mathcal{M}-\mathcal{N})\Big]^{-1},
\end{aligned} \tag{24}$$

which we proved to be exactly equivalent to a series of results published in recent literature (Kim, 2011; Challagulla and Georgiades, 2011; Bravo-Castillero et al., 2008). Some examples of application of Eq. (24) will be discussed in Section 5. We remark that our expressions for $\mathcal{L}_{\mathit{eff}}$, $\mathcal{R}_{\mathit{eff}}$ and $\mathcal{S}_{\mathit{eff}}$ are exact when we are interested in the nonlinear behavior up the the third order. If needed, they can be simply generalized by repeating the entire procedure with a desired number of nonlinear terms.

4. Generalization to an arbitrary lamination direction

In this Section we prove that the previous formalism can be simply generalized in order to consider an arbitrary direction $\vec{n}$ of lamination. In particular, we will prove that previous results can be applied with a specific form of operators $\mathcal{P}_{\vec{n}}$, $\mathcal{Q}_{\vec{n}}$, $\mathcal{N}_{\vec{n}}$ and $\mathcal{M}_{\vec{n}}$, now depending on $\vec{n}$. We suppose to deal with N different materials described by tensors $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$ in a given coordinate system $\mathbf{e}$ (x_1, x_2, x_3). We consider another system $\mathbf{f}$ (x_1', x_2', x_3'), rotated with respect to $\mathbf{e}$ in order to get the axis x_3' oriented along the unit vector $\vec{n}=(\cos\varphi\sin\vartheta, \sin\varphi\sin\vartheta, \cos\vartheta)$ on the base $\mathbf{e}$. Here ϑ and φ are the standard nutation and precession angles, respectively. It means that we must rotate the system $\mathbf{e}$ of an angle ϑ along the unit vector $\vec{\nu}=(-\sin\varphi, \cos\varphi, 0)$ (with the right-hand grip rule). To do this we can use a rotation matrix given by the following expression

$$\hat{\mathbb{R}}(\vartheta,\varphi)=\begin{pmatrix} s_\varphi^2+c_\vartheta c_\varphi^2 & s_\varphi c_\varphi(c_\vartheta-1) & c_\varphi s_\vartheta\\ s_\varphi c_\varphi(c_\vartheta-1) & c_\varphi^2+c_\vartheta s_\varphi^2 & s_\varphi s_\vartheta\\ -c_\varphi s_\vartheta & -s_\varphi s_\vartheta & c_\vartheta \end{pmatrix}, \tag{25}$$

where $c_\vartheta=\cos\vartheta$, $c_\varphi=\cos\varphi$, $s_\vartheta=\sin\vartheta$ and $s_\varphi=\sin\varphi$. For an arbitrary vector $\vec{w}$ we have $\vec{w}^{\mathbf{e}}=\hat{\mathbb{R}}(\vartheta,\varphi):\vec{w}^{\mathbf{f}}$ where $\vec{w}^{\mathbf{e}}$ are the coordinates of $\vec{w}$ on the base $\mathbf{e}$ and $\vec{w}^{\mathbf{f}}$ are those on the base $\mathbf{f}$. This is the law of transformation of all the electric and magnetic vector fields.

Concerning the strain and stress tensors we have to introduce a more complicated procedure. The following relations hold on between different frames: $\hat{\varepsilon}^{\mathbf{e}}=\hat{\mathbb{R}}(\vartheta,\varphi):\hat{\varepsilon}^{\mathbf{f}}:\hat{\mathbb{R}}(\vartheta,\varphi)^T$ for the strain and, similarly, $\hat{T}^{\mathbf{e}}=\hat{\mathbb{R}}(\vartheta,\varphi):\hat{T}^{\mathbf{f}}:\hat{\mathbb{R}}(\vartheta,\varphi)^T$ for the stress. They can be converted to the generalized Voigt notation defining a matrix ω, which acts as a rotation matrix on vectors $\vec{\tau}$ or $\vec{\varepsilon}$ defined in Eqs. (5) and (6). In other words, we can write $\vec{\tau}^{\mathbf{e}}=\omega:\vec{\tau}^{\mathbf{f}}$ and $\vec{\varepsilon}^{\mathbf{e}}=\omega:\vec{\varepsilon}^{\mathbf{f}}$. The entries of the matrix ω can be easily identified by the comparison between the relations $\hat{T}^{\mathbf{e}}=\hat{\mathbb{R}}(\vartheta,\varphi):\hat{T}^{\mathbf{f}}:\hat{\mathbb{R}}(\vartheta,\varphi)^T$ and $\vec{\tau}^{\mathbf{e}}=\omega:\vec{\tau}^{\mathbf{f}}$. Summing up, we can write the laws of transformation of the generalized stress

$$\mathcal{K}^{\mathbf{e}}=\begin{pmatrix}\vec{\tau}^{\mathbf{e}}\\ \vec{D}^{\mathbf{e}}\\ \vec{B}^{\mathbf{e}}\end{pmatrix}=\begin{pmatrix}\omega & 0 & 0\\ 0 & \hat{\mathbb{R}} & 0\\ 0 & 0 & \hat{\mathbb{R}}\end{pmatrix}:\begin{pmatrix}\vec{\tau}^{\mathbf{f}}\\ \vec{D}^{\mathbf{f}}\\ \vec{B}^{\mathbf{f}}\end{pmatrix}\quad \Omega:\mathcal{K}^{\mathbf{f}}, \tag{26}$$

and, similarly, for the strain $\mathcal{Z}^{\mathbf{e}}=\Omega:\mathcal{Z}^{\mathbf{f}}$, where we have introduced the generalized rotation matrix Ω. We underline that the use of the modified Voigt notation (see Eqs. (5) and (6)) has profitably conducted to the same transformation law for $\mathcal{K}$ and $\mathcal{Z}$ (operated by the same rotation matrix Ω). As final result we obtain the transformation rules

$$\mathcal{L}_i^{\mathbf{f}}=\Omega^{-1}:\mathcal{L}_i^{\mathbf{e}}:\Omega \tag{27}$$

$$\mathcal{R}_i^{\mathbf{f}}=\Omega^{-1}:\mathcal{R}_i^{\mathbf{e}}\,\dot{:}\,\Omega\otimes\Omega \tag{28}$$

$$\mathcal{S}_i^{\mathbf{f}}=\Omega^{-1}:\mathcal{S}_i^{\mathbf{e}}::\Omega\otimes\Omega\otimes\Omega \tag{29}$$

for the tensor properties $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$ describing the (linear and nonlinear) coupled response of each material.

Now, we consider the laminated structure represented in Fig. 2. On the base $\mathbf{f}$ the direction of lamination is x_3' and, therefore, we can use Eqs. (18), (21) and (22) for quantities $\mathcal{L}_i^{\mathbf{f}}$, $\mathcal{R}_i^{\mathbf{f}}$ and $\mathcal{S}_i^{\mathbf{f}}$. Then, we can apply Eqs. (27), (28) and (29) in order to obtain, after a long but straightforward calculation, the effective properties on the original base $\mathbf{e}$. The remarkable result is that the structure of Eqs. (18), (21) and (22) remains unchanged (for an arbitrary lamination direction) provided that we use new operators $\mathcal{P}_{\vec{n}}$, $\mathcal{Q}_{\vec{n}}$, $\mathcal{N}_{\vec{n}}$ and $\mathcal{M}_{\vec{n}}$ in place of old operators $\mathcal{P}$, $\mathcal{Q}$, $\mathcal{N}$ and $\mathcal{M}$ defined in Eq. (11). The new definitions follow

$$\mathcal{P}_{\vec{n}}=\mathcal{P}:\Omega^{-1},\quad \mathcal{P}_{\vec{n}}^T=\Omega:\mathcal{P}^T, \tag{30}$$

$$\mathcal{Q}_{\vec{n}}=\mathcal{Q}:\Omega^{-1},\quad ,\mathcal{Q}_{\vec{n}}^T=\Omega:\mathcal{Q}^T, \tag{31}$$

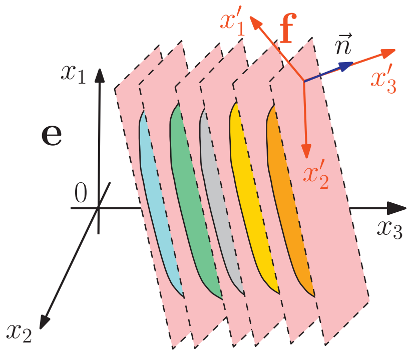

Fig. 2. Multilayered structure with an arbitrary lamination direction $\vec{n}$ (represented on the base **e**). All tensor properties $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$ (and the effective ones $\mathcal{L}_{eff}$, $\mathcal{R}_{eff}$ and $\mathcal{S}_{eff}$) are measured on the base **e**. The base **f**, with x_3' aligned with $\vec{n}$, is represented as well. The rotation matrix $\hat{\mathbb{R}}(\vartheta, \varphi)$ linking base **f** to base **e** is given in Eq. (25).

$$\begin{aligned}\mathcal{N}_{\vec{n}} &= \Omega : \mathcal{N} : \Omega^{-1} \\ &= \Omega : \mathcal{P}^T : \mathcal{P} : \Omega^{-1} = \mathcal{P}_{\vec{n}}^T : \mathcal{P}_{\vec{n}},\end{aligned} \tag{32}$$

$$\begin{aligned}\mathcal{M}_{\vec{n}} &= \Omega : \mathcal{M} : \Omega^{-1} \\ &= \Omega : \mathcal{Q}^T : \mathcal{Q} : \Omega^{-1} = \mathcal{Q}_{\vec{n}}^T : \mathcal{Q}_{\vec{n}},\end{aligned} \tag{33}$$

and they fulfil the properties $\mathcal{N}_{\vec{n}}^2 = \mathcal{N}_{\vec{n}}$, $\mathcal{M}_{\vec{n}}^2 = \mathcal{M}_{\vec{n}}$ and $\mathcal{N}_{\vec{n}} + \mathcal{M}_{\vec{n}} = \mathcal{I}_{n+m}$, as can be easily verified. It means that, with an arbitrary lamination direction, operators $\mathcal{N}_{\vec{n}}$ and $\mathcal{M}_{\vec{n}}$ are non-trivial idempotent linear transformations and they decompose $\Re^{n+m}$ in the direct sum of two non-canonical subspaces. It is important to remember that, when we apply Eqs. (18), (21) and (22) with the new operators $\mathcal{N}_{\vec{n}}$ and $\mathcal{M}_{\vec{n}}$, the tensor properties $\mathcal{L}_i$, $\mathcal{R}_i$ and $\mathcal{S}_i$ (and the effective ones $\mathcal{L}_{eff}$, $\mathcal{R}_{eff}$ and $\mathcal{S}_{eff}$) are all measured on the base **e**. The factorization of $\mathcal{N}_{\vec{n}}$ with $\mathcal{P}_{\vec{n}}$, in this case with an arbitrary lamination direction, is very convenient because the mathematical structure of $\mathcal{P}_{\vec{n}}$ is much simpler than that of $\mathcal{N}_{\vec{n}}$ (see Eq. (34) below). It means that the knowledge of the operator $\mathcal{P}_{\vec{n}}$ is sufficient to apply the whole homogenization procedure with an arbitrary lamination direction since we can determine $\mathcal{N}_{\vec{n}} = \mathcal{P}_{\vec{n}}^T : \mathcal{P}_{\vec{n}}$ and $\mathcal{M}_{\vec{n}} = \mathcal{I}_{n+m} - \mathcal{P}_{\vec{n}}^T : \mathcal{P}_{\vec{n}}$. Therefore, the only information concerning the lamination direction $\vec{n}$ is included within the tensor $\mathcal{P}_{\vec{n}}$. We eventually obtained its following explicit form

$$\mathcal{P}_{\vec{n}}^T = \begin{pmatrix} c_\varphi^2 s_\vartheta^2 & -\sqrt{2} s_\varphi c_\varphi^2 s_\vartheta (1-c_\vartheta) & \sqrt{2} c_\varphi s_\vartheta \left(s_\varphi^2 + c_\varphi^2 c_\vartheta\right) & 0 & 0 \\ s_\varphi^2 s_\vartheta^2 & \sqrt{2} s_\varphi s_\vartheta \left(c_\varphi^2 + s_\varphi^2 c_\vartheta\right) & -\sqrt{2} s_\varphi^2 c_\varphi s_\vartheta (1-c_\vartheta) & 0 & 0 \\ c_\vartheta^2 & -\sqrt{2} s_\varphi s_\vartheta c_\vartheta & -\sqrt{2} c_\varphi s_\vartheta c_\vartheta & 0 & 0 \\ \sqrt{2} s_\varphi s_\vartheta c_\vartheta & s_\varphi^2 \left(c_\vartheta^2 - s_\vartheta^2\right) + c_\varphi^2 c_\vartheta & s_\varphi c_\varphi \left(2c_\vartheta^2 - c_\vartheta - 1\right) & 0 & 0 \\ \sqrt{2} c_\varphi s_\vartheta c_\vartheta & s_\varphi c_\varphi \left(2c_\vartheta^2 - c_\vartheta - 1\right) & c_\varphi^2 \left(c_\vartheta^2 - s_\vartheta^2\right) + s_\varphi^2 c_\vartheta & 0 & 0 \\ \sqrt{2} s_\varphi c_\varphi s_\vartheta^2 & c_\varphi s_\vartheta \left(c_\varphi^2 - s_\varphi^2 + 2c_\vartheta s_\varphi^2\right) & s_\varphi s_\vartheta \left(s_\varphi^2 - c_\varphi^2 + 2c_\vartheta c_\varphi^2\right) & 0 & 0 \\ 0 & 0 & 0 & c_\varphi s_\vartheta & 0 \\ 0 & 0 & 0 & s_\varphi s_\vartheta & 0 \\ 0 & 0 & 0 & c_\vartheta & 0 \\ 0 & 0 & 0 & 0 & c_\varphi s_\vartheta \\ 0 & 0 & 0 & 0 & s_\varphi s_\vartheta \\ 0 & 0 & 0 & 0 & c_\vartheta \end{pmatrix}, \tag{34}$$

where as before, $c_\vartheta = \cos\vartheta$, $c_\varphi = \cos\varphi$, $s_\vartheta = \sin\vartheta$ and $s_\varphi = \sin\varphi$.

The main result of the present paper can be finally stated as follows: *a nonlinear laminated material composed of layers with tensor properties $\mathcal{L}_i, \mathcal{R}_i, \mathcal{S}_i \ldots (i = 1, \ldots, N)$ is described by effective parameters $\mathcal{L}_{eff}$, $\mathcal{R}_{eff}$, $\mathcal{S}_{eff}$..., which can be explicitly found through the following scheme*

$$\begin{array}{ccc} \left\{\mathcal{L}_i, \mathcal{R}_i, \mathcal{S}_i \ldots\right\}_{i=1,\ldots,N} & \overset{\psi_{\vec{n}}}{\Rightarrow} & \left\{\mathcal{A}_i, \mathcal{B}_i, \mathcal{C}_i \ldots\right\}_{i=1,\ldots,N} \\ \Downarrow \ \text{NH} & & \Downarrow \ \text{WAM} \\ \left\{\mathcal{L}_{eff}, \mathcal{R}_{eff}, \mathcal{S}_{eff} \ldots\right\} & \overset{\psi_{\vec{n}}^{-1}}{\Longleftarrow} & \left\{\mathcal{A}_{eff}, \mathcal{B}_{eff}, \mathcal{C}_{eff} \ldots\right\} \end{array} \tag{35}$$

where $\psi_{\vec{n}}$ is the nonlinear mapping given in Eq. (18) (used with the general form of $\mathcal{N}_{\vec{n}}$ and $\mathcal{M}_{\vec{n}}$, valid for an arbitrary lamination direction) and $\psi_{\vec{n}}^{-1}$ is its inverse given in Eq. (22). It means that the nonlinear homogenization (NH) can be performed through three steps: the application of $\psi_{\vec{n}}$, the determination of the weighted arithmetic mean (WAM) defined in Eq. (21) and, finally, the application of $\psi_{\vec{n}}^{-1}$. We remark that the mathematical form of $\psi_{\vec{n}}$ can be simply generalized for considering an arbitrary number of nonlinear terms.

The technique presented in this paper allows for the direct determination of the effective properties without calculating the distribution of the physical fields within the layers of the structure. If it is important to evaluate the elastic and electromagnetic fields in each layer, then it is necessary to generalize standard theories originally developed in the pure mechanical context (Pagano, 1978; Mittelstedt and Becker, 2007).

5. Particular cases and applications

In this Section we show the application of the general procedure to some specific cases. The following structures will be considered:

1 linear analysis of artificial multiferroics: we apply the general solution given in Eq. (24) in order to numerically obtain the magnetoelectric response of piezoelectric/magnetoelastic laminates. We will stress the good agreement with some results published in recent literature;
2 linear and nonlinear analysis of pure electric or magnetic isotropic systems: we approach the problem analytically and we

obtain the classical results for the linear response (Tartar, 1979; Milton, 2004) and new explicit expressions for the nonlinear one;
3 linear and nonlinear analysis of pure elastic isotropic systems: again, we approach the problem analytically and we obtain the classical results for the linear response (Postma, 1955; Backus, 1962; Milton, 2004) and new explicit expressions for the nonlinear one;
4 linear and nonlinear properties of coupled magnetoelectric isotropic systems: as in the previous cases we perform a theoretical analysis providing the complete effective constitutive equation of the laminated system.

5.1. Linear analysis of artificial multiferroics

A first example of application of the linear theory (see Eq. (24)) is discussed here. We consider some configurations involving piezoelectric (PE) and magnetoelastic (ME) phases. In particular, we utilize the PE ceramic $BaTiO_3$ (barium titanate) and the ME compound $CoFe_2O_4$ (cobalt ferrite), whose physical linear properties are reported by Kim (2011). Both materials exhibit a uniaxial behavior and therefore their crystal orientation can be defined through a given poling direction. For simplicity, we consider here the case with both the poling directions aligned to the lamination direction $\vec{n}=(0,0,1)$. We suppose to have a pure elastic isotropic layer (EL) between the PE and the ME ones. It has a bulk modulus $k = 150\times10^9$Pa and a shear modulus $\mu = 40\times10^9$Pa. The volume fractions are defined as follows: $\phi_{PE} = cx$, $\phi_{ME} = x(1-c)$ and $\phi_{EL} = 1-x$, where the stoichiometric coefficients c and x vary in the range (0,1).

The final overall properties of the laminated system are written as

$$\begin{pmatrix}\langle\vec{\tau}\rangle\\ \langle\vec{D}\rangle\\ \langle\vec{B}\rangle\end{pmatrix}=\underbrace{\begin{pmatrix}\mathbf{C} & \mathbf{E} & \mathbf{F}\\ \mathbf{E}^T & -\mathbf{P} & -\mathbf{G}\\ \mathbf{F}^T & -\mathbf{G}^T & -\mathbf{M}\end{pmatrix}}_{\mathcal{L}_{eff}}\begin{pmatrix}\langle\vec{\varepsilon}\rangle\\ -\langle\vec{E}\rangle\\ -\langle\vec{H}\rangle\end{pmatrix}\qquad(36)$$

where $\mathcal{L}_{eff}$ can be found through Eq. (24). We present here the results concerning the magneto-electric properties. It is indeed interesting to observe the emergence of the magneto-electric response generated by the combination of piezoelectric and magnetoelastic properties of two materials that have no intrinsic magneto-electric coupling. The result is given by tensor $\mathbf{G}=\{g_{ij}\}$, in general non symmetric, describing the relation $\vec{D}=\mathbf{G}\vec{H}$ (with $\hat{\varepsilon}=0$ and $\vec{E}=0$) or, equivalently, $\vec{B}=\mathbf{G}^T\vec{E}$ (with $\hat{\varepsilon}=0$ and $\vec{H}=0$). In Fig. 3 the components g_{ij} are represented in terms of x and c. In Fig. 3a g_{ij} is shown versus x for three different values (1/2,1/5,1/8) of c. We can observe the absence of magneto-electric effect for $x=0$ (pure elastic system EL) and the strongest magneto-electric effect for $x=1$ (absence of elastic interphase between ME and PE subsystems). In Fig. 3b we fixed $x=1$ and we studied g_{ij} versus c. Of course, we always have $g_{ij}=0$ if $c=0$ (pure ME response) or $c=1$ (pure PE response). In all cases the effective behavior corresponds to a transversely isotropic symmetry, similarly to the PE and ME phases composing the system. Moreover, the overall poling direction is clearly aligned to the poling axes of the PE and ME materials. From a quantitative point of view, we observe that the largest magneto-electric response $g_{11}=g_{22}\cong-3.5\times10^{-8}$s/m is obtained for $x=1$ and $c=1/2$ (however, g_{33} is quite negligible). We underline that the results corresponding to Fig. 3b are in perfect agreement with those reported in Fig.7 by Kim (2011). Unfortunately, the nonlinear properties of barium titanate and cobalt ferrite are not available in literature. Nevertheless, we show the explicit implementation of our nonlinear homogenization in the following examples.

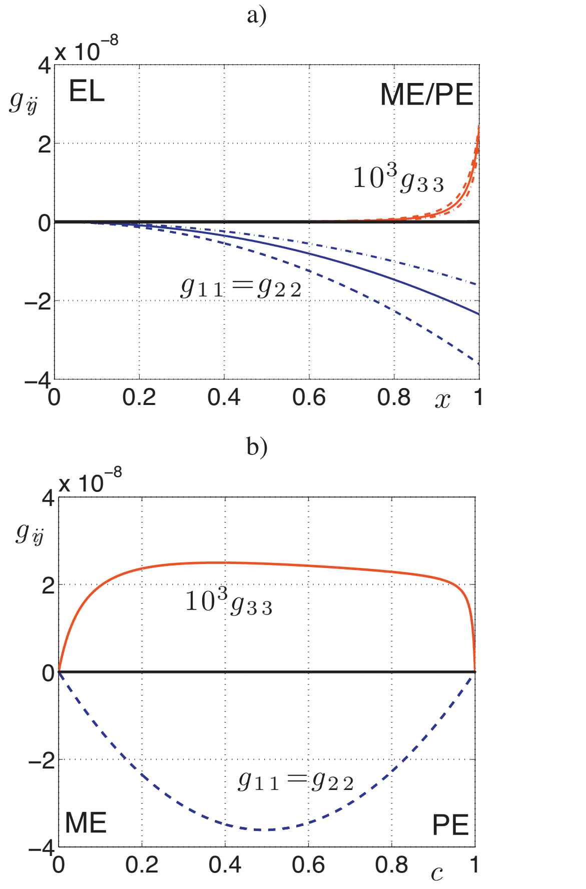

Fig. 3. Magneto-electric response of the combination PE/EL/ME: (a) g_{ij} is shown versus x for three different values of c (c=1/2 dashed lines, c=1/5 continuous lines and c=1/8 dashed-dotted lines) and (b) we fixed x = 1 and we studied g_{ij} versus c. We represented only the magnetoelectric components different from zero.

5.2. Pure electric or magnetic isotropic system: linear and nonlinear analysis

We introduce the simplest laminated nonlinear system, which can be explicitly homogenized by means of the proposed procedure. It is composed of pure electric or magnetic layers with an isotropic response. To fix the ideas we consider the dielectric case and we determine the most general isotropic nonlinear constitutive equation. The free energy w defined in Eq. (3) must be a scalar isotropic function of one vector $\vec{E}$. It means that $w(\vec{E})=w(\mathbb{R}^T:\vec{E})$ for any rotation tensor $\hat{\mathbb{R}}$. Therefore, w can only depend on the single invariant that can be defined through the vector $\vec{E}$, i.e. the scalar product $\vec{E}\cdot\vec{E}$. As result, we have $w(\vec{E})=w(\vec{E}\cdot\vec{E})$. By considering the first two terms of the series expansion of w we obtain the following mathematical form

$$w(\vec{E})=-\frac{1}{2}\varepsilon\vec{E}\cdot\vec{E}-\frac{1}{4}\alpha\left(\vec{E}\cdot\vec{E}\right)^2,\qquad(37)$$

where ε is the permittivity and α the dielectric susceptibility. By differentiating Eq. (37) we obtain the displacement-field relation

$$D_i=-\frac{\partial w}{\partial E_i}=\varepsilon E_i+\alpha E_jE_jE_i,\qquad(38)$$

or, equivalently in vector notation $\vec{D}=\varepsilon\vec{E}+\alpha\left(\vec{E}\cdot\vec{E}\right)\vec{E}$. This is the so-called Kerr behavior of a dielectric material. We apply our general theory to homogenize a laminated system with $\vec{n}=(0,0,1)$, where each layer is a Kerr material having parameters ε_i and α_i

($i = 1 \ldots N$). Since we are dealing with a pure elastic system, we can adopt the following simplified quantities

$$\mathcal{D} = \begin{pmatrix} D_1 \\ D_2 \\ D_3 \end{pmatrix} = \begin{pmatrix} y_1 \\ y_2 \\ x_3 \end{pmatrix}, \quad \mathcal{E} = \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix} = \begin{pmatrix} x_1 \\ x_2 \\ -y_3 \end{pmatrix}, \tag{39}$$

corresponding to Eqs. (14) and (15) and the following ones

$$\mathcal{X} = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix} = \begin{pmatrix} E_1 \\ E_2 \\ D_3 \end{pmatrix}, \quad \mathcal{Y} = \begin{pmatrix} y_1 \\ y_2 \\ y_3 \end{pmatrix} = \begin{pmatrix} D_1 \\ D_2 \\ -E_3 \end{pmatrix}, \tag{40}$$

corresponding to Eqs. (12) and (13). By using Eq. (18) (i.e. by applying $\psi_{\vec{n}}$), we can eventually found the response of each layer in the form defined in Eq. (17), i.e. in the form $\mathcal{Y} = \mathcal{Y}(\mathcal{X})$

$$\mathcal{Y} = \varepsilon \begin{pmatrix} x_1 \\ x_2 \\ -x_3/\varepsilon^2 \end{pmatrix} + \alpha \left(x_1^2 + x_2^2 + \frac{x_3^2}{\varepsilon^2} \right) \begin{pmatrix} x_1 \\ x_2 \\ x_3/\varepsilon^2 \end{pmatrix}. \tag{41}$$

Hence, we perform the average (WAM) defined in Eq. (20) to obtain the constitutive equation in the form $\langle\mathcal{Y}\rangle = \mathcal{Y}(\mathcal{X})$

$$\langle\mathcal{Y}\rangle = \begin{pmatrix} \langle\varepsilon\rangle x_1 \\ \langle\varepsilon\rangle x_2 \\ -\langle 1/\varepsilon\rangle x_3 \end{pmatrix} + \left(x_1^2 + x_2^2\right) \begin{pmatrix} \langle\alpha\rangle x_1 \\ \langle\alpha\rangle x_2 \\ -\langle\alpha/\varepsilon^2\rangle x_3 \end{pmatrix} \tag{42}$$

$$+ x_3^2 \begin{pmatrix} \langle\alpha/\varepsilon^2\rangle x_1 \\ \langle\alpha/\varepsilon^2\rangle x_2 \\ -\langle\alpha/\varepsilon^4\rangle x_3 \end{pmatrix}.$$

To conclude, we apply Eq. (22) (or, equivalently, $\psi_{\vec{n}}^{-1}$) to obtain the homogenized relation in the final form

$$\langle D_1\rangle = \langle\varepsilon\rangle\langle E_1\rangle + \langle\alpha\rangle\langle E_1\rangle^3 + \langle\alpha\rangle\langle E_1\rangle\langle E_2\rangle^2 + \frac{\langle\frac{\alpha}{\varepsilon^2}\rangle}{\langle\frac{1}{\varepsilon}\rangle^2}\langle E_1\rangle\langle E_3\rangle^2, \tag{43}$$

$$\langle D_2\rangle = \langle\varepsilon\rangle\langle E_2\rangle + \langle\alpha\rangle\langle E_2\rangle\langle E_1\rangle^2 + \langle\alpha\rangle\langle E_2\rangle^3 + \frac{\langle\frac{\alpha}{\varepsilon^2}\rangle}{\langle\frac{1}{\varepsilon}\rangle^2}\langle E_2\rangle\langle E_3\rangle^2, \tag{44}$$

$$\langle D_3\rangle = \frac{1}{\langle\frac{1}{\varepsilon}\rangle}\langle E_3\rangle + \frac{\langle\frac{\alpha}{\varepsilon^2}\rangle}{\langle\frac{1}{\varepsilon}\rangle^2}\langle E_3\rangle\langle E_1\rangle^2 + \frac{\langle\frac{\alpha}{\varepsilon^2}\rangle}{\langle\frac{1}{\varepsilon}\rangle^2}\langle E_3\rangle\langle E_2\rangle^2 + \frac{\langle\frac{\alpha}{\varepsilon^4}\rangle}{\langle\frac{1}{\varepsilon}\rangle^4}\langle E_3\rangle^3. \tag{45}$$

The first term in each equation represents the linear response and it is in perfect agreement with the isotropic version of the classical Tartar (1979) result (see also Milton (2004)): the effective permittivity perpendicular to the layers is the harmonic mean of the permittivities of constituents; on the other hand, the effective permittivity along the axes parallel to the layers is the arithmetic mean of the permittivities of constituents. The other terms in Eq. (43) represent the effective nonlinear response of the laminated system, a peculiar result of our procedure. This nonlinear behavior is described by three parameters defined as follows

$$\mathcal{C}_i = \langle\frac{\alpha}{\varepsilon^{2i}}\rangle / \left[\langle\frac{1}{\varepsilon}\rangle^{2i}\right], \quad i = 0, 1, 2. \tag{46}$$

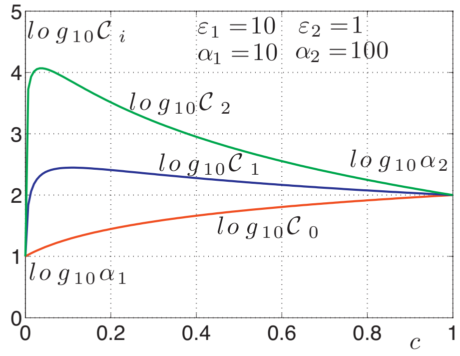

Fig. 4. Nonlinear response of the dielectric laminated system. The coefficients $\mathcal{C}_i$ are plotted versus the volume fraction c. We underline the amplification of the nonlinearities $\mathcal{C}_1 > \alpha_2$ and $\mathcal{C}_2 > \alpha_2$ for a given range of volume fraction.

In the specific case with two layers (with stoichiometric coefficients $\phi_1 = 1 - c$ and $\phi_2 = c$) we can write the explicit expressions for $\mathcal{C}_i$

$$\mathcal{C}_i = \left[(1-c)\frac{\alpha_1}{\varepsilon_1^{2i}} + c\frac{\alpha_2}{\varepsilon_2^{2i}}\right] / \left[(1-c)\frac{1}{\varepsilon_1} + c\frac{1}{\varepsilon_2}\right]^{2i}. \tag{47}$$

It is interesting to investigate the nonlinear results for materials satisfying $\varepsilon_1 \gg \varepsilon_2$ and $\alpha_1 \ll \alpha_2$; we have the approximation

$$\mathcal{C}_i \cong \left[c\frac{\alpha_2}{\varepsilon_2^{2i}}\right] / \left[c\frac{1}{\varepsilon_2}\right]^{2i} = \frac{\alpha_2}{c^{2i-1}}. \tag{48}$$

This expression leads to $\mathcal{C}_0 < \alpha_2$, $\mathcal{C}_1 > \alpha_2$ and $\mathcal{C}_2 > \alpha_2$ for a given range of volume fraction; it means that we can obtain an amplification of the nonlinear parameters $\mathcal{C}_1$ and $\mathcal{C}_2$. An example of such an amplification can be found in Fig. 4. Similar phenomena have been observed in different dielectric structures composed of nonlinear particles dispersed in a linear matrix (Giordano and Rocchia, 2005, 2006).

5.3. Pure elastic isotropic system: linear and nonlinear analysis

As before, we introduce the constitutive equation for an isotropic nonlinear elastic material. This case is described by the Green elasticity that is based on the following hypothesis: we suppose that the stress power, in a given deformation, is absorbed into a strain energy function $u(\hat{\epsilon})$, representing the density of elastic potential energy (Asaro and Lubarda, 2006; Landau and Lifshitz, 1986). The existence of such a function and the consideration of energy balance in the continuum, lead to the evolution equation $\frac{du(\hat{\epsilon})}{dt} = T_{ij}(\hat{\epsilon})\frac{d\epsilon_{ij}}{dt}$ affirming that the function $u(\hat{\epsilon})$ is an exact differential form such that $T_{ij}(\hat{\epsilon}) = \frac{\partial u(\hat{\epsilon})}{\partial\epsilon_{ij}}$ (Asaro and Lubarda, 2006). These relations are a particular case of Eqs. (1) and (2). From the thermodynamics point of view, the strain energy function can be identified with the internal energy per unit volume in an isentropic process, or with the Helmholtz free energy per unit volume in an isothermal process (Landau and Lifshitz, 1986). Such an approach can be further developed for isotropic media: in this case, the function $u(\hat{\epsilon})$ must satisfy the relation

$$u(\hat{\epsilon}) = u\left(\hat{\mathbb{R}}^T : \hat{\epsilon} : \hat{\mathbb{R}}\right), \tag{49}$$

for any rotation tensor $\hat{\mathbb{R}}$. If Eq. (49) is satisfied, then it follows that the function $u(\hat{\epsilon})$ can depend only on the principal invariants of the strain tensor

$$u = u\left(\mathrm{Tr}(\hat{\epsilon}), \mathrm{Tr}(\hat{\epsilon}^2), \mathrm{Tr}(\hat{\epsilon}^3)\right). \tag{50}$$

We may expand Eq. (50) up to the third order in the strain components, obtaining (Landau and Lifshitz, 1986)

$$u(\hat{\epsilon}) = \mu \text{Tr}\left(\hat{\epsilon}^2\right) + \frac{\lambda}{2}\left[\text{Tr}\left(\hat{\epsilon}\right)\right]^2 + \frac{A}{3}\text{Tr}\left(\hat{\epsilon}^3\right) \quad (51)$$

$$+B\text{Tr}\left(\hat{\epsilon}\right)\text{Tr}\left(\hat{\epsilon}^2\right) + \frac{C}{3}\left[\text{Tr}\left(\hat{\epsilon}\right)\right]^3,$$

where λ and μ are the standard Lamé coefficients of linear isotropic elasticity, and A, B and C are the so-called Landau moduli, measuring the first deviation from the linearity (Landau and Lifshitz, 1986; Giordano et al., 2008; Colombo and Giordano, 2011). We remark that the use of the energy function u or its Legendre transform w leads to the same formalism for the pure elastic case (see Eq. (3)). Finally, performing the derivatives, we obtain the nonlinear isotropic constitutive equation (within the Green approach) expanded up to the second order in the strain tensor

$$\hat{T} = 2\mu\hat{\epsilon} + \lambda\text{Tr}\left(\hat{\epsilon}\right)\hat{I} + A\hat{\epsilon}^2 \quad (52)$$

$$+B\left\{\text{Tr}\left(\hat{\epsilon}^2\right)\hat{I} + 2\hat{\epsilon}\text{Tr}\left(\hat{\epsilon}\right)\right\} + C\left[\text{Tr}\left(\hat{\epsilon}\right)\right]^2\hat{I}.$$

As an example, we homogenize a laminated structure ($\vec{n} = (0, 0, 1)$) where each layer is described by a model with only one nonlinear parameter, namely the Landau modulus C. Each layer is therefore described by the constitutive equation

$$\hat{T} = 2\mu_i\hat{\epsilon} + \lambda_i\text{Tr}\left(\hat{\epsilon}\right)\hat{I} + C_i\left[\text{Tr}\left(\hat{\epsilon}\right)\right]^2\hat{I}, \quad (53)$$

for $i = 1 \ldots N$, representing a particular case of Eq. (16). Since we are dealing with a pure elastic system, we can adopt the following simplified quantities

$$\mathcal{T} = \begin{pmatrix} T_{11} \\ T_{22} \\ T_{33} \\ T_{23} \\ T_{13} \\ T_{12} \end{pmatrix} = \begin{pmatrix} y_1 \\ y_2 \\ x_3 \\ x_4 \\ x_5 \\ y_6 \end{pmatrix}, \quad \mathcal{E} = \begin{pmatrix} \epsilon_{11} \\ \epsilon_{22} \\ \epsilon_{33} \\ \epsilon_{23} \\ \epsilon_{13} \\ \epsilon_{12} \end{pmatrix} = \begin{pmatrix} x_1 \\ x_2 \\ -y_3 \\ -y_4 \\ -y_5 \\ x_6 \end{pmatrix}, \quad (54)$$

corresponding to Eqs. (14) and (15) and the following ones

$$\mathcal{X} = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \\ x_5 \\ x_6 \end{pmatrix} = \begin{pmatrix} \epsilon_{11} \\ \epsilon_{22} \\ T_{33} \\ T_{23} \\ T_{13} \\ \epsilon_{12} \end{pmatrix}, \quad \mathcal{Y} = \begin{pmatrix} y_1 \\ y_2 \\ y_3 \\ y_4 \\ y_5 \\ y_6 \end{pmatrix} = \begin{pmatrix} T_{11} \\ T_{22} \\ -\epsilon_{33} \\ -\epsilon_{23} \\ -\epsilon_{13} \\ T_{12} \end{pmatrix}, \quad (55)$$

corresponding to Eqs. (12) and (13). By using Eq. (18) (i.e. by applying $\psi_{\vec{n}}$), we can eventually found the response of each layer in the form defined in Eq. (17), i.e. in the form $\mathcal{Y} = \mathcal{Y}(\mathcal{X})$

$$y_1 = \frac{4\mu_i(\lambda_i + \mu_i)x_1 + 2\lambda_i\mu_i x_2 + \lambda_i x_3}{2\mu_i + \lambda_i} + \frac{2C_i\mu_i(2\mu_i x_1 + 2\mu_i x_2 + x_3)^2}{(2\mu_i + \lambda_i)^3} \quad (56)$$

$$y_2 = \frac{2\lambda_i\mu_i x_1 + 4\mu_i(\lambda_i + \mu_i)x_2 + \lambda_i x_3}{2\mu_i + \lambda_i} + \frac{2C_i\mu_i(2\mu_i x_1 + 2\mu_i x_2 + x_3)^2}{(2\mu_i + \lambda_i)^3} \quad (57)$$

$$y_3 = \frac{\lambda_i x_1 + \lambda_i x_2 - x_3}{2\mu_i + \lambda_i} + \frac{C_i\mu_i(2\mu_i x_1 + 2\mu_i x_2 + x_3)^2}{(2\mu_i + \lambda_i)^3} \quad (58)$$

$$y_4 = -\frac{1}{2\mu_i}x_4 \quad (59)$$

$$y_5 = -\frac{1}{2\mu_i}x_5 \quad (60)$$

$$y_6 = 2\mu_i x_6 \quad (61)$$

Further, in order to perform the average defined in Eq. (20) we introduce the following parameters describing the linear behavior

$$\begin{aligned} a &= \langle\frac{4\mu(\mu+\lambda)}{2\mu+\lambda}\rangle, \quad b = \langle\frac{2\mu\lambda}{2\mu+\lambda}\rangle, \quad c = \langle\frac{\lambda}{2\mu+\lambda}\rangle, \\ d &= \langle\frac{1}{2\mu+\lambda}\rangle, \quad e = \langle\frac{1}{\mu}\rangle, \quad f = \langle\mu\rangle, \end{aligned} \quad (62)$$

where $\langle z\rangle = \sum_{i=1}^{N}\phi_i z_i$ is the weighted arithmetic mean (WAM) over the sequence of layers. On the other hand, for the nonlinear response we introduce the quantities

$$\begin{aligned} g &= \langle\frac{8\mu^3 C}{(2\mu+\lambda)^3}\rangle, \quad h = \langle\frac{4\mu^2 C}{(2\mu+\lambda)^3}\rangle, \\ q &= \langle\frac{2\mu C}{(2\mu+\lambda)^3}\rangle, \quad r = \langle\frac{C}{(2\mu+\lambda)^3}\rangle, \end{aligned} \quad (63)$$

depending on the Landau moduli C_i of all layers. Eqs. (62) and (63) correspond to Eq. (21). Hence, the averaged constitutive equation in the form $\langle\mathcal{Y}\rangle = \mathcal{Y}(\mathcal{X})$ can be written as

$$\langle y_1\rangle = ax_1 + bx_2 + cx_3 + g(x_1 + x_2)^2 + 2h(x_1 + x_2)x_3 + qx_3^2 \quad (64)$$

$$\langle y_2\rangle = bx_1 + ax_2 + cx_3 + g(x_1 + x_2)^2 + 2h(x_1 + x_2)x_3 + qx_3^2 \quad (65)$$

$$\langle y_3\rangle = cx_1 + cx_2 - dx_3 + h(x_1 + x_2)^2 + 2q(x_1 + x_2)x_3 + rx_3^2 \quad (66)$$

$$\langle y_4\rangle = -\frac{1}{2}ex_4 \quad (67)$$

$$\langle y_5\rangle = -\frac{1}{2}ex_5 \quad (68)$$

$$\langle y_6\rangle = 2fx_6 \quad (69)$$

The last step of the procedure consists in applying Eq. (22) (or, equivalently, $\psi_{\vec{n}}^{-1}$) in order to obtain the homogenized stress–strain relation in the final form

$$\begin{aligned} \langle T_{11}\rangle = &\left(a + \frac{c^2}{d}\right)\langle\epsilon_{11}\rangle + \left(b + \frac{c^2}{d}\right)\langle\epsilon_{22}\rangle + \frac{c^2}{d}\langle\epsilon_{33}\rangle \\ &+ \frac{3cd^2h + 3dc^2q + c^3r + d^3g}{d^3}(\langle\epsilon_{11}\rangle + \langle\epsilon_{22}\rangle)^2 \\ &+ 2\frac{2cdq + c^2r + d^2h}{d^3}(\langle\epsilon_{11}\rangle + \langle\epsilon_{22}\rangle)\langle\epsilon_{33}\rangle \\ &+ \frac{cr + dq}{d^3}\langle\epsilon_{33}\rangle^2 \end{aligned} \quad (70)$$

$$\langle T_{22}\rangle = \left(b+\frac{c^2}{d}\right)\langle\epsilon_{11}\rangle + \left(a+\frac{c^2}{d}\right)\langle\epsilon_{22}\rangle + \frac{c^2}{d}\langle\epsilon_{33}\rangle + \frac{3cd^2h+3dc^2q+c^3r+d^3g}{d^3}(\langle\epsilon_{11}\rangle+\langle\epsilon_{22}\rangle)^2 + 2\frac{2cdq+c^2r+d^2h}{d^3}(\langle\epsilon_{11}\rangle+\langle\epsilon_{22}\rangle)\langle\epsilon_{33}\rangle + \frac{cr+dq}{d^3}\langle\epsilon_{33}\rangle^2 \quad (71)$$

$$\langle T_{33}\rangle = \frac{c}{d}(\langle\epsilon_{11}\rangle+\langle\epsilon_{22}\rangle) + \frac{1}{d}\langle\epsilon_{33}\rangle + \frac{2cdq+c^2r+d^2h}{d^3}(\langle\epsilon_{11}\rangle+\langle\epsilon_{22}\rangle)^2 + 2\frac{cr+dq}{d^3}(\langle\epsilon_{11}\rangle+\langle\epsilon_{22}\rangle)\langle\epsilon_{33}\rangle + \frac{r}{d^3}\langle\epsilon_{33}\rangle^2 \quad (72)$$

$$\langle T_{23}\rangle = \frac{2}{e}\langle\epsilon_{23}\rangle \quad (73)$$

$$\langle T_{13}\rangle = \frac{2}{e}\langle\epsilon_{13}\rangle \quad (74)$$

$$\langle T_{12}\rangle = 2f\langle\epsilon_{12}\rangle \quad (75)$$

This completes the linear and nonlinear homogenization of the laminated structure. The stratified material is described by 5 linear parameters (please note that $a-b=2f$) and 4 nonlinear ones. The linear results are in perfect agreement with classical findings of Postma (1955) and Backus (1962); moreover, the nonlinear ones are new results obtained with our generalization. The same procedure can be applied to find the mixing laws of the other Landau parameters A and B.

Interesting enough, the resulting transversely isotropic material can be described, from the linear point of view, by the so called Hill parameters defined as follows

$$\mathcal{T} = \begin{pmatrix} k+m & k-m & l & 0 & 0 & 0 \\ k-m & k+m & l & 0 & 0 & 0 \\ l & l & n & 0 & 0 & 0 \\ 0 & 0 & 0 & 2m & 0 & 0 \\ 0 & 0 & 0 & 0 & 2p & 0 \\ 0 & 0 & 0 & 0 & 0 & 2p \end{pmatrix}\mathcal{E}. \quad (76)$$

They assume the following explicit expressions

$$k = \frac{a+b}{2} + \frac{c^2}{d} = \langle\frac{\mu(2\mu+3\lambda)}{2\mu+\lambda}\rangle + \langle\frac{\lambda}{2\mu+\lambda}\rangle^2\langle\frac{1}{2\mu+\lambda}\rangle^{-1}, \quad (77)$$

$$m = \frac{a-b}{2} = f = \langle\mu\rangle, \quad (78)$$

$$l = \frac{c}{d} = \langle\frac{\lambda}{2\mu+\lambda}\rangle\langle\frac{1}{2\mu+\lambda}\rangle^{-1}, \quad (79)$$

$$n = \frac{1}{d} = \langle\frac{1}{2\mu+\lambda}\rangle^{-1}, \quad (80)$$

$$p = \frac{1}{e} = \langle\frac{1}{\mu}\rangle^{-1}. \quad (81)$$

To conclude, the 4 nonlinear parameters are given below

$$\mathcal{C}_{111} = \frac{3cd^2h+3dc^2q+c^3r+d^3g}{d^3}, \quad (82)$$

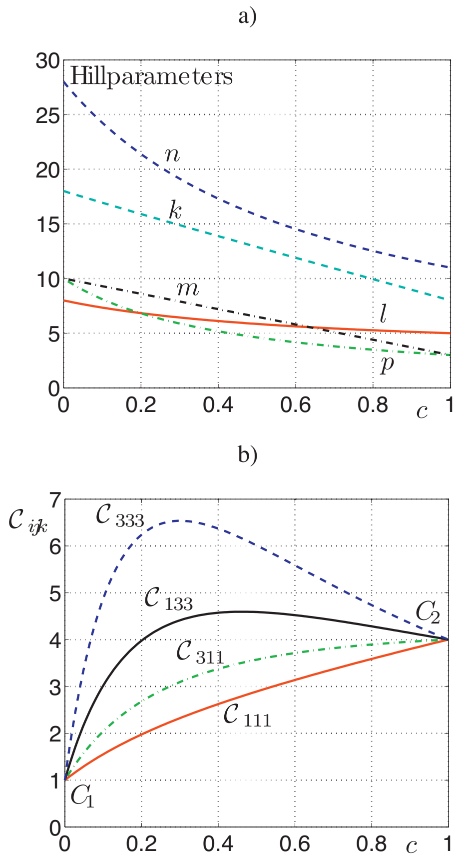

Fig. 5. Linear and nonlinear elastic response of a two-layer system defined by the following parameters: $\mu_1=10$, $\mu_2=3$, $\lambda_1=8$, $\lambda_2=5$, $C_1=1$ and $C_2=4$ (in arbitrary units). We represented the Hill parameters (a) and the nonlinear moduli $\mathcal{C}_{ijk}$ (b) versus the volume fraction c.

$$\mathcal{C}_{311} = \frac{2cdq+c^2r+d^2h}{d^3}, \quad (83)$$

$$\mathcal{C}_{133} = \frac{cr+dq}{d^3}, \quad (84)$$

$$\mathcal{C}_{333} = \frac{r}{d^3}. \quad (85)$$

An example of application of these results can be found in Fig. 5. The linear and nonlinear effective properties of a two-layer structure are shown versus the volume fraction c of the second phase. While we can observe a monotone behavior for the linear effective parameters, the system exhibits possible amplifications of the nonlinear coefficients. Similar intensification phenomena of the second order response have been observed in dispersions of nonlinear elastic particles embedded in a different matrix (Giordano et al., 2008, 2009; Colombo and Giordano, 2011; Giordano, 2013).

5.4. Magneto-electric isotropic system: linear and nonlinear analysis

We introduce a simple isotropic and nonlinear magneto-electric laminated system. To begin, we determine the most general isotropic nonlinear magneto-electric constitutive equation. The free energy w defined in Eq. (3) must be a scalar isotropic function of two vectors $\vec{E}$ and $\vec{H}$. It means that $w(\vec{E},\vec{H}) = w(\mathbb{R}^T:\vec{E},\mathbb{R}^T:\vec{H})$ for any rotation tensor $\hat{\mathbb{R}}$. Therefore, w can only depend on the invariants $\vec{E}\cdot\vec{E}$, $\vec{H}\cdot\vec{H}$ and $\vec{E}\cdot\vec{H}$. By considering the first terms in the series expansion of w we obtain the following mathematical form

$$w(\vec{E},\vec{H}) = -\frac{1}{2}\varepsilon\vec{E}\cdot\vec{E} - \frac{1}{2}\mu\vec{H}\cdot\vec{H} - g\vec{E}\cdot\vec{H} - \frac{1}{4}\alpha\left(\vec{E}\cdot\vec{E}\right)^2 - \frac{1}{4}\beta\left(\vec{H}\cdot\vec{H}\right)^2 - \frac{1}{2}\eta\left(\vec{E}\cdot\vec{H}\right)^2 - \gamma\left(\vec{E}\cdot\vec{H}\right)\left(\vec{E}\cdot\vec{E}\right) - \delta\left(\vec{E}\cdot\vec{H}\right)\left(\vec{H}\cdot\vec{H}\right) - \frac{1}{2}\xi\left(\vec{E}\cdot\vec{E}\right)\left(\vec{H}\cdot\vec{H}\right), \tag{86}$$

which corresponds to the following constitutive equations

$$D_i = \varepsilon E_i + gH_i + \alpha\left(\vec{E}\cdot\vec{E}\right)E_i + \eta\left(\vec{E}\cdot\vec{H}\right)H_i + \gamma\left(\vec{E}\cdot\vec{E}\right)H_i + 2\gamma\left(\vec{E}\cdot\vec{H}\right)E_i + \delta\left(\vec{H}\cdot\vec{H}\right)H_i + \xi\left(\vec{H}\cdot\vec{H}\right)E_i, \tag{87}$$

$$B_i = gE_i + \mu H_i + \beta\left(\vec{H}\cdot\vec{H}\right)H_i + \eta\left(\vec{E}\cdot\vec{H}\right)E_i + \delta\left(\vec{H}\cdot\vec{H}\right)E_i + 2\delta\left(\vec{E}\cdot\vec{H}\right)H_i + \gamma\left(\vec{E}\cdot\vec{E}\right)E_i + \xi\left(\vec{E}\cdot\vec{E}\right)H_i. \tag{88}$$

The possibility to obtain and to use these nonlinear terms in real systems was demonstrated by Rose et al. (2012) by means of metamaterials at microwave frequencies. In these systems, the nonlinear magnetoelectric coupling is detected through the second-harmonic generation in which incident magnetic fields at frequency ω drive an electric polarization at frequency 2ω. Here, we study a simple laminated system (along x_3) with only the nonlinear term described by α. This hypothesis allows us to solve the problem analytically. However, our procedure can be easily numerically implemented with all the possible combinations of nonlinear modes. Each layer is therefore governed by the laws

$$D_i = \varepsilon E_i + gH_i + \alpha\left(\vec{E}\cdot\vec{E}\right)E_i, \tag{89}$$

$$B_i = gE_i + \mu H_i. \tag{90}$$

In order to apply our homogenization procedure, we can adopt the following simplified quantities

$$\mathcal{K} = \begin{pmatrix} D_1\\ D_2\\ D_3\\ B_1\\ B_2\\ B_3\end{pmatrix} = \begin{pmatrix} y_1\\ y_2\\ x_3\\ y_4\\ y_5\\ x_6\end{pmatrix}, \quad \mathcal{Z} = \begin{pmatrix} E_1\\ E_2\\ E_3\\ H_1\\ H_2\\ H_3\end{pmatrix} = \begin{pmatrix} x_1\\ x_2\\ -y_3\\ x_4\\ x_5\\ -y_6\end{pmatrix}, \tag{91}$$

corresponding to Eqs. (14) and (15) and the following ones

$$\mathcal{X} = \begin{pmatrix} x_1\\ x_2\\ x_3\\ x_4\\ x_5\\ x_6\end{pmatrix} = \begin{pmatrix} E_1\\ E_2\\ D_3\\ H_1\\ H_2\\ B_3\end{pmatrix}, \quad \mathcal{Y} = \begin{pmatrix} y_1\\ y_2\\ y_3\\ y_4\\ y_5\\ y_6\end{pmatrix} = \begin{pmatrix} D_1\\ D_2\\ -E_3\\ B_1\\ B_2\\ -H_3\end{pmatrix}, \tag{92}$$

corresponding to Eqs. (12) and (13). By using Eq. (18), we can eventually found the response of each layer in the form defined in Eq. (17)

$$y_1 = \varepsilon x_1 + gx_4 + \alpha x_1\frac{\mathcal{N}}{\Delta^2}, \tag{93}$$

$$y_2 = \varepsilon x_2 + gx_5 + \alpha x_2\frac{\mathcal{N}}{\Delta^2}, \tag{94}$$

$$y_3 = \frac{-\mu x_3 + gx_6}{\Delta} + \alpha\mu(\mu x_3 - gx_6)\frac{\mathcal{N}}{\Delta^4}, \tag{95}$$

$$y_4 = \mu x_4 + gx_1, \tag{96}$$

$$y_5 = \mu x_5 + gx_2, \tag{97}$$

$$y_6 = \frac{-\varepsilon x_6 + gx_3}{\Delta} - \alpha g(\mu x_3 - gx_6)\frac{\mathcal{N}}{\Delta^4}, \tag{98}$$

where $\Delta = \varepsilon\mu - g^2$ and $\mathcal{N} = \Delta^2(x_1^2 + x_2^2) + (\mu x_3 - gx_6)^2$. To perform the average value defined in Eq. (20) (WAM) we introduce the following parameters

$$\varepsilon_1 = \langle\frac{\varepsilon}{\Delta}\rangle, \quad \mu_1 = \langle\frac{\mu}{\Delta}\rangle, \quad g_1 = \langle\frac{g}{\Delta}\rangle; \tag{99}$$

$$h_i = \langle\frac{\alpha g^i\mu^{2-i}}{\Delta^2}\rangle \quad \forall i = 0, 1, 2; \tag{100}$$

$$k_i = \langle\frac{\alpha g^i\mu^{4-i}}{\Delta^4}\rangle \quad \forall i = 0, 1, 2, 3, 4. \tag{101}$$

Therefore, we obtain the following averaged constitutive equation

$$\langle y_1\rangle = \langle\varepsilon\rangle x_1 + \langle g\rangle x_4 + \langle\alpha\rangle x_1(x_1^2 + x_2^2) + h_0x_1x_3^2 - 2h_1x_1x_3x_6 + h_2x_1x_6^2, \tag{102}$$

$$\langle y_2\rangle = \langle\varepsilon\rangle x_2 + \langle g\rangle x_5 + \langle\alpha\rangle x_2(x_1^2 + x_2^2) + h_0x_2x_3^2 - 2h_1x_2x_3x_6 + h_2x_2x_6^2, \tag{103}$$

$$\langle y_3\rangle = -\mu_1x_3 + g_1x_6 + h_0x_3(x_1^2 + x_2^2) - h_1x_6(x_1^2 + x_2^2) + k_0x_3^3 - 3k_1x_3^2x_6 + 3k_2x_3x_6^2 - k_3x_6^3, \tag{104}$$

$$\langle y_4\rangle = \langle\mu\rangle x_4 + \langle g\rangle x_1, \tag{105}$$

$$\langle y_5\rangle = \langle\mu\rangle x_5 + \langle g\rangle x_2, \tag{106}$$

$$\langle y_6\rangle = -\varepsilon_1x_6 + g_1x_3 - h_1x_3(x_1^2 + x_2^2) + h_2x_6(x_1^2 + x_2^2) - k_1x_3^3 + 3k_2x_3^2x_6 - 3k_3x_3x_6^2 + k_4x_6^3. \tag{107}$$

To conclude, we apply Eq. (22) to obtain, after very long but straightforward calculations, the homogenized relation in the final form

$$\langle D_1\rangle = \langle\varepsilon\rangle\langle E_1\rangle + \langle g\rangle\langle H_1\rangle + \langle\alpha\rangle\langle E_1\rangle(\langle E_1\rangle^2 + \langle E_2\rangle^2) + \mathcal{C}\langle E_1\rangle\langle E_3\rangle^2 + \mathcal{D}\langle E_1\rangle\langle H_3\rangle^2 + 2\mathcal{E}\langle E_1\rangle\langle E_3\rangle\langle H_3\rangle, \tag{108}$$

$$\langle D_2\rangle = \langle\varepsilon\rangle\langle E_2\rangle + \langle g\rangle\langle H_2\rangle + \langle\alpha\rangle\langle E_2\rangle(\langle E_1\rangle^2 + \langle E_2\rangle^2) + \mathcal{C}\langle E_2\rangle\langle E_3\rangle^2 + \mathcal{D}\langle E_2\rangle\langle H_3\rangle^2 + 2\mathcal{E}\langle E_2\rangle\langle E_3\rangle\langle H_3\rangle, \tag{109}$$

$$\langle D_3\rangle = \varepsilon_{eff}\langle E_3\rangle + g_{eff}\langle H_3\rangle + \mathcal{E}\langle E_3\rangle(\langle E_1\rangle^2 + \langle E_2\rangle^2) + \mathcal{D}\langle H_3\rangle(\langle E_1\rangle^2 + \langle E_2\rangle^2) + \mathcal{F}\langle E_3\rangle^3 + 3\mathcal{G}\langle E_3\rangle^2\langle H_3\rangle + 3\mathcal{H}\langle E_3\rangle\langle H_3\rangle^2 + \mathcal{I}\langle H_3\rangle^3, \tag{110}$$

$$\langle B_1\rangle = \langle\mu\rangle\langle H_1\rangle + \langle g\rangle\langle E_1\rangle, \tag{111}$$

$$\langle B_2\rangle = \langle\mu\rangle\langle H_2\rangle + \langle g\rangle\langle E_2\rangle, \tag{112}$$

$$\langle B_3\rangle = \mu_{eff}\langle H_3\rangle + g_{eff}\langle E_3\rangle + \mathcal{E}\langle E_3\rangle(\langle E_1\rangle^2+\langle E_2\rangle^2) + \mathcal{D}\langle H_3\rangle(\langle E_1\rangle^2 + \langle E_2\rangle^2) + \mathcal{L}\langle H_3\rangle^3 + 3\mathcal{H}\langle E_3\rangle^2\langle H_3\rangle + 3\mathcal{I}\langle E_3\rangle\langle H_3\rangle^2 + \mathcal{G}\langle E_3\rangle^3, \tag{113}$$

where the linear effective parameters are given by

$$\varepsilon_{eff} = \frac{\langle\frac{\varepsilon}{\varepsilon\mu-g^2}\rangle}{\langle\frac{\varepsilon}{\varepsilon\mu-g^2}\rangle\langle\frac{\mu}{\varepsilon\mu-g^2}\rangle - \langle\frac{g}{\varepsilon\mu-g^2}\rangle^2}, \tag{114}$$

$$\mu_{eff} = \frac{\langle\frac{\mu}{\varepsilon\mu-g^2}\rangle}{\langle\frac{\varepsilon}{\varepsilon\mu-g^2}\rangle\langle\frac{\mu}{\varepsilon\mu-g^2}\rangle - \langle\frac{g}{\varepsilon\mu-g^2}\rangle^2}, \tag{115}$$

$$g_{eff} = \frac{\langle\frac{g}{\varepsilon\mu-g^2}\rangle}{\langle\frac{\varepsilon}{\varepsilon\mu-g^2}\rangle\langle\frac{\mu}{\varepsilon\mu-g^2}\rangle - \langle\frac{g}{\varepsilon\mu-g^2}\rangle^2}, \tag{116}$$

and the nonlinear ones by the following expressions

$$\mathcal{C} = \frac{\sum_{i=0}^{2}(-1)^i\binom{2}{i}h_i g_1^i\varepsilon_1^{2-i}}{(\varepsilon_1\mu_1-g_1^2)^2}, \tag{117}$$

$$\mathcal{D} = \frac{\sum_{i=0}^{2}(-1)^i\binom{2}{i}h_i\mu_1^i g_1^{2-i}}{(\varepsilon_1\mu_1-g_1^2)^2}, \tag{118}$$

$$\mathcal{E} = \frac{g_1h_2\mu_1 + g_1h_0\varepsilon_1 - h_1\varepsilon_1\mu_1 - h_1g_1^2}{(\varepsilon_1\mu_1-g_1^2)^2}, \tag{119}$$

$$\mathcal{F} = \frac{\sum_{i=0}^{4}(-1)^i\binom{4}{i}k_i g_1^i\varepsilon_1^{4-i}}{(\varepsilon_1\mu_1-g_1^2)^4}, \tag{120}$$

$$\mathcal{G} = \frac{g_1\sum_{i=0}^{3}(-1)^i\binom{3}{i}k_i g_1^i\varepsilon_1^{3-i}}{(\varepsilon_1\mu_1-g_1^2)^4} - \frac{\mu_1\sum_{i=0}^{3}(-1)^i\binom{3}{i}k_{i+1} g_1^i\varepsilon_1^{3-i}}{(\varepsilon_1\mu_1-g_1^2)^4}, \tag{121}$$

$$\mathcal{H} = \frac{g_1^2\sum_{i=0}^{2}(-1)^i\binom{2}{i}k_i g_1^i\varepsilon_1^{2-i}}{(\varepsilon_1\mu_1-g_1^2)^4} - \frac{2\mu_1g_1\sum_{i=0}^{2}(-1)^i\binom{2}{i}k_{i+1} g_1^i\varepsilon_1^{2-i}}{(\varepsilon_1\mu_1-g_1^2)^4} + \frac{\mu_1^2\sum_{i=0}^{2}(-1)^i\binom{2}{i}k_{i+2} g_1^i\varepsilon_1^{2-i}}{(\varepsilon_1\mu_1-g_1^2)^4}, \tag{122}$$

$$\mathcal{I} = \frac{\varepsilon_1\sum_{i=0}^{3}(-1)^i\binom{3}{i}k_i g_1^{3-i}\mu_1^i}{(\varepsilon_1\mu_1-g_1^2)^4} - \frac{g_1\sum_{i=0}^{3}(-1)^i\binom{3}{i}k_{i+1} g_1^{3-i}\mu_1^i}{(\varepsilon_1\mu_1-g_1^2)^4}, \tag{123}$$

$$\mathcal{L} = \frac{\sum_{i=0}^{4}(-1)^i\binom{4}{i}k_i g_1^{4-i}\mu_1^i}{(\varepsilon_1\mu_1-g_1^2)^4}. \tag{124}$$

This is an example where our theory is able to provide new results for both the linear and the nonlinear effective response of the laminated structure. It interesting to observe that a single uncoupled nonlinear term (controlled by α) drives the emergence of a series of nonlinear modes, also with an explicit magneto-electric coupling not present in the original separated materials. Finally, we underline that, in absence of coupling (i.e. with $g=0$ everywhere), we obtain the same results of Section 5.2 for the nonlinear dielectric response and a pure linear behavior from the magnetic point of view, as expected.

6. Conclusions

We elaborated a homogenization scheme for nonlinear laminated materials. Each layer of these composite structures is described by a coupled magneto-electro-elastic behavior. The proposed technique is based on an extension of the Backus–Tartar method, i.e. on the separation between continuous and discontinuous variables across each interface. Following this idea, the nonlinear constitutive equation of all materials is written as a direct relationship between continuous and discontinuous quantities. This is the central point of the procedure, which simply leads to average (through a weighted arithmetic mean) these constitutive equations in order to find the overall behavior of the system. We firstly developed our method with a fixed direction of lamination and, afterwards, we generalized it for considering an arbitrary interfaces orientation. To do this, we exploited the tensor character of the entire procedure combined with a generalized rotation matrix, able to represent the physical properties of each layer in an arbitrary reference frame. We underline that all closed form expressions can be easily implemented in a software code through the standard operations of tensor calculus. Finally, we presented a series of examples of application of the theory, resolved with both numerical and analytical approaches.

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