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ON THE EFFECTIVE BEHAVIOR OF PERIODIC NONLINEAR ELECTRO-MECHANICAL COMPOSITES

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Abstract. *Here, the problem of estimating the effective behavior of nonlinear electro-mechanical composite materials with periodic microstructures is addressed. The approach adopted is a combination of asymptotic and variational homogenization techniques, which has provided remarkable results in previous applications to nonlinear uncoupled (dielectric and incompressible elastic) problems. Specifically, the nonlinear problem is addressed via a generalization to nonlinear electro-mechanics of the celebrated Hashin-Shtrikman variational methodology, which relies on the introduction of a comparison material taken here to be a linear piezoelectric composite exhibiting the same microstructure as the nonlinear composite under study. Then, the resulting estimates for the nonlinear effective behavior will depend on estimates of the effective behavior of the comparison material, which in turn are obtained via the corresponding applications of the well-known asymptotic homogenization method.*

Keywords: *composites, electro-mechanics, nonlinearity, homogenization*

1. INTRODUCTION

Starting on the information at hand, one of the ways of estimating the effective response of nonlinear composites bases on limiting, over all the microstructures consistent with such an information, the range of possible responses. Mathematically speaking, such a range takes the form of bounds which include the given information. Obtention and improvement of such bounds has been the focus of many efforts in the last decades. One of these, the generalization to nonlinear behavior of the Hashin-Strikman variational methodology, was originated in a work by Willis (1983) and it is employed mainly in bounding the effective energy density of nonlinear composites in various different physical frameworks. The nonlinear Hashin-Shtrikman variational methodology bases on the introduction of a comparison material which is taken to be linear in most cases, and its theoretical basis was rigorously stablished by Talbot & Willis (1985). An alternative approach, which was introduced by Ponte Castañeda (1991) consist on taking the comparison material to be a linear composite exhibiting the same microstructure as the nonlinear composite, was incorporated to the Hashin-Shtrikman methodology by Talbot & Willis (1992). The advantage of taking such a comparison composite is that the resulting bounds for the nonlinear composite will depend on the effective properties of the linear composite, which have been extensively studied. For instance, Talbot (1999, 2000) applied such a methodology to bound the effective energy density of nonlinear dielectric and incompressible elastic matrix-imclusion random composites, respectively, and estimated the effective dielectric constant and shear modulus of the comparison composites by using the linear bounds by Bruno (1991) and Bruno & Leo (1993), respectively. In the related studies by Pérez-Fernández et al. (2005, 2007) concerning periodic versions of the composites addressed by Talbot (1999, 2000), we use estimations of the effective properties of the linear comparison composites obtained via the asymptotic homogenization method by Rodríguez-Ramos et al. (2001) and Guinovart-Díaz et al. (2001), respectively, and compared the resulting nonlinear bounds to those using the corresponding versions of the linear bounds by Bruno (1991) and Bruno & Leo (1993), respectively. As periodicity is accounted for with greater accuracy, the bounds using the asymptotic homogenization estimations resulted in remarkable improvement over those using the bounds by Bruno (1991) and Bruno & Leo (1993) with extreme narrowing of the effective behavior range. Similar results were obtained by León-Mecías et al. (2008) and López-Realpozo et al. (2008) for other types of composites.

In this work, we present preliminary results concerning the extension to nonlinear coupled behavior of our approach combining nonlinear variational bounds with linear asymptotic homogenization estimations. The chosen framework is that of third-order electro-mechanics which include the nonlinear coupled effects of electrostriction and nonlinear piezoelectricity besides the nonlinear elastic and dielectric uncoupled effects together with the usual linear elastic, piezoelectric and dielectric effects. To the best of our knowledge, little has been done concerning both coupled behavior and nonlinearity. Some of these works are: Bisegna & Luciano (1996) generalized the Hashin-Strikman variational principles to electro-mechanics including nonlinear and nonlocal behavior but applied the developed methodology to study linear composites; Bustamante et al. (2006) obtained so-called universal relations for isotropic nonlinear incompressible magneto-elastic composites; Dokmeci (1988) obtained variational principles for piezoelectric materials with up to fourth-order elastic effects; He (2000) applied a semi-inverse method to obtain generalized variational principles for third-order piezoelectricity; Joshi (1992) obtained nonlinear constitutive relations for piezoelectricity; Li & Rao (2004) developed a micromechanical

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nonlinear model for electrostrictive composites; Rodríguez-Ramos et al. (2004) generalized the Hashin-Strikman variational principles to nonlinear piezoelectricity with an invariant formulation; Telega et al. (1998) applied $\Gamma-$convergence to homogenize periodic nonlinear piezoelectric composites.

This paper is organized as follows: section 2 deals with the setting of the problem; section 3 contains the development of the new bounds; section 4 presents an example dealing with centrosymmetric materials; and some concluding remarks are given in section 5.

2. PROBLEM SETTING

Consider a two-phase matrix-inclusion composite exhibiting nonlinear electro-mechanical constitutive behavior and periodic microstructure. It is assumed that matrix and inclusions are made of homogeneous materials occupying phases 1 and 2, respectively, so phase $r = 1, 2$ is described by the convex energy potential $W_r(\boldsymbol{\sigma}, \mathbf{E})$ and characteristic function $\chi_r(\mathbf{x})$, where $\boldsymbol{\sigma}$, $\mathbf{E}$ and $\mathbf{x}$ stand for the mechanical stress tensor, the electric field vector and the position vector, respectively. Then, the constitutive energy is given by the piecewise-convex potential

$$W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E}) = \chi_r(\mathbf{x}) W_r(\boldsymbol{\sigma}, \mathbf{E}), \tag{1}$$

where Einstein's summation convention over repeated indexes is adopted. Then, with the mean-value operator represented by angular brackets, the effective behavior is given by the effective energy potential $\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})$ defined via the minimum energy principle

$$\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) = \inf_{(\boldsymbol{\sigma}, \mathbf{E}) \in S(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})} \langle W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E}) \rangle, \tag{2}$$

where $S(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})$ is the admissibility set of divergence-free stress tensors $\boldsymbol{\sigma}$ and anti-irrotational electric fields $\mathbf{E}$, both $\Omega-$periodic, with mean values $\langle \boldsymbol{\sigma} \rangle = \bar{\boldsymbol{\sigma}}$ and $\langle \mathbf{E} \rangle = \bar{\mathbf{E}}$, respectively.

Now, consider the dual formulation by taking the Legendre transform $W^*(\mathbf{x}; \varepsilon, \mathbf{D})$ of $W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E})$, that is,

$$W^*(\mathbf{x}; \varepsilon, \mathbf{D}) = \sup_{(\boldsymbol{\sigma}, \mathbf{E})} \{ \varepsilon : \boldsymbol{\sigma} + \mathbf{D} \cdot \mathbf{E} - W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E}) \}, \tag{3}$$

where ε and $\mathbf{D}$ are, respectively, the mechanical strain tensor and the electric displacement vector. Then, the effective behavior can be stated by the complementary effective energy potential $\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}})$ defined via the complementary minimum energy principle

$$\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}}) = \inf_{(\varepsilon, \mathbf{D}) \in S^*(\bar{\varepsilon}, \bar{\mathbf{D}})} \left\langle W^*(\mathbf{x}; \bar{\varepsilon}, \bar{\mathbf{D}}) \right\rangle, \tag{4}$$

where $S^*(\bar{\varepsilon}, \bar{\mathbf{D}})$ is the admissibility set of compatible strain tensors ε and divergence-free electric displacements $\mathbf{D}$, both $\Omega-$periodic, with mean values $\langle \varepsilon \rangle = \bar{\varepsilon}$ and $\langle \mathbf{D} \rangle = \bar{\mathbf{D}}$, respectively, so the effective potentials are Legendre duals, that is,

$$\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}}) = \sup_{(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})} \left\{ \bar{\varepsilon} : \bar{\boldsymbol{\sigma}} + \bar{\mathbf{D}} \cdot \bar{\mathbf{E}} - \hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \right\}. \tag{5}$$

In what follows, it is assumed that the matrix material is linear whereas the inclusions exhibit third-order nonlinearity, that is,

$$W_1(\boldsymbol{\sigma}, \mathbf{E}) = \frac{1}{2} \boldsymbol{\sigma} : \mathbf{S}_1^{\mathbf{E}} : \boldsymbol{\sigma} + \mathbf{E} \cdot \mathbf{d}_1 : \boldsymbol{\sigma} + \frac{1}{2} \mathbf{E} \cdot \boldsymbol{\kappa}_1^{\boldsymbol{\sigma}} \cdot \mathbf{E} \equiv W_1^L(\boldsymbol{\sigma}, \mathbf{E}) \tag{6}$$

and (Joshi, 1992)

$$W_2(\boldsymbol{\sigma}, \mathbf{E}) = (W_2^L + W_2^N)(\boldsymbol{\sigma}, \mathbf{E}) \tag{7}$$

with

$$W_2^L(\boldsymbol{\sigma}, \mathbf{E}) = \frac{1}{2} \boldsymbol{\sigma} : \mathbf{S}_2^{\mathbf{E}} : \boldsymbol{\sigma} + \mathbf{E} \cdot \mathbf{d}_2 : \boldsymbol{\sigma} + \frac{1}{2} \mathbf{E} \cdot \boldsymbol{\kappa}_2^{\boldsymbol{\sigma}} \cdot \mathbf{E} \tag{8}$$

and

$$W_2^N(\boldsymbol{\sigma}, \mathbf{E}) = \frac{1}{6} \boldsymbol{\sigma} : \left(\mathcal{S}_2^{\mathbf{E}} : \boldsymbol{\sigma} \right) : \boldsymbol{\sigma} + \frac{1}{2} \mathbf{E} \cdot (\boldsymbol{\delta}_2 : \boldsymbol{\sigma}) : \boldsymbol{\sigma} + \frac{1}{2} \mathbf{E} \cdot (\mathbf{E} \cdot \mathcal{D}_2) : \boldsymbol{\sigma} + \frac{1}{6} \mathbf{E} \cdot (\mathcal{K}_2^{\boldsymbol{\sigma}} \cdot \mathbf{E}) \cdot \mathbf{E}, \tag{9}$$

so Eq. (1) specializes to

$$W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E}) = \chi_r(\mathbf{x}) W_r^L(\boldsymbol{\sigma}, \mathbf{E}) + \chi_2(\mathbf{x}) W_2^N(\boldsymbol{\sigma}, \mathbf{E}). \tag{10}$$

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In Eqs. (6)-(9), $\mathbf{S}_r^{\mathbf{E}}$, $\mathbf{d}_r$ and $\boldsymbol{\kappa}_r^{\boldsymbol{\sigma}}$ are the linear material properties of phase r, that is, the linear compliance tensor at constant electric field (indicated by superscript $\mathbf{E}$), the linear piezoelectric tensor and the linear dielectric tensor at constant stress (indicated by superscript $\boldsymbol{\sigma}$), respectively, whereas $\mathcal{S}_2^{\mathbf{E}}$, $\boldsymbol{\delta}_2$, $\mathcal{D}_2$ and $\mathcal{K}_2^{\boldsymbol{\sigma}}$ are the nonlinear material properties of phase 2, that is, the nonlinear compliance tensor at constant electric field, the nonlinear piezoelectric tensor, the electrostrictive tensor and the nonlinear dielectric tensor at constant stress, respectively. Single- and double-dot notations represent the usual vector and diadic products, respectively.

3. THE BOUNDS

3.1 Classical and elementary bounds

Taking $\boldsymbol{\sigma} = \bar{\boldsymbol{\sigma}}$ and $\mathbf{E} = \bar{\mathbf{E}}$ in Eq. (2), $\varepsilon = \bar{\varepsilon}$ and $\mathbf{D} = \bar{\mathbf{D}}$ in Eq. (4) and then applying Eq. (5) yields the classical Reuss-type lower and Voigt-type upper bounds of the electro-mechanical framework, that is,

$$\langle W^* \rangle^* (\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \le \hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \le \langle W \rangle (\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}), \tag{11}$$

which are valid for any composite having the constitutive behavior stated by $W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E})$ as the only microstructural information they contain is that of the phase volume fractions $c_r = \langle \chi_r(x) \rangle$, $r = 1, 2$. On the other hand, by applying Eq. (2) directly to Eq. (10) yields the elementary lower bound

$$\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \ge \hat{W}^L(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) + c_2 \min_{(\boldsymbol{\sigma}, \mathbf{E})} \left\{ W_2^N(\boldsymbol{\sigma}, \mathbf{E}) \right\}, \tag{12}$$

where $\hat{W}^L(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})$ is the effective energy potential of a linear composite with phase potentials given by W_r^L, $r = 1, 2$ (Eqs. (6) and (8)), and exhibiting the same microstructure as the nonlinear composite.

3.2 Improved bounds

3.2.1 A lower bound

Here, the Hashin-Shtrikman variational structure is generalized to nonlinear electro-mechanics. Consider a comparison linear piezoelectric composite with the same microstructure as the nonlinear composite and energy potential $W^0(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E})$. In this case, it is convenient to choose $W_1^0(\boldsymbol{\sigma}, \mathbf{E}) \equiv W_1^L(\boldsymbol{\sigma}, \mathbf{E})$ and

$$W_2^0(\boldsymbol{\sigma}, \mathbf{E}) = \frac{1}{2} \boldsymbol{\sigma} : \mathbf{S}_0^{\mathbf{E}} : \boldsymbol{\sigma} + \mathbf{E} \cdot \mathbf{d}_0 : \boldsymbol{\sigma} + \frac{1}{2} \mathbf{E} \cdot \boldsymbol{\kappa}_0^{\boldsymbol{\sigma}} \cdot \mathbf{E}, \tag{13}$$

where $\mathbf{S}_0^{\mathbf{E}}$, $\mathbf{d}_0$ and $\boldsymbol{\kappa}_0^{\boldsymbol{\sigma}}$ are regarded as free parameters to be chosen optimally later on. Now, by solving for $W(\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E})$ the definition

$$\left(W - W^0 \right)^* (\mathbf{x}; \mathbf{s}, \mathbf{e}) = \sup_{(\boldsymbol{\sigma}, \mathbf{E})} \left\{ \mathbf{s} : \boldsymbol{\sigma} + \mathbf{e} \cdot \mathbf{E} - \left(W - W^0 \right) (\mathbf{x}; \boldsymbol{\sigma}, \mathbf{E}) \right\}, \tag{14}$$

applying Eq. (2) and then taking $\mathbf{s} \equiv \mathbf{0}$ and $\mathbf{e} \equiv \mathbf{0}$, it follows the Ponte Castañeda-type lower bound

$$\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \ge \hat{W}^0(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) + c_2 \min_{(\boldsymbol{\sigma}, \mathbf{E})} \left\{ \left(W_2 - W_2^0 \right) (\boldsymbol{\sigma}, \mathbf{E}) \right\}, \tag{15}$$

where $\hat{W}^0(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})$ is the effective energy potential of the comparison composite. Notice that Eq. (15) can be maximize with respect to the parameters $\mathbf{S}_0^{\mathbf{E}}$, $\mathbf{d}_0$ and $\boldsymbol{\kappa}_0^{\boldsymbol{\sigma}}$, that is, to choose the comparison composite whose properties provide the best lower bound for $\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})$. Also, notice that Eq. (12) can be obtained from Eq. (15) by taking $\mathbf{S}_0^{\mathbf{E}} = \mathbf{S}_2^{\mathbf{E}}$, $\mathbf{d}_0 = \mathbf{d}_2$ and $\boldsymbol{\kappa}_0^{\boldsymbol{\sigma}} = \boldsymbol{\kappa}_2^{\boldsymbol{\sigma}}$.

3.2.2 An upper bound

It follows from the Legendre transform of $W_2(\boldsymbol{\sigma}, \mathbf{E})$ that, for any $(\boldsymbol{\sigma}, \mathbf{E})$,

$$W_2^*(\varepsilon, \mathbf{D}) \ge \varepsilon : \boldsymbol{\sigma} + \mathbf{D} \cdot \mathbf{E} - W_2(\boldsymbol{\sigma}, \mathbf{E}). \tag{16}$$

Then, by subsituting Eq. (16) into Eq. (4), it yields the following lower bound for $\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}})$:

$$\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}}) \ge \inf_{(\boldsymbol{\varepsilon}, \mathbf{D}) \in S^*(\bar{\boldsymbol{\varepsilon}}, \bar{\mathbf{D}})} \langle \chi_1(\mathbf{x}) W_1^*(\varepsilon, \mathbf{D}) + \chi_2(\mathbf{x}) \left[\varepsilon : \boldsymbol{\sigma} + \mathbf{D} \cdot \mathbf{E} - W_2(\boldsymbol{\sigma}, \mathbf{E}) \right] \rangle, \tag{17}$$

where

$$W_1^*(\varepsilon, \mathbf{D}) = \frac{1}{2} \varepsilon : \mathbf{C}_1^{\mathbf{D}} : \varepsilon - \mathbf{D} \cdot \mathbf{h}_1 : \varepsilon + \frac{1}{2} \mathbf{D} \cdot \boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}} \cdot \mathbf{D} \tag{18}$$

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is the Legendre transform of Eq. (6), with $\mathbf{C}_1^{\mathbf{D}}$, $\mathbf{h}_1$ and $\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}$ being the dual linear material properties of phase 1, that is, the linear rigidity tensor at constant electric displacement (indicated by superscript $\mathbf{D}$), the linear piezoelectric tensor and the linear dielectric tensor at constant strain (indicated by superscript ε), respectively, which are given by

$$\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}} = \left[\boldsymbol{\kappa}_1^{\boldsymbol{\sigma}} - \mathbf{d}_1 : \left[S_1^{\mathbf{E}}\right]^{-1} : \mathbf{d}_1^T\right]^{-1}, \mathbf{h}_1 = \boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}} \cdot \mathbf{d}_1 : \left[S_1^{\mathbf{E}}\right]^{-1}, \text{ and } \mathbf{C}_1^{\mathbf{D}} = \left[S_1^{\mathbf{E}}\right]^{-1} + \left[S_1^{\mathbf{E}}\right]^{-1} : \mathbf{d}_1^T \cdot \mathbf{h}_1, \tag{19}$$

where superscripts -1 and T stand for relevant tensor inversion and transposition, respectively. Now, by assuming that $(\boldsymbol{\sigma}, \mathbf{E})$ is constant, it follows from Eq. (17) that

$$\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}}) \geq \inf_{(\boldsymbol{\varepsilon},\mathbf{D})\in S^*(\bar{\varepsilon},\bar{\mathbf{D}})} \langle \chi_1(\mathbf{x}) W_1^*(\boldsymbol{\varepsilon}, \mathbf{D}) + \chi_2(\mathbf{x}) [\boldsymbol{\varepsilon} : \boldsymbol{\sigma} + \mathbf{D} \cdot \mathbf{E}] \rangle - c_2 W_2(\boldsymbol{\sigma}, \mathbf{E}). \tag{20}$$

Manipulation of Eq. (20) together with the application of a result of Talbot & Willis (1992) yields the lower bound

$$\hat{W}^*(\bar{\varepsilon}, \bar{\mathbf{D}}) \geq \hat{W}_-^*(\bar{\varepsilon}) + \hat{W}_-^*(\bar{\mathbf{D}}) - c_2 W_2(\boldsymbol{\sigma}, \mathbf{E}), \tag{21}$$

where

$$\hat{W}_-^*(\bar{\varepsilon}) = \frac{1}{2} c_1 \bar{\varepsilon} : \mathbf{C}_1^{\mathbf{D}} : \bar{\varepsilon} + \bar{\varepsilon} : \bar{\mathbf{s}} - \frac{1}{2} \left\{ \left[\bar{\varepsilon} + \left[\mathbf{C}_1^{\mathbf{D}}\right]^{-1} : (\mathbf{s}_1 - \mathbf{s}_2)\right] : \left(c_1 \mathbf{C}_1^{\mathbf{D}} - \hat{\mathbf{C}}_0^{\bar{\mathbf{D}}}\right) : \left[\bar{\varepsilon} + \left[\mathbf{C}_1^{\mathbf{D}}\right]^{-1} : (\mathbf{s}_1 - \mathbf{s}_2)\right] \right\} \tag{22}$$

and

$$\hat{W}_-^*(\bar{\mathbf{D}}) = \frac{1}{2} c_1 \bar{\mathbf{D}} \cdot \boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}} \cdot \bar{\mathbf{D}} + \bar{\mathbf{D}} \cdot \bar{\mathbf{e}} - \frac{1}{2} \left\{ \left[\bar{\mathbf{D}} + [\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}]^{-1} \cdot (\mathbf{e}_1 - \mathbf{e}_2)\right] \cdot \left(c_1 \boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}} - \hat{\boldsymbol{\beta}}_0^{\bar{\boldsymbol{\varepsilon}}}\right) \cdot \left[\bar{\mathbf{D}} + [\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}]^{-1} \cdot (\mathbf{e}_1 - \mathbf{e}_2)\right] \right\} \tag{23}$$

with the effective rigidity tensor $\hat{\mathbf{C}}_0^{\bar{\mathbf{D}}}$ and the effective inverse dielectric tensor $\hat{\boldsymbol{\beta}}_0^{\bar{\boldsymbol{\varepsilon}}}$ of the comparison composite (indicated by subscript 0), and

$$\bar{\mathbf{s}} = c_1 \mathbf{s}_1 + c_2 \mathbf{s}_2 \text{ and } \bar{\mathbf{e}} = c_1 \mathbf{e}_1 + c_2 \mathbf{e}_2. \tag{24}$$

Then, the upper bound for $\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}})$ follows by applying the Legendre transform to Eq. (21) with Eqs. (22) and (23), that is,

$$\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \leq \sup_{(\bar{\varepsilon},\bar{\mathbf{D}})} \left\{ \bar{\boldsymbol{\sigma}} : \bar{\varepsilon} + \bar{\mathbf{E}} \cdot \bar{\mathbf{D}} - \left(\hat{W}_-^*(\bar{\varepsilon}) + \hat{W}_-^*(\bar{\mathbf{D}}) - c_2 W_2(\boldsymbol{\sigma}, \mathbf{E}) \right) \right\}, \tag{25}$$

where the supremum is attained when

$$\bar{\varepsilon} = \left[\hat{\mathbf{C}}_0^{\bar{\mathbf{D}}}\right]^{-1} : (\bar{\boldsymbol{\sigma}} - \mathbf{s}_2) - \left[\mathbf{C}_1^{\mathbf{D}}\right]^{-1} : (\mathbf{s}_1 - \mathbf{s}_2) \text{ and } \bar{\varepsilon} = \left[\hat{\boldsymbol{\beta}}_0^{\bar{\boldsymbol{\varepsilon}}}\right]^{-1} \cdot \left(\bar{\mathbf{E}} - \mathbf{e}_2\right) - [\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}]^{-1} \cdot (\mathbf{e}_1 - \mathbf{e}_2). \tag{26}$$

Finally, substitution of Eqs. (26) into Eq. (21) yields the Talbot-type upper bound

$$\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \leq \hat{W}_+(\bar{\boldsymbol{\sigma}}) + \hat{W}_+(\bar{\mathbf{E}}) + c_2 W_2(\boldsymbol{\sigma}, \mathbf{E}), \tag{27}$$

where

$$\begin{aligned} \hat{W}_+(\bar{\boldsymbol{\sigma}}) &= \frac{1}{2} \bar{\boldsymbol{\sigma}} : \left[\hat{\mathbf{C}}_0^{\bar{\mathbf{D}}}\right]^{-1} : \bar{\boldsymbol{\sigma}} + \bar{\boldsymbol{\sigma}} : \left[\left[\mathbf{C}_1^{\mathbf{D}}\right]^{-1} - \left[\hat{\mathbf{C}}_0^{\bar{\mathbf{D}}}\right]^{-1}\right] : \boldsymbol{\sigma} - \frac{1}{2} \boldsymbol{\sigma} : \left[(1 + c_2) \left[\mathbf{C}_1^{\mathbf{D}}\right]^{-1} - \left[\hat{\mathbf{C}}_0^{\bar{\mathbf{D}}}\right]^{-1}\right] : \boldsymbol{\sigma} \\ &+ \left[\bar{\boldsymbol{\sigma}} - c_2 \boldsymbol{\sigma} + \frac{1}{2} c_1 \mathbf{p}\right] : \left[\mathbf{C}_1^{\mathbf{D}}\right]^{-1} : \mathbf{p} \end{aligned} \tag{28}$$

and

$$\begin{aligned} \hat{W}_+(\bar{\mathbf{E}}) &= \frac{1}{2} \bar{\mathbf{E}} \cdot \left[\hat{\boldsymbol{\beta}}_0^{\bar{\boldsymbol{\varepsilon}}}\right]^{-1} \cdot \bar{\mathbf{E}} + \bar{\mathbf{E}} \cdot \left[[\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}]^{-1} - \left[\hat{\boldsymbol{\beta}}_0^{\bar{\boldsymbol{\varepsilon}}}\right]^{-1}\right] \cdot \mathbf{E} - \frac{1}{2} \mathbf{E} \cdot \left[(1 + c_2) [\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}]^{-1} - \left[\hat{\boldsymbol{\beta}}_0^{\bar{\boldsymbol{\varepsilon}}}\right]^{-1}\right] \cdot \mathbf{E} \\ &+ \left[\bar{\mathbf{E}} - c_2 \mathbf{E} + \frac{1}{2} c_1 \mathbf{q}\right] \cdot [\boldsymbol{\beta}_1^{\boldsymbol{\varepsilon}}]^{-1} \cdot \mathbf{q} \end{aligned} \tag{29}$$

being $\mathbf{p}$ and $\mathbf{q}$ some tensor and vector parameters, respectively, related to the linear electro-mechanical coupling. Specifically, $\mathbf{p} = \mathbf{0}$ and $\mathbf{q} = \mathbf{0}$ for uncoupled linear behavior. Notice that Eq. (27) can be minimize with respect to the $\boldsymbol{\sigma}$, $\mathbf{E}$, $\mathbf{p}$ and $\mathbf{q}$.

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4. AN EXAMPLE

4.1 Composites with centrosymmetric phases

In this case, the only coupling between mechanical and electric magnitudes occurs in the inclusions material through the electrostrictive effect. In this example, it is also assumed that electrostriction is the only nonlinearity present. Then, Eqs. (6)-(9) become

$$W_1(\boldsymbol{\sigma}, \mathbf{E}) = \frac{1}{2}\boldsymbol{\sigma} : \mathbf{S}_1^{\mathbf{E}} : \boldsymbol{\sigma} + \frac{1}{2}\mathbf{E} \cdot \boldsymbol{\kappa}_1^{\boldsymbol{\sigma}} \cdot \mathbf{E} \tag{30}$$

and

$$W_2(\boldsymbol{\sigma}, \mathbf{E}) = \frac{1}{2}\boldsymbol{\sigma} : \mathbf{S}_2^{\mathbf{E}} : \boldsymbol{\sigma} + \frac{1}{2}\mathbf{E} \cdot \boldsymbol{\kappa}_2^{\boldsymbol{\sigma}} \cdot \mathbf{E} + \frac{1}{2}\mathbf{E} \cdot (\mathbf{E} \cdot \mathcal{D}_2) : \boldsymbol{\sigma}. \tag{31}$$

Then, we choose the comparison composite to have also uncoupled behavior, that is, $\mathbf{d}_0 = \mathbf{0}$, so Eq. (15) specializes to

$$\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \geq \frac{1}{2}\bar{\boldsymbol{\sigma}} : \hat{\mathbf{S}}_0^{\bar{\mathbf{E}}} : \bar{\boldsymbol{\sigma}} + \frac{1}{2}\bar{\mathbf{E}} \cdot \hat{\boldsymbol{\kappa}}_0^{\bar{\boldsymbol{\sigma}}} \cdot \bar{\mathbf{E}} + \frac{1}{2}c_2\left(\boldsymbol{\kappa}_2^{\boldsymbol{\sigma}} - \boldsymbol{\kappa}_0^{\boldsymbol{\sigma}}\right) : \mathcal{D}_2^{-1} : \left(\mathbf{S}_2^{\mathbf{E}} - \mathbf{S}_0^{\mathbf{E}}\right) : \left[\mathcal{D}_2^{-1}\right]^T : \left(\boldsymbol{\kappa}_2^{\boldsymbol{\sigma}} - \boldsymbol{\kappa}_0^{\boldsymbol{\sigma}}\right), \tag{32}$$

where superscript -1 indicates inversion with respect to the mechanical indexes while superscript T stands for transposition of the mechanical indexes with respect to the electric indexes.

On the other hand, in view of the presence of linear uncoupling, Eq. (27) specializes to

$$\begin{aligned}
\hat{W}(\bar{\boldsymbol{\sigma}}, \bar{\mathbf{E}}) \quad \leq \quad & \frac{1}{2}\bar{\boldsymbol{\sigma}} : \hat{\mathbf{S}}_0^{\bar{\mathbf{E}}} : \bar{\boldsymbol{\sigma}} + \bar{\boldsymbol{\sigma}} : \left[\mathbf{S}_1^{\mathbf{E}} - \hat{\mathbf{S}}_0^{\bar{\mathbf{E}}}\right] : \boldsymbol{\sigma} - \frac{1}{2}\boldsymbol{\sigma} : \left[(1+c_2)\mathbf{S}_1^{\mathbf{E}} - \hat{\mathbf{S}}_0^{\bar{\mathbf{E}}}\right] : \boldsymbol{\sigma} \\
+ \quad & \frac{1}{2}\bar{\mathbf{E}} \cdot \hat{\boldsymbol{\kappa}}_0^{\bar{\boldsymbol{\sigma}}} \cdot \bar{\mathbf{E}} + \bar{\mathbf{E}} \cdot \left[\boldsymbol{\kappa}_1^{\boldsymbol{\sigma}} - \hat{\boldsymbol{\kappa}}_0^{\bar{\boldsymbol{\sigma}}}\right] \cdot \mathbf{E} - \frac{1}{2}\mathbf{E} \cdot \left[(1+c_2)\boldsymbol{\kappa}_1^{\boldsymbol{\sigma}} - \hat{\boldsymbol{\kappa}}_0^{\bar{\boldsymbol{\sigma}}}\right] \cdot \mathbf{E} \quad . \\
+ \quad & c_2\left[\frac{1}{2}\boldsymbol{\sigma} : \mathbf{S}_2^{\mathbf{E}} : \boldsymbol{\sigma} + \frac{1}{2}\mathbf{E} \cdot \boldsymbol{\kappa}_2^{\boldsymbol{\sigma}} \cdot \mathbf{E} + \frac{1}{2}\mathbf{E} \cdot (\mathbf{E} \cdot \mathcal{D}_2) : \boldsymbol{\sigma}\right]
\end{aligned} \tag{33}$$

4.2 Discussion

Both Eqs. (32) and (33) depend on the linear effective properties $\hat{\mathbf{S}}_0^{\bar{\mathbf{E}}}$ and $\hat{\boldsymbol{\kappa}}_0^{\bar{\boldsymbol{\sigma}}}$, which are different for each bound, and for which estimations must be available before carrying out the optimization procedures mentioned above. In this case, as there is no linear coupling, we can use the asymptotic homogenization estimations developed for purely elastic and dielectric composites. For instance, the estimations obtained by Rodríguez-Ramos et al. (2001) and Guinovart-Díaz et al. (2001) for fiber-reinforced composites with square and hexagonal periodic distributions can be employed.

5. CONCLUSIONS

Here, we presented a generalization of the Hashin-Shtrikman variational methodology to nonlinear electro-mechanics from which bounds on the effective energy of nonlinear composites are obtained. Particular realizations of the bounds for two-phase materials with centrosymmetric constituents are provided and their implementation is discussed.

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