

## HOMOGENIZATION OF ELASTIC DIELECTRIC COMPOSITES WITH RAPIDLY OSCILLATING PASSIVE AND ACTIVE SOURCE TERMS\*

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**Abstract.** This paper presents the derivation of the homogenized equations for the macroscopic response of elastic dielectric composites containing space charges (i.e., electric source terms) that oscillate rapidly at the length scale of the microstructure. The derivation is carried out in the setting of small deformations and moderate electric fields by means of a two-scale asymptotic analysis. Two types of rapidly oscillating space charges are considered: passive and active. The latter type corresponds to space charges that appear within the composite in response to externally applied electrical stimuli, while the former corresponds to space charges that are present within the composite from the outset. The obtained homogenized equations reveal that the presence of (passive or active) space charges within elastic dielectric composites can have a significant and even dominant effect on their macroscopic response, possibly leading to extreme behaviors ranging from unusually large permittivities and electrostriction coefficients to metamaterial-type properties featuring negative permittivities. These results suggest a promising strategy to design deformable dielectric composites—such as electrets and dielectric elastomer composites—with exceptional electromechanical properties.

**Key words.** electrets, dielectric elastomer composites, metamaterials, multiscale asymptotic expansions

**AMS subject classifications.** 35B27, 35J47, 35J66, 35Q74

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**1. The problem.** In this paper, we derive the homogenized equations governing the macroscopic response of elastic dielectric composites, containing space charges that oscillate at the length scale of the microstructure, in the so-called limit of small deformations and moderate electric fields. The focus is on elastic dielectric composites with even electromechanical coupling (such type of deformable dielectrics are often times referred to in the literature as dielectric elastomers) and periodic microstructure, which contain rapidly oscillating space charges of two types: *passive* and *active*.

Passive space charges refer to space charges that are present within the elastic dielectric composite from the outset, in its ground state. A prominent class of materials that can be viewed as elastic dielectric composites containing passive space charges is electrets (see, e.g., [18, 5, 15, 8]). On the other hand, active space charges refer to space charges that are *not* present within the elastic dielectric composite in its ground state. Instead, they appear within the composite as a result of externally applied stimuli, for instance, by a charge injection process [21, 30]. Dielectric elastomers filled with (semi)conducting or high-dielectric nanoparticles are thought to be an example of such a class of materials [25, 21, 26, 20]. In this work, we shall consider active charges that appear in proportionality to the electric field induced within the composite by externally applied electrical stimuli.

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*Ergo*, consider an elastic dielectric composite with periodic microstructure of period  $\delta$  that occupies a bounded domain  $\Omega \subset \mathbb{R}^N$  ( $N = 1, 2, 3$ ), with smooth boundary  $\partial\Omega$  and closure  $\bar{\Omega} = \Omega \cup \partial\Omega$ , in its undeformed configuration; throughout this work, again, attention is restricted to elastic dielectrics with *even* electromechanical coupling. In the classical setting of small deformations and moderate electric fields (see the Appendix), the permittivity, elasticity, and electrostriction tensors that characterize the local elastic dielectric response of the composite at any material point  $\mathbf{X} \in \Omega$  are taken to be, without loss of generality and with help of the notation  $Y = (0, 1)^N$ , of the forms

$$(1) \quad \begin{aligned} \varepsilon_{ij}^\delta(\mathbf{X}) &\in \mathbb{R}, & \varepsilon_{ij}^\delta(\mathbf{X}) &= \varepsilon_{ij}(\delta^{-1}\mathbf{X}) \quad \text{with } \varepsilon_{ij}(\mathbf{y}) \text{ } Y\text{-periodic,} \\ L_{ijkl}^\delta(\mathbf{X}) &\in \mathbb{R}, & L_{ijkl}^\delta(\mathbf{X}) &= L_{ijkl}(\delta^{-1}\mathbf{X}) \quad \text{with } L_{ijkl}(\mathbf{y}) \text{ } Y\text{-periodic,} \\ M_{ijkl}^\delta(\mathbf{X}) &\in \mathbb{R}, & M_{ijkl}^\delta(\mathbf{X}) &= M_{ijkl}(\delta^{-1}\mathbf{X}) \quad \text{with } M_{ijkl}(\mathbf{y}) \text{ } Y\text{-periodic,} \end{aligned}$$

respectively. Basic physical considerations dictate that<sup>1</sup>

$$(2) \quad \varepsilon_{ij}^\delta = \varepsilon_{ji}^\delta, \quad \varepsilon_{ij}^\delta \xi_i \xi_j \geq \varepsilon_0 \xi_k \xi_k \quad \forall \xi \in \mathbb{R}^N,$$

$$(3) \quad L_{ijkl}^\delta = L_{klij}^\delta = L_{jikl}^\delta = L_{ijlk}^\delta, \quad L_{ijkl}^\delta \Xi_{ij} \Xi_{kl} \geq \theta \Xi_{pq} \Xi_{pq} \quad \forall \Xi \in \mathbb{R}^{N \times N},$$

$$(4) \quad M_{ijkl}^\delta = M_{jikl}^\delta = M_{ijlk}^\delta,$$

where  $\varepsilon_0 \approx 8.85 \times 10^{-12}$  F/m stands for the permittivity of vacuum and  $\theta$  is some positive constant, namely, the smallest eigenvalue of  $L_{ijkl}^\delta$ , which is required to be positive. For mathematical expediency, we assume the following regularity properties:

$$(5) \quad \varepsilon_{ij}^\delta \in \mathcal{C}^1(\bar{\Omega}), \quad L_{ijkl}^\delta \in \mathcal{L}^\infty(\Omega), \quad M_{ijkl}^\delta \in \mathcal{L}^\infty(\Omega).$$

Here, we remark that the relatively strong regularity (5)<sub>1</sub> of the components of the permittivity tensor  $\varepsilon^\delta(\mathbf{X})$  is invoked in order to leverage standard theorems that will warrant mathematical well-posedness; more precisely, as elaborated below, the regularity (5)<sub>1</sub> is invoked here in order to obtain the sufficient regularity for the electric fields needed to prove existence of solution for the mechanical fields via the Lax–Milgram theorem. Such a regularity can be relaxed to allow for a general class of piecewise constant values of  $\varepsilon^\delta(\mathbf{X})$ —for instance, the piecewise constant values of  $\varepsilon^\delta(\mathbf{X})$  associated with particulate composites wherein the inclusions have smooth boundaries—at the expense of possibly invoking more technical theorems (see, e.g., [4, 22]). We assume further that the composite is subjected to a prescribed electric potential and a prescribed displacement,

$$(6) \quad \phi \in H^{3/2}(\partial\Omega) \quad \text{and} \quad \mathbf{v} \in H^{1/2}(\partial\Omega; \mathbb{R}^N),$$

on the entirety of its boundary  $\partial\Omega$ ; Neumann or mixed boundary conditions could be considered at no significant further conceptual expense. Moreover, we assume that the composite contains a distribution of space charges with density (per unit undeformed volume)

$$(7) \quad q^\delta \in \mathcal{L}^2(\Omega).$$

Figure 1 illustrates a schematic of the composite and of its microstructure and space charge content.

<sup>1</sup>Throughout this work we make use of the Einstein summation convention.

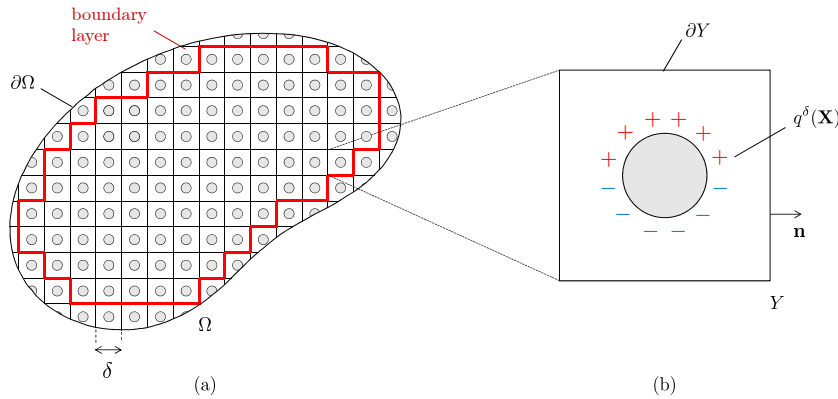


FIG. 1. (a) Schematic of the elastic dielectric composite in its undeformed configuration  $\Omega$ ; the boundary layer of incomplete unit cells needed to conform with the arbitrary geometry of its boundary  $\partial\Omega$  is marked in red. (b) Schematic of the unit cell  $Y$  that defines the periodic microstructure (of period  $\delta$ ) of the composite with the explicit illustration of the distribution of space charges characterized by the space-charge density  $q^\delta(\mathbf{X})$ .

In the limit of small deformations and moderate electric fields (see the Appendix), the relevant equations of Maxwell and of balance of linear momentum can be shown to reduce to the following *one-way coupled* boundary-value problems:

$$(8) \quad \begin{cases} \frac{\partial}{\partial X_i} \left[ -\varepsilon_{ij}^\delta(\mathbf{X}) \frac{\partial \varphi^\delta}{\partial X_j}(\mathbf{X}) \right] = q^\delta(\mathbf{X}), & \mathbf{X} \in \Omega, \\ \varphi^\delta(\mathbf{X}) = \phi(\mathbf{X}), & \mathbf{X} \in \partial\Omega \end{cases}$$

and

$$(9) \quad \begin{cases} \frac{\partial}{\partial X_j} \left[ L_{ijkl}^\delta(\mathbf{X}) \frac{\partial u_k^\delta}{\partial X_l}(\mathbf{X}) + M_{ijkl}^\delta(\mathbf{X}) \frac{\partial \varphi^\delta}{\partial X_k}(\mathbf{X}) \frac{\partial \varphi^\delta}{\partial X_l}(\mathbf{X}) \right] = 0, & \mathbf{X} \in \Omega, \\ u_i^\delta(\mathbf{X}) = v_i(\mathbf{X}), & \mathbf{X} \in \partial\Omega \end{cases}$$

for the electric potential  $\varphi^\delta(\mathbf{X})$  and the displacement field  $\mathbf{u}^\delta(\mathbf{X})$ . The PDE (8)<sub>1</sub> is the standard equation that governs the electrostatic field within a dielectric medium that contains a distribution of space charges. We remark that its restriction to the domain  $\Omega$  occupied by the solid (as opposed to the entire space  $\mathbb{R}^N$  where Maxwell's equations ought to be solved) is sufficient in the present context thanks to the prescription of the Dirichlet boundary condition (8)<sub>2</sub>. On the other hand, the PDE (9)<sub>1</sub> governs the deformation of the solid that results from the electric field in addition to the applied displacement boundary condition (9)<sub>2</sub>.

From a mathematical point of view, we remark that while the coupled system of boundary-value problems (8)–(9) is *nonlinear*, the boundary-value problem (8) is *linear* in the electric potential  $\varphi^\delta(\mathbf{X})$  and the boundary-value problem (9) is *linear* in the displacement field  $\mathbf{u}^\delta(\mathbf{X})$ . For any fixed  $\delta > 0$  then, granted the ellipticity (2)<sub>2</sub> and regularity (5)<sub>1</sub> of the components of the permittivity tensor  $\varepsilon^\delta(\mathbf{X})$ , the properties (6)<sub>1</sub> and (7) of the boundary data and source term, and the smoothness of  $\partial\Omega$ , the Lax–Milgram theorem ensures existence and uniqueness of the solution of (8) for  $\varphi^\delta(\mathbf{X})$  in the Sobolev space  $H^1(\Omega)$ . The regularity (5)<sub>1</sub>, (6)<sub>1</sub>, (7) together with the smoothness of  $\partial\Omega$  imply in fact the stronger regularity result that  $\varphi^\delta \in H^2(\Omega)$ ,

and hence that  $\text{Grad}\varphi^\delta \in H^1(\Omega; \mathbb{R}^N) \subset \mathcal{L}^4(\Omega; \mathbb{R}^N)$ ; see, e.g., Chapter 8 in [12], Chapter 6.3 in [10], Theorem 8.3 in [23], and Theorem 9.16 in [7]. In turn, granted the ellipticity (3)<sub>2</sub> and boundedness (5)<sub>2</sub> of the components of the elasticity tensor  $\mathbf{L}^\delta(\mathbf{X})$ , the boundedness (5)<sub>3</sub> of the components of the electrostriction tensor  $\mathbf{M}^\delta(\mathbf{X})$ , the fact that  $\text{Grad}\varphi^\delta \in \mathcal{L}^4(\Omega; \mathbb{R}^N)$  so that by (the generalized) Hölder’s inequality  $\text{Grad}\varphi^\delta \otimes \text{Grad}\varphi^\delta \in \mathcal{L}^2(\Omega; \mathbb{R}^{N \times N})$ , the regularity (6)<sub>2</sub> of the boundary data, and the smoothness of  $\partial\Omega$ , the Lax–Milgram theorem ensures existence and uniqueness of the solution of (9) for the displacement field  $\mathbf{u}^\delta(\mathbf{X})$  in the Sobolev space  $H^1(\Omega; \mathbb{R}^N)$ .

A specific class of space-charge densities  $q^\delta(\mathbf{X})$ . In this work, we shall restrict attention to space-charge densities  $q^\delta(\mathbf{X})$  of the following divergence form:

$$(10) \quad \begin{aligned} q^\delta(\mathbf{X}) &= -\delta \frac{\partial}{\partial X_i} \left[ f_k(\mathbf{X}) \frac{\partial}{\partial X_i} [\psi_k(\delta^{-1}\mathbf{X})] \right] \\ &= \delta^{-1} f_k(\mathbf{X}) g_k(\delta^{-1}\mathbf{X}) - \frac{\partial f_k}{\partial X_i}(\mathbf{X}) \tau_{ki}(\delta^{-1}\mathbf{X}). \end{aligned}$$

Here,

$$(11) \quad \mathbf{f} \in H^2(\Omega; \mathbb{R}^N), \quad \mathbf{g} \text{ is } Y\text{-periodic}, \quad \mathbf{g} \in \mathcal{L}^\infty(Y; \mathbb{R}^N), \quad \int_Y \mathbf{g}(\mathbf{y}) d\mathbf{y} = \mathbf{0},$$

and  $\tau_{ki}(\mathbf{y}) = \partial\psi_k(\mathbf{y})/\partial y_i$  with  $\psi(\mathbf{y})$  defined in terms of  $\mathbf{g}(\mathbf{y})$  as the unique solution in  $H^2(Y; \mathbb{R}^N)$  of the linear elliptic boundary-value problem

$$(12) \quad \begin{cases} -\frac{\partial^2 \psi_k}{\partial y_i \partial y_i}(\mathbf{y}) = g_k(\mathbf{y}), & \mathbf{y} \in Y, \\ -\frac{\partial \psi_k}{\partial y_i}(\mathbf{y}) n_i = 0, & \mathbf{y} \in \partial Y, \\ \int_Y \psi_k(\mathbf{y}) d\mathbf{y} = 0, \end{cases}$$

where  $\mathbf{n}$  in (12)<sub>2</sub> stands for the outward unit normal to the boundary  $\partial Y$  of the unit cell  $Y$  (see Figure 1(b)).

The choice (10) with (11)<sub>2,4</sub> and (12) of space-charge density is motivated by physical requirements/observations as well as by mathematical expediency. Indeed, the divergence form (10) together with the zero-average condition (11)<sub>4</sub> and the boundary condition (12)<sub>2</sub> ensure global charge neutrality in  $\Omega$  up to a boundary layer of thickness  $\delta$  (see Figure 1(a)). Moreover, the leading order term in (10) being  $O(\delta^{-1})$  implies that the content of charges at the “microscopic” length scale  $\delta$  remains finite even in the limit as  $\delta \rightarrow 0$  (in this limit, the space-charge density  $q^\delta(\mathbf{X})$  blows up within a vanishingly small volume to lead to a microscopic distribution of finite charges), consistent with physical expectations. We further remark that the form (10) comprises two constitutive inputs: the functions  $\mathbf{f}(\mathbf{X})$  and  $\mathbf{g}(\delta^{-1}\mathbf{X})$ . Roughly speaking, the latter dictates the local distribution of charges at the microscopic length scale  $\delta$  of each unit cell. The former, on the other hand, dictates the possibly nonuniform distribution of charges at the macroscopic length scale of  $\Omega$ . Finally, it is also interesting to note that source terms of the asymptotic form (10)—with leading  $O(\delta^{-1})$  and correction  $O(\delta^0)$ —can appear naturally when converting elliptic boundary-value problems with nonhomogeneous Dirichlet boundary conditions to problems with homogeneous Dirichlet boundary conditions; see, e.g. section 18 of Chapter 1 in the monograph by Bensoussan et al. [6].

The first objective of this work is to determine the elastic dielectric behavior of the above-defined composite, as governed by the coupled boundary-value problems

(8)–(9), for arbitrary but fixed or *passive* distribution of space charges with density of the form (10) in the limit when the period of the microstructure  $\delta \rightarrow 0$ . Given that the permittivity, elasticity, and electrostriction tensors (1) of the composite, as well as the distribution of space charges (10) in it, vary spatially at the length scale of  $\delta$ , one expects the electric potential  $\varphi^\delta(\mathbf{X})$  and the displacement field  $\mathbf{u}^\delta(\mathbf{X})$  to oscillate rapidly around smoothly varying macroscopic fields in such a limit. We show in section 2 that this is indeed the case and work out the governing equations—that is, the *homogenized equations*—for these macroscopic fields. The second objective of this work is to determine the homogenized equations resulting from (8)–(9) for the case when the space-charge density (10) is not fixed but *active*, in the sense that it is taken to depend on the resulting macroscopic field for the electric potential  $\varphi^\delta(\mathbf{X})$ . We work out the pertinent derivation in section 3. With the compound purpose of demonstrating the use of the resulting homogenized equations and of illustrating the dominant effect that space charges can have on the macroscopic behavior, we conclude by presenting in section 4 sample results for a porous electret containing passive charges on the walls of the pores.

**2. Passive charges: The limit as  $\delta \rightarrow 0$  by the method of two-scale asymptotic expansions.** In this section, we present the derivation of the homogenized equations that emerge from the boundary-value problems (8)–(9) in the limit as  $\delta \rightarrow 0$  by means of the method of two-scale asymptotic expansions [31, 6]. In the present context, this method amounts to looking for an asymptotic solution of the equations (8)–(9) as  $\delta \rightarrow 0$  of the form

$$(13) \quad \varphi^\delta(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k \varphi^{(k)}(\mathbf{X}, \delta^{-1}\mathbf{X}) \quad \text{and} \quad u_i^\delta(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k u_i^{(k)}(\mathbf{X}, \delta^{-1}\mathbf{X}),$$

where the functions  $\varphi^{(k)}(\mathbf{X}, \delta^{-1}\mathbf{X})$  and  $\mathbf{u}^{(k)}(\mathbf{X}, \delta^{-1}\mathbf{X})$  are  $Y$ -periodic in their second argument and, according to the boundary conditions (8)<sub>2</sub> and (9)<sub>2</sub>, such that  $\varphi^{(0)}(\mathbf{X}, \delta^{-1}\mathbf{X}) = \phi(\mathbf{X})$ ,  $\varphi^{(k)}(\mathbf{X}, \delta^{-1}\mathbf{X}) = 0$  for  $k \neq 0$ ,  $\mathbf{u}^{(0)}(\mathbf{X}, \delta^{-1}\mathbf{X}) = \mathbf{v}(\mathbf{X})$ , and  $\mathbf{u}^{(k)}(\mathbf{X}, \delta^{-1}\mathbf{X}) = \mathbf{0}$  for  $k \neq 0$  on  $\partial\Omega$ . In view of the one-way coupling of the boundary-value problems (8)–(9), we begin in section 2.1 by working out the limit for the electric potential  $\varphi^\delta(\mathbf{X})$  and subsequently make use of this result to then work out the limit for the displacement field  $\mathbf{u}^\delta(\mathbf{X})$  in section 2.2.

A few words about the presentation are in order. A number of the results that are obtained in section 2.1 are classical, yet we opt to include their presentation in order to preserve the continuity of the derivation and, more critically, to better be able to point to how the presence of space charges affects the homogenized equations. Similarly, some of the results that are obtained in section 2.2 have been previously obtained by Tian [37] (see also [38]) via the two-scale convergence method [1]. In addition to providing an alternative derivation for those, their inclusion in the presentation here preserves the continuity of the derivation and, more critically, aids in illustrating how the addition of space charges (*not* present in the work of Tian [37]) impacts the homogenized equations.

Before proceeding with the derivation *per se*, it is important to remark that while the method of two-scale asymptotic expansions typically yields the right homogenized equations (see, e.g., [6]), it is not a rigorous proof of the homogenization limit; this is because the two-scale ansatz, (13) for the problem of interest here, may possibly be incorrect beyond  $O(\delta)$  due, for instance, to boundary-layer effects in the vicinity of  $\partial\Omega$ ; see, e.g., [32, 3] and references therein. The rigorous proof that the homogenized equations derived here from the two-scale asymptotic expansion are indeed correct

turns out to be quite technical because of the quadratic term  $\text{Grad}\varphi^\delta(\mathbf{X}) \otimes \text{Grad}\varphi^\delta(\mathbf{X})$  in  $(9)_1$  for the balance of linear momentum. Such a rigorous proof will be presented elsewhere.

**2.1. The limit of the electric potential  $\varphi^\delta(\mathbf{X})$  as  $\delta \rightarrow 0$ .** Upon introducing the variables  $\mathbf{x} = \mathbf{X}$  and  $\mathbf{y} = \delta^{-1}\mathbf{X}$  and the operator

$$(14) \quad \begin{aligned} \mathbf{A}^\delta &= \delta^{-2}\mathbf{A}^{(1)} + \delta^{-1}\mathbf{A}^{(2)} + \delta^0\mathbf{A}^{(3)} \quad \text{with} \\ \mathbf{A}^{(1)} &= -\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial}{\partial y_j} \right], \quad \mathbf{A}^{(2)} = -\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial}{\partial x_j} \right] - \frac{\partial}{\partial x_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial}{\partial y_j} \right], \\ \mathbf{A}^{(3)} &= -\frac{\partial}{\partial x_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial}{\partial x_j} \right], \end{aligned}$$

where  $\frac{\partial}{\partial x_i}$  and  $\frac{\partial}{\partial y_i}$  denote partial derivatives with respect to  $\mathbf{x}$  and  $\mathbf{y}$ , we begin by recasting the PDE  $(8)_1$  for the electric potential  $\varphi^\delta(\mathbf{X})$  in the more convenient form

$$(15) \quad \mathbf{A}^\delta \varphi^\delta = q^\delta.$$

Substituting the ansatz  $(13)_1$  in the PDE (15) and expanding in powers of  $\delta$  leads to a hierarchy of equations of a very distinctive structure for the functions  $\varphi^{(k)}(\mathbf{x}, \mathbf{y})$ . The first three of these equations turn out to be enough for our purposes here, namely, to determine the first two terms  $\varphi^{(0)}(\mathbf{x}, \mathbf{y})$  and  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  in the expansion  $(13)_1$ . They are of  $O(\delta^{-2})$ ,  $O(\delta^{-1})$ ,  $O(\delta^0)$  and in terms of the operators (14) read as

$$(16) \quad \mathbf{A}^{(1)}\varphi^{(0)} = 0,$$

$$(17) \quad \mathbf{A}^{(1)}\varphi^{(1)} + \mathbf{A}^{(2)}\varphi^{(0)} = f_k(\mathbf{x})g_k(\mathbf{y}),$$

$$(18) \quad \mathbf{A}^{(1)}\varphi^{(2)} + \mathbf{A}^{(2)}\varphi^{(1)} + \mathbf{A}^{(3)}\varphi^{(0)} = -\frac{\partial f_k}{\partial x_i}(\mathbf{x}) \frac{\partial \psi_k}{\partial y_i}(\mathbf{y}).$$

**The equation of order  $\delta^{-2}$ .** The equation (16) of leading order is a PDE for the function  $\varphi^{(0)}(\mathbf{x}, \mathbf{y})$ , where  $\mathbf{y}$  is the independent variable and  $\mathbf{x}$  plays the role of a parameter. Its unique solution (with respect to  $\mathbf{y}$ ) is simply a function of  $\mathbf{x}$  that does not depend on  $\mathbf{y}$ . We write

$$(19) \quad \varphi^{(0)}(\mathbf{x}, \mathbf{y}) = \varphi(\mathbf{x}).$$

**The equation of order  $\delta^{-1}$ .** Making direct use of relation (19), the equation (17) of order  $\delta^{-1}$  reduces to

$$(20) \quad -\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial \varphi^{(1)}}{\partial y_j}(\mathbf{x}, \mathbf{y}) \right] = \frac{\partial \varepsilon_{ij}}{\partial y_i}(\mathbf{y}) \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) + f_k(\mathbf{x})g_k(\mathbf{y}), \quad \mathbf{y} \in Y,$$

which, for a given function  $\varphi(\mathbf{x})$  and a given  $\mathbf{x}$ , can be thought of as a PDE for the function  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  in the periodic unit cell  $Y$  with  $\mathbf{x}$  playing the role of a parameter. By introducing the  $Y$ -periodic functions  $\omega_i(\mathbf{y})$  and  $\varpi_i(\mathbf{y})$  defined implicitly as the unique solutions of the linear elliptic PDEs

$$(21) \quad \begin{cases} \frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial \omega_k}{\partial y_j}(\mathbf{y}) \right] = \frac{\partial \varepsilon_{ik}}{\partial y_i}(\mathbf{y}), \quad \mathbf{y} \in Y, \\ \int_Y \omega_k(\mathbf{y}) d\mathbf{y} = 0, \end{cases} \quad \begin{cases} \frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial \varpi_k}{\partial y_j}(\mathbf{y}) \right] = g_k(\mathbf{y}), \quad \mathbf{y} \in Y, \\ \int_Y \varpi_k(\mathbf{y}) d\mathbf{y} = 0, \end{cases}$$

the unique solution (with respect to  $\mathbf{y}$ ) of (20) can be written as

$$(22) \quad \varphi^{(1)}(\mathbf{x}, \mathbf{y}) = -\omega_k(\mathbf{y}) \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) - \varpi_k(\mathbf{y}) f_k(\mathbf{x}) + r^{(1)}(\mathbf{x}),$$

where  $r^{(1)}(\mathbf{x})$  is an arbitrary function of  $\mathbf{x}$ .

**The equation of order  $\delta^0$ .** Making again direct use of relation (19), the equation (18) of order  $\delta^0$  can be simplified to

$$(23) \quad -\frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial \varphi^{(2)}}{\partial y_j}(\mathbf{x}, \mathbf{y}) \right] = \frac{\partial}{\partial y_i} \left[ \varepsilon_{ij}(\mathbf{y}) \frac{\partial \varphi^{(1)}}{\partial x_j}(\mathbf{x}, \mathbf{y}) \right] \\ + \frac{\partial}{\partial x_i} \left[ \varepsilon_{ij}(\mathbf{y}) \left( \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) + \frac{\partial \varphi^{(1)}}{\partial y_j}(\mathbf{x}, \mathbf{y}) \right) \right] - \frac{\partial f_k}{\partial x_i}(\mathbf{x}) \frac{\partial \psi_k}{\partial y_i}(\mathbf{y}), \quad \mathbf{y} \in Y.$$

For a given function  $\varphi(\mathbf{x})$  and a given  $\mathbf{x}$  (since  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  is given by (22) in terms of  $\varphi(\mathbf{x})$ ), this equation can be thought of as a PDE for the function  $\varphi^{(2)}(\mathbf{x}, \mathbf{y})$  in the periodic unit cell  $Y$  with  $\mathbf{x}$  playing the role of a parameter.

Now, the PDE (23) admits a solution (with respect to  $\mathbf{y}$  and unique up to an additive constant) for  $\varphi^{(2)}(\mathbf{x}, \mathbf{y})$  if its right-hand side has zero average over  $Y$ ; this is the so-called Fredholm alternative. Consequently, after some manipulation employing the divergence theorem together with the  $Y$ -periodicity of  $\varepsilon(\mathbf{y})$  and  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$ , we require that

$$(24) \quad \frac{\partial}{\partial x_i} \int_Y \left[ \varepsilon_{ij}(\mathbf{y}) \left( \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) + \frac{\partial \varphi^{(1)}}{\partial y_j}(\mathbf{x}, \mathbf{y}) \right) - f_k(\mathbf{x}) \frac{\partial \psi_k}{\partial y_i}(\mathbf{y}) \right] d\mathbf{y} = 0.$$

Making use of the representation (22) for  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  in terms of the  $Y$ -periodic functions  $\omega_i(\mathbf{y})$  and  $\varpi_i(\mathbf{y})$ , applying the divergence theorem repeatedly, and exploiting the  $Y$ -periodicity of the PDEs (21), this equation can be simplified to

$$(25) \quad \frac{\partial}{\partial x_i} \left[ -\hat{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) \right] = \hat{q}(\mathbf{x}),$$

where

$$(26) \quad \hat{\varepsilon}_{ij} = \int_Y \varepsilon_{ik}(\mathbf{y}) \left( \delta_{jk} - \frac{\partial \omega_j}{\partial y_k}(\mathbf{y}) \right) d\mathbf{y} \quad \text{and} \quad \hat{q}(\mathbf{x}) = -\frac{\partial}{\partial x_i} [\hat{\alpha}_{ij} f_j(\mathbf{x})]$$

with

$$(27) \quad \hat{\alpha}_{ij} = \int_Y \left( \varepsilon_{ik}(\mathbf{y}) \frac{\partial \varpi_j}{\partial y_k}(\mathbf{y}) + y_i g_j(\mathbf{y}) \right) d\mathbf{y} = \int_Y (y_i - \omega_i(\mathbf{y})) g_j(\mathbf{y}) d\mathbf{y}.$$

Equation (25) is the homogenized PDE in  $\Omega$  that, together with the boundary condition  $\varphi(\mathbf{x}) = \phi(\mathbf{x})$  on  $\partial\Omega$ , completely determines the macroscopic electric potential  $\varphi(\mathbf{x})$ . The following remarks are in order:

i. *Physical interpretation of the homogenized equation (25) for  $\varphi(\mathbf{x})$ .* Equation (25), together with the boundary condition  $\varphi(\mathbf{x}) = \phi(\mathbf{x})$  on  $\partial\Omega$ , corresponds to the governing equation for the electrostatic field within a *homogeneous* dielectric medium, with constant effective permittivity tensor  $\hat{\varepsilon}$ , which contains a distribution of space charges characterized by the effective space-charge density  $\hat{q}(\mathbf{x})$ .

ii. *The effective permittivity tensor  $\hat{\varepsilon}$ .* The effective permittivity tensor (26)<sub>1</sub> that emerges in the homogenized equation (25) is independent of the choice of the domain  $\Omega$  occupied by the composite, the boundary conditions on  $\partial\Omega$ , and the presence of

space charges. Moreover, it follows from the properties (2) and (5)<sub>1</sub> of the local permittivity  $\epsilon(\mathbf{y})$  and the definition (21)<sub>1</sub> of the function  $\omega_i(\mathbf{y})$  that  $\widehat{\epsilon}$  satisfies the standard properties

$$(28) \quad \widehat{\epsilon}_{ij} = \widehat{\epsilon}_{ji}, \quad \widehat{\epsilon}_{ij}\xi_i\xi_j \geq \epsilon_0\xi_k\xi_k \quad \forall \xi \in \mathbb{R}^N, \quad \widehat{\epsilon}_{ij} \in \mathcal{L}^\infty(\Omega)$$

of a homogeneous dielectric medium; see, e.g., section 2.3 of Chapter 1 in [6].

iii. *The effective space-charge density  $\widehat{q}(\mathbf{x})$ .* The effective space-charge density (26)<sub>2</sub> that emerges in the homogenized equation (25) depends fundamentally on the presence of space charges through both of the constitutive functions  $\mathbf{f}(\mathbf{x})$  and  $\mathbf{g}(\mathbf{y})$  defining their density (10). It follows from the regularity (11)<sub>1</sub> of the function  $\mathbf{f}(\mathbf{x})$  and the definiteness of the integrals in (27) that

$$(29) \quad \widehat{q} \in H^1(\Omega).$$

It is also interesting to note that the total content of macroscopic space charges implied by the effective space-charge density (26)<sub>2</sub>,

$$(30) \quad \int_{\Omega} \widehat{q}(\mathbf{x})d\mathbf{x} = - \int_{\Omega} \widehat{\alpha}_{ij} \frac{\partial f_j}{\partial x_i}(\mathbf{x})d\mathbf{x},$$

need *not* be necessarily zero (only certain choices of the constitutive function  $\mathbf{f}(\mathbf{x})$  render macroscopic charge neutrality).

iv. *Mathematical well-posedness.* In view of the properties (28)<sub>2,3</sub> and (29) of  $\widehat{\epsilon}$  and  $\widehat{q}(\mathbf{x})$ , and of the smoothness of  $\partial\Omega$ , it follows from the Lax–Milgram theorem that the solution of the homogenized equation (25), supplemented by the boundary condition  $\varphi(\mathbf{x}) = \phi(\mathbf{x})$  on  $\partial\Omega$ , for the macroscopic electric potential  $\varphi(\mathbf{x})$  exists and is unique in  $H^1(\Omega)$ . The fact that the effective permittivity  $\widehat{\epsilon}$  is a constant together with the regularity  $\phi \in H^{3/2}(\partial\Omega)$  and the smoothness of the boundary  $\partial\Omega$  imply in fact the following stronger regularity result for  $\varphi(\mathbf{x})$ :

$$(31) \quad \varphi \in H^2(\Omega) \quad \text{and} \quad \text{Grad } \varphi \in H^1(\Omega; \mathbb{R}^N) \subset \mathcal{L}^4(\Omega; \mathbb{R}^N);$$

see, e.g., Chapter 8 in [12], Chapter 6.3 in [10], Theorem 8.3 in [23], and Theorem 9.16 in [7]. The higher regularity  $\varphi \in H^3(\Omega)$  can be obtained by considering boundary data  $\phi \in H^{5/2}(\partial\Omega)$ . We shall invoke this higher regularity in section 3.

v. *Computation of  $\widehat{\epsilon}$  and  $\widehat{q}(\mathbf{x})$ .* Evaluation of the formula (26)<sub>1</sub> for the effective permittivity tensor  $\widehat{\epsilon}$  requires knowledge of the  $Y$ -periodic function  $\omega_i \in H^1_{\#}(Y)$  defined by the PDE (21)<sub>1</sub>. In general, this PDE does not admit an analytical solution and hence must be solved numerically; being linear elliptic, the PDE (21)<sub>1</sub> can be readily solved, for instance, by the finite element method. Similarly, evaluation of the formula (26)<sub>2</sub> for the effective space-charge density  $\widehat{q}(\mathbf{x})$  requires knowledge of the  $Y$ -periodic function  $\varpi_i \in H^1_{\#}(Y)$  defined by the PDE (21)<sub>2</sub>. Given the alternative representation for  $\widehat{\alpha}_{ij}$  in the second equality of (27)<sub>2</sub>—which is a simple consequence of the divergence theorem and the  $Y$ -periodicity of the PDEs (21)—the effective space-charge density  $\widehat{q}(\mathbf{x})$  can also be obtained directly from knowledge of  $\omega_i(\mathbf{y})$ .

vi. *The correction function  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$ .* Having completely determined the function  $\varphi(\mathbf{x})$  in terms of equation (25) allows one to determine (up to an additive function of  $\mathbf{x}$ ) the correction function  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  in the expansion (13)<sub>1</sub> by virtue of relation (22). Knowledge of  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  allows one in turn to determine the leading-order term of the corresponding asymptotic expansion for the electric field  $\mathbf{E}^\delta(\mathbf{X})$  in the limit as  $\delta \rightarrow 0$ :



$$(32) \quad E_i^\delta(\mathbf{X}) = -\frac{\partial \varphi^\delta}{\partial X_i}(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k E_i^{(k)}(\mathbf{x}, \mathbf{y}) = -\left( \frac{\partial \varphi}{\partial x_i}(\mathbf{x}) + \frac{\partial \varphi^{(1)}}{\partial y_i}(\mathbf{x}, \mathbf{y}) \right) + O(\delta)$$

and, by the same token, the leading-order term of the expansion for the electric displacement field  $\mathbf{D}^\delta(\mathbf{X})$ :

$$(33) \quad D_i^\delta(\mathbf{X}) = \varepsilon_{ij}^\delta(\mathbf{X}) E_j^\delta(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k D_i^{(k)}(\mathbf{x}, \mathbf{y}) = \varepsilon_{ij}(\mathbf{y}) E_j^{(0)}(\mathbf{x}, \mathbf{y}) + O(\delta).$$

vii. *The macro-variables.* In addition to identifying  $\varphi(\mathbf{x})$  as the macro-variable for the electric potential in the homogenized equation (25), a quick glance at (25) suffices to recognize the macroscopic electric field

$$(34) \quad E_i(\mathbf{x}) \doteq -\frac{\partial \varphi}{\partial x_i}(\mathbf{x})$$

and the macroscopic electric displacement field

$$(35) \quad D_i(\mathbf{x}) \doteq -\widehat{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x})$$

as the corresponding macro-variables that complete the electrostatics characterization of the resulting effective dielectric medium.

The macro-variable (34) turns out to be identical to the one that arises in the classical context of dielectric composites *without* rapidly oscillating source terms (see, e.g., Chapter 2 in [6]). Namely, it corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (32) of the electric field  $\mathbf{E}^\delta(\mathbf{X})^2$ :

$$(36) \quad E_i(\mathbf{x}) = \int_Y E_i^{(0)}(\mathbf{x}, \mathbf{y}) d\mathbf{y}.$$

By the same token, the macro-variable (34) is consistent with the classical heuristic definition of macro-variables—in the absence of source terms—due to Hill [13, 14].

By contrast, the macro-variable (35) is *not* in accord with the classical result; instead relation (35) corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (33) of the electric displacement field  $\mathbf{D}^\delta(\mathbf{X})$  plus an additional contribution due to the presence of charges, specifically,

$$(37) \quad D_i(\mathbf{x}) = \int_Y D_i^{(0)}(\mathbf{x}, \mathbf{y}) d\mathbf{y} + \left( \int_Y \omega_i(\mathbf{y}) g_j(\mathbf{y}) d\mathbf{y} \right) f_j(\mathbf{x}).$$

viii. *An alternative set of macro-variables.* By exploiting the divergence form of the effective space-charge density (26)<sub>2</sub> and rewriting the homogenized equation (25) as

$$(38) \quad \frac{\partial}{\partial x_i} \left[ -\widehat{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) + \widehat{\alpha}_{ij} f_j(\mathbf{x}) \right] = 0,$$

one can alternatively define the same macroscopic electric field

$$(39) \quad E_i(\mathbf{x}) \doteq -\frac{\partial \varphi}{\partial x_i}(\mathbf{x})$$

<sup>2</sup>From a mathematical point of view, the macro-variable (34) corresponds to the weak  $\mathcal{L}^2$  limit of  $\mathbf{E}^\delta(\mathbf{X})$  as  $\delta \rightarrow 0$ .

as in remark vii above, but the different macro-variable

$$(40) \quad D_i(\mathbf{x}) \doteq -\widehat{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) + \widehat{\alpha}_{ij} f_j(\mathbf{x})$$

for the macroscopic electric displacement field instead of (35). Similar to the definition (35), the macro-variable (40) corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (33) of the electric displacement field  $\mathbf{D}^\delta(\mathbf{X})$  plus an additional contribution due to the presence of charges; in this case,

$$(41) \quad D_i(\mathbf{x}) = \int_Y D_i^{(0)}(\mathbf{x}, \mathbf{y}) d\mathbf{y} + \left( \int_Y y_i g_j(\mathbf{y}) d\mathbf{y} \right) f_j(\mathbf{x}).$$

Alternatively, this relation can be recast as a surface integral, namely,

$$(42) \quad D_i(\mathbf{x}) = \int_{\partial Y} y_i D_j^{(0)}(\mathbf{x}, \mathbf{y}) n_j d\mathbf{y}.$$

We conclude this remark by emphasizing that, in the alternative view (38) of the homogenized equation (25), the homogenized material is no longer a standard homogenous dielectric that contains a distribution of space charges, but rather some sort of source-free polarized dielectric with the term  $\widehat{\alpha}_{ij} f_j(\mathbf{x})$  playing the role of an initial polarization in the electric displacement field.

**2.2. The limit of the displacement field  $\mathbf{u}^\delta(\mathbf{X})$  as  $\delta \rightarrow 0$ .** Next, we turn to the asymptotic analysis for the displacement field  $\mathbf{u}^\delta(\mathbf{X})$ . Similar to the preceding asymptotic analysis for the electric potential field  $\varphi^\delta(\mathbf{X})$ , it proves helpful to introduce the operators

$$(43) \quad \begin{aligned} \mathbf{B}_{ik}^\delta &= \delta^{-2} \mathbf{B}_{ik}^{(1)} + \delta^{-1} \mathbf{B}_{ik}^{(2)} + \delta^0 \mathbf{B}_{ik}^{(3)} \quad \text{with} \\ \mathbf{B}_{ik}^{(1)} &= \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial}{\partial y_l} \right], \quad \mathbf{B}_{ik}^{(2)} = \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial}{\partial x_l} \right] + \frac{\partial}{\partial x_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial}{\partial y_l} \right], \\ \mathbf{B}_{ik}^{(3)} &= \frac{\partial}{\partial x_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial}{\partial x_l} \right] \end{aligned}$$

and

$$\mathbf{C}_i^\delta(h_1, h_2) = \delta^{-3} \mathbf{C}_i^{(0)}(h_1, h_2) + \delta^{-2} \mathbf{C}_i^{(1)}(h_1, h_2) + \delta^{-1} \mathbf{C}_i^{(2)}(h_1, h_2) + \delta^0 \mathbf{C}_i^{(3)}(h_1, h_2) \quad \text{with}$$

$$(44) \quad \begin{aligned} \mathbf{C}_i^{(0)}(h_1, h_2) &= -\frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial y_l} \right], \\ \mathbf{C}_i^{(1)}(h_1, h_2) &= -\frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \left( \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial x_l} + \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial y_l} \right) \right] - \frac{\partial}{\partial x_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial y_l} \right], \\ \mathbf{C}_i^{(2)}(h_1, h_2) &= -\frac{\partial}{\partial x_j} \left[ M_{ijkl}(\mathbf{y}) \left( \frac{\partial h_1}{\partial y_k} \frac{\partial h_2}{\partial x_l} + \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial y_l} \right) \right] - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial x_l} \right], \\ \mathbf{C}_i^{(3)}(h_1, h_2) &= -\frac{\partial}{\partial x_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial h_1}{\partial x_k} \frac{\partial h_2}{\partial x_l} \right] \end{aligned}$$

in order to recast the PDE (9)<sub>1</sub> for the displacement field  $\mathbf{u}^\delta(\mathbf{X})$  in the more convenient form

$$(45) \quad \mathbf{B}_{ik}^\delta u_k^\delta = \mathbf{C}_i^\delta(\varphi^\delta, \varphi^\delta).$$

Substituting the ansatz (13) in the PDE (45) and expanding in powers of  $\delta$  leads to a hierarchy of equations for the functions  $\mathbf{u}^{(k)}(\mathbf{x}, \mathbf{y})$ . Only the first four of these, of  $O(\delta^{-3})$ ,  $O(\delta^{-2})$ ,  $O(\delta^{-1})$ , and  $O(\delta^0)$ , turn out to be needed for our purposes here. In terms of the operators (43) and (44), they read as

$$(46) \quad 0 = \mathbf{C}_i^{(0)}(\varphi^{(0)}, \varphi^{(0)}),$$

$$(47) \quad \mathbf{B}_{ik}^{(1)} u_k^{(0)} = \mathbf{C}_i^{(1)}(\varphi^{(0)}, \varphi^{(0)}) + \mathbf{C}_i^{(0)}(\varphi^{(0)}, \varphi^{(1)}) + \mathbf{C}_i^{(0)}(\varphi^{(1)}, \varphi^{(0)}),$$

$$(48) \quad \mathbf{B}_{ik}^{(1)} u_k^{(1)} + \mathbf{B}_{ik}^{(2)} u_k^{(0)} = \mathbf{C}_i^{(2)}(\varphi^{(0)}, \varphi^{(0)}) + \mathbf{C}_i^{(1)}(\varphi^{(0)}, \varphi^{(1)}) + \mathbf{C}_i^{(1)}(\varphi^{(1)}, \varphi^{(0)}) \\ + \mathbf{C}_i^{(0)}(\varphi^{(0)}, \varphi^{(2)}) + \mathbf{C}_i^{(0)}(\varphi^{(2)}, \varphi^{(0)}) + \mathbf{C}_i^{(0)}(\varphi^{(1)}, \varphi^{(1)}),$$

$$(49) \quad \mathbf{B}_{ik}^{(1)} u_k^{(2)} + \mathbf{B}_{ik}^{(2)} u_k^{(1)} + \mathbf{B}_{ik}^{(3)} u_k^{(0)} = \mathbf{C}_i^{(3)}(\varphi^{(0)}, \varphi^{(0)}) + \mathbf{C}_i^{(2)}(\varphi^{(0)}, \varphi^{(1)}) + \mathbf{C}_i^{(2)}(\varphi^{(1)}, \varphi^{(0)}) \\ + \mathbf{C}_i^{(1)}(\varphi^{(0)}, \varphi^{(2)}) + \mathbf{C}_i^{(1)}(\varphi^{(2)}, \varphi^{(0)}) + \mathbf{C}_i^{(1)}(\varphi^{(1)}, \varphi^{(1)}) \\ + \mathbf{C}_i^{(0)}(\varphi^{(0)}, \varphi^{(3)}) + \mathbf{C}_i^{(0)}(\varphi^{(3)}, \varphi^{(0)}) + \mathbf{C}_i^{(0)}(\varphi^{(1)}, \varphi^{(2)}) \\ + \mathbf{C}_i^{(0)}(\varphi^{(2)}, \varphi^{(1)}).$$

In connection with these equations, we emphasize that the function  $\varphi^{(0)}(\mathbf{x}, \mathbf{y}) = \varphi(\mathbf{x})$  has been completely determined in the preceding subsection, while the function  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  has been partially determined (up to an additive function of  $\mathbf{x}$ ). On the other hand, the functions  $\varphi^{(2)}(\mathbf{x}, \mathbf{y})$  and  $\varphi^{(3)}(\mathbf{x}, \mathbf{y})$  were not solved for since the relevant hierarchical equations were not considered. In the sequel, it will become evident that, in spite of their appearance in (48) and (49), the functions  $\varphi^{(2)}(\mathbf{x}, \mathbf{y})$  and  $\varphi^{(3)}(\mathbf{x}, \mathbf{y})$  are actually not needed for our purposes here, namely, to work out the solution for the first two terms  $\mathbf{u}^{(0)}(\mathbf{x}, \mathbf{y})$  and  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$  in the expansion (13)<sub>2</sub>.

**The equation of order  $\delta^{-3}$ .** Granted the fact that the macroscopic electric potential  $\varphi^{(0)}(\mathbf{x}, \mathbf{y}) = \varphi(\mathbf{x})$  is independent of  $\mathbf{y}$ , the equation (46) of leading order is trivially satisfied.

**The equation of order  $\delta^{-2}$ .** By invoking again the independence of  $\varphi(\mathbf{x})$  on  $\mathbf{y}$ , the equation (47) of order  $\delta^{-2}$  reduces to a PDE for the function  $\mathbf{u}^{(0)}(\mathbf{x}, \mathbf{y})$  where  $\mathbf{y}$  is the independent variable and  $\mathbf{x}$  plays the role of a parameter. We write its unique solution (with respect to  $\mathbf{y}$ ) as

$$(50) \quad \mathbf{u}^{(0)}(\mathbf{x}, \mathbf{y}) = \mathbf{u}(\mathbf{x}).$$

**The equation of order  $\delta^{-1}$ .** Next, the equation (48) of order  $\delta^{-1}$  can be written as

$$(51) \quad \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial u_k^{(1)}}{\partial y_l}(\mathbf{x}, \mathbf{y}) \right] = - \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial u_k}{\partial x_l}(\mathbf{x}) \right] \\ - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \left( \frac{\partial \varphi^{(1)}}{\partial y_k}(\mathbf{x}, \mathbf{y}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) + \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi^{(1)}}{\partial y_l}(\mathbf{x}, \mathbf{y}) \right) \right] \\ - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) \right] - \frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial \varphi^{(1)}}{\partial y_k}(\mathbf{x}, \mathbf{y}) \frac{\partial \varphi^{(1)}}{\partial y_l}(\mathbf{x}, \mathbf{y}) \right], \quad \mathbf{y} \in Y.$$

For a given function  $\mathbf{u}(\mathbf{x})$  and a given  $\mathbf{x}$ , this equation is a PDE for the function  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$  in the periodic unit cell  $Y$  with  $\mathbf{x}$  playing the role of a parameter. With

help of the representation (22) for the function  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  and the introduction of the  $Y$ -periodic functions  $\chi_{ijk}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(1)}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(2)}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(3)}(\mathbf{y})$ , and  $\check{\chi}_{ijk}^{(4)}(\mathbf{y})$  defined implicitly as the unique solutions of the following linear elliptic PDEs for  $\mathbf{y} \in Y$ ,

$$(52) \quad \begin{cases} \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial \chi_{kpq}}{\partial y_l}(\mathbf{y}) \right] = -\frac{\partial L_{ijpq}}{\partial y_j}(\mathbf{y}), \\ \int_Y \chi_{kpq}(\mathbf{y}) d\mathbf{y} = 0, \\ \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial \check{\chi}_{kpq}^{(1)}}{\partial y_l}(\mathbf{y}) \right] = -\frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \left( \delta_{kp} - \frac{\partial \omega_p}{\partial y_k}(\mathbf{y}) \right) \left( \delta_{lq} - \frac{\partial \omega_q}{\partial y_l}(\mathbf{y}) \right) \right], \\ \int_Y \check{\chi}_{kpq}^{(1)}(\mathbf{y}) d\mathbf{y} = 0, \\ \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial \check{\chi}_{kpq}^{(2)}}{\partial y_l}(\mathbf{y}) \right] = \frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \left( \delta_{kp} - \frac{\partial \omega_p}{\partial y_k}(\mathbf{y}) \right) \frac{\partial \varpi_q}{\partial y_l}(\mathbf{y}) \right], \\ \int_Y \check{\chi}_{kpq}^{(2)}(\mathbf{y}) d\mathbf{y} = 0, \\ \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial \check{\chi}_{kpq}^{(3)}}{\partial y_l}(\mathbf{y}) \right] = \frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial \varpi_p}{\partial y_k}(\mathbf{y}) \left( \delta_{lq} - \frac{\partial \omega_q}{\partial y_l}(\mathbf{y}) \right) \right], \\ \int_Y \check{\chi}_{kpq}^{(3)}(\mathbf{y}) d\mathbf{y} = 0, \\ \frac{\partial}{\partial y_j} \left[ L_{ijkl}(\mathbf{y}) \frac{\partial \check{\chi}_{kpq}^{(4)}}{\partial y_l}(\mathbf{y}) \right] = -\frac{\partial}{\partial y_j} \left[ M_{ijkl}(\mathbf{y}) \frac{\partial \varpi_p}{\partial y_k}(\mathbf{y}) \frac{\partial \varpi_q}{\partial y_l}(\mathbf{y}) \right], \\ \int_Y \check{\chi}_{kpq}^{(4)}(\mathbf{y}) d\mathbf{y} = 0, \end{cases}$$

the unique solution (with respect to  $\mathbf{y}$ ) of (51) can be written in the form

$$(53) \quad u_i^{(1)}(\mathbf{x}, \mathbf{y}) = \chi_{ipq}(\mathbf{y}) \frac{\partial u_p}{\partial x_q}(\mathbf{x}) + \check{\chi}_{ipq}^{(1)}(\mathbf{y}) \frac{\partial \varphi}{\partial x_p}(\mathbf{x}) \frac{\partial \varphi}{\partial x_q}(\mathbf{x}) + \check{\chi}_{ipq}^{(2)}(\mathbf{y}) \frac{\partial \varphi}{\partial x_p}(\mathbf{x}) f_q(\mathbf{x}) \\ + \check{\chi}_{ipq}^{(3)}(\mathbf{y}) f_p(\mathbf{x}) \frac{\partial \varphi}{\partial x_q}(\mathbf{x}) + \check{\chi}_{ipq}^{(4)}(\mathbf{y}) f_p(\mathbf{x}) f_q(\mathbf{x}) + s_i^{(1)}(\mathbf{x}),$$

where  $s^{(1)}(\mathbf{x})$  is an arbitrary function of  $\mathbf{x}$ .

**The equation of order  $\delta^0$ .** For a given function  $\mathbf{u}(\mathbf{x})$  and a given  $\mathbf{x}$  (since  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$  is given by (53) in terms of  $\mathbf{u}(\mathbf{x})$ ), the equation (49) of order  $\delta^0$  can be thought of as a PDE for the function  $\mathbf{u}^{(2)}(\mathbf{x}, \mathbf{y})$  in the periodic unit cell  $Y$  with  $\mathbf{x}$  playing the role of a parameter. By invoking yet again the Fredholm alternative, such a PDE admits a solution (with respect to  $\mathbf{y}$  and unique up to an additive constant) for  $\mathbf{u}^{(2)}(\mathbf{x}, \mathbf{y})$  so long as the condition

$$(54) \quad \int_Y \frac{\partial}{\partial x_j} \left[ L_{ijkl}(\mathbf{y}) \left( \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \frac{\partial u_k^{(1)}}{\partial y_l}(\mathbf{x}, \mathbf{y}) \right) \right] d\mathbf{y} \\ + \int_Y \frac{\partial}{\partial x_j} \left[ M_{ijkl}(\mathbf{y}) \left( \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) + \frac{\partial \varphi^{(1)}}{\partial y_k}(\mathbf{x}, \mathbf{y}) \right) \left( \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) + \frac{\partial \varphi^{(1)}}{\partial y_l}(\mathbf{x}, \mathbf{y}) \right) \right] d\mathbf{y} = 0$$

is satisfied; in the derivation of this condition, use has been made of the divergence theorem together with the  $Y$ -periodicity of  $\mathbf{L}(\mathbf{y})$ ,  $\mathbf{M}(\mathbf{y})$ ,  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$ ,  $\varphi^{(2)}(\mathbf{x}, \mathbf{y})$ , and  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$ .

Making now use of the representation (22) for  $\varphi^{(1)}(\mathbf{x}, \mathbf{y})$  in terms of the  $Y$ -periodic functions  $\omega_i(\mathbf{y})$ ,  $\varpi_i(\mathbf{y})$ , the representation (53) for  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$  in terms of the  $Y$ -periodic functions  $\chi_{ijk}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(1)}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(2)}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(3)}(\mathbf{y})$ ,  $\check{\chi}_{ijk}^{(4)}(\mathbf{y})$ , and repeated use of the divergence theorem, equation (54) simplifies to

$$(55) \quad \frac{\partial}{\partial x_j} \left[ \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \widehat{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) \right] = - \widehat{b}_i(\mathbf{x}; \varphi(\mathbf{x})),$$

where

$$(56)$$

$$\tilde{L}_{ijkl} = \int_Y L_{ijpq}(\mathbf{y}) \left( \delta_{pk} \delta_{ql} + \frac{\partial \chi_{pkl}}{\partial y_q}(\mathbf{y}) \right) d\mathbf{y},$$

$$(57)$$

$$\begin{aligned} \widehat{M}_{ijkl} &= \int_Y \left\{ L_{ijpq}(\mathbf{y}) \frac{\partial \check{\chi}_{pkl}^{(1)}}{\partial y_q}(\mathbf{y}) + M_{ijpq}(\mathbf{y}) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(\mathbf{y}) \right) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(\mathbf{y}) \right) \right\} d\mathbf{y} \\ &= \int_Y M_{rspq}(\mathbf{y}) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(\mathbf{y}) \right) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(\mathbf{y}) \right) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(\mathbf{y}) \right) d\mathbf{y}, \end{aligned}$$

and

$$(58) \quad \widehat{b}_i(\mathbf{x}; \varphi(\mathbf{x})) = \frac{\partial}{\partial x_j} \left[ -\widehat{B}_{ijkl}^{(1)} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) f_l(\mathbf{x}) - \widehat{B}_{ijkl}^{(2)} f_k(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) + \widehat{B}_{ijkl}^{(3)} f_k(\mathbf{x}) f_l(\mathbf{x}) \right]$$

with

$$(59) \quad \widehat{B}_{ijkl}^{(1)} = - \int_Y \left\{ L_{ijpq}(\mathbf{y}) \frac{\partial \check{\chi}_{pkl}^{(2)}}{\partial y_q}(\mathbf{y}) - M_{ijpq}(\mathbf{y}) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(\mathbf{y}) \right) \frac{\partial \varpi_l}{\partial y_q}(\mathbf{y}) \right\} d\mathbf{y}$$

$$= \int_Y M_{rspq}(\mathbf{y}) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(\mathbf{y}) \right) \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(\mathbf{y}) \right) \frac{\partial \varpi_l}{\partial y_q}(\mathbf{y}) d\mathbf{y},$$

$$(60) \quad \widehat{B}_{ijkl}^{(2)} = - \int_Y \left\{ L_{ijpq}(\mathbf{y}) \frac{\partial \check{\chi}_{pkl}^{(3)}}{\partial y_q}(\mathbf{y}) - M_{ijpq}(\mathbf{y}) \frac{\partial \varpi_k}{\partial y_p}(\mathbf{y}) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(\mathbf{y}) \right) \right\} d\mathbf{y}$$

$$= \int_Y M_{rspq}(\mathbf{y}) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(\mathbf{y}) \right) \frac{\partial \varpi_k}{\partial y_p}(\mathbf{y}) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(\mathbf{y}) \right) d\mathbf{y},$$

$$(61) \quad \widehat{B}_{ijkl}^{(3)} = \int_Y \left\{ L_{ijpq}(\mathbf{y}) \frac{\partial \check{\chi}_{pkl}^{(4)}}{\partial y_q}(\mathbf{y}) + M_{ijpq}(\mathbf{y}) \frac{\partial \varpi_k}{\partial y_p}(\mathbf{y}) \frac{\partial \varpi_l}{\partial y_q}(\mathbf{y}) \right\} d\mathbf{y}$$

$$= \int_Y M_{rspq}(\mathbf{y}) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(\mathbf{y}) \right) \frac{\partial \varpi_k}{\partial y_p}(\mathbf{y}) \frac{\partial \varpi_l}{\partial y_q}(\mathbf{y}) d\mathbf{y}.$$

For a given macroscopic electric field  $\varphi(\mathbf{x})$  defined by the PDE (25) and a given boundary condition  $\mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x})$  on  $\partial\Omega$ , (55) is the homogenized PDE in  $\Omega$  that completely determines the macroscopic displacement field  $\mathbf{u}(\mathbf{x})$ . The following remarks are in order:

i. *Physical interpretation of the homogenized equation (55) for  $\mathbf{u}(\mathbf{x})$ .* Equation (55), together with the one-way coupled PDE (25) for the macroscopic electric field  $\varphi(\mathbf{x})$  and the boundary conditions  $\varphi(\mathbf{x}) = \phi(\mathbf{x})$  and  $\mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x})$  on  $\partial\Omega$ , constitutes the governing equation for the displacement field within a *homogeneous* elastic

dielectric medium, with constant effective elasticity tensor  $\tilde{\mathbf{L}}$  and constant effective electrostriction tensor  $\widehat{\mathbf{M}}$ , which contains a distribution of body forces characterized by the effective body-force density  $\widehat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x}))$ . Figure 2 provides a schematic of this physical interpretation of the homogenized equations (25) and (55).

ii. *The effective elasticity and electrostriction tensors  $\tilde{\mathbf{L}}$  and  $\widehat{\mathbf{M}}$ .* Much like the effective permittivity tensor  $\widehat{\epsilon}$  in the homogenized equation (25) for  $\varphi(\mathbf{x})$ , the effective elasticity tensor (56) and the effective electrostriction tensor (57) in the homogenized equation (55) for  $\mathbf{u}(\mathbf{x})$  are independent of the choice of the domain  $\Omega$  occupied by the composite, the boundary conditions on  $\partial\Omega$ , and the presence of space charges. Moreover, it follows from the properties (3)<sub>2</sub> and (5)<sub>2</sub> of the local elasticity tensor  $\mathbf{L}(\mathbf{y})$  and the definition (52)<sub>1</sub> of the function  $\chi_{ijk}(\mathbf{y})$  that  $\tilde{\mathbf{L}}$  satisfies the standard properties

$$(62) \quad \tilde{L}_{ijkl} = \tilde{L}_{klij} = \tilde{L}_{jikl} = \tilde{L}_{ijlk}, \quad \tilde{L}_{ijkl}\Xi_{ij}\Xi_{kl} > \theta\Xi_{pq}\Xi_{pq} \quad \forall \Xi \in \mathbb{R}^{N \times N}, \quad \tilde{L}_{ijkl} \in \mathcal{L}^\infty(\Omega),$$

for some positive constant  $\theta$ , of a homogeneous elastic dielectric medium; see, e.g., section 2.3 of Chapter 1 in [6] and Chapter 1.1.4 in [2]. Similarly, it follows from the properties (3)<sub>2</sub> and (5)<sub>2</sub> of the local elasticity tensor  $\mathbf{L}(\mathbf{y})$ , the property (5)<sub>3</sub> of the local electrostriction tensor  $\mathbf{M}(\mathbf{y})$ , and the definition (52)<sub>1</sub> of the function  $\chi_{ijk}(\mathbf{y})$  that  $\widehat{\mathbf{M}}$  also satisfies the standard properties

$$(63) \quad \widehat{M}_{ijkl} = \widehat{M}_{jikl} = \widehat{M}_{ijlk}, \quad \widehat{M}_{ijkl} \in \mathcal{L}^\infty(\Omega)$$

of a homogeneous elastic dielectric medium.

iii. *The effective body-force density  $\widehat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x}))$ .* In spite of the fact that there are no body forces in the original boundary-value problem (9) for  $\mathbf{u}^\delta(\mathbf{X})$ , body forces appear in the homogenized equation (55) for  $\mathbf{u}(\mathbf{x})$ . These emerge as a result of the presence of space charges in the *coupled* boundary-value problem (8) for  $\varphi^\delta(\mathbf{X})$ . In particular, the effective body-force density (58) that emerges in the homogenized equation (55) is independent of the choice of the domain  $\Omega$  occupied by the composite and the boundary conditions on  $\partial\Omega$ . Its dependence on the presence of space charges is through both constitutive functions  $\mathbf{f}(\mathbf{x})$  and  $\mathbf{g}(\mathbf{y})$  defining their density (10), as

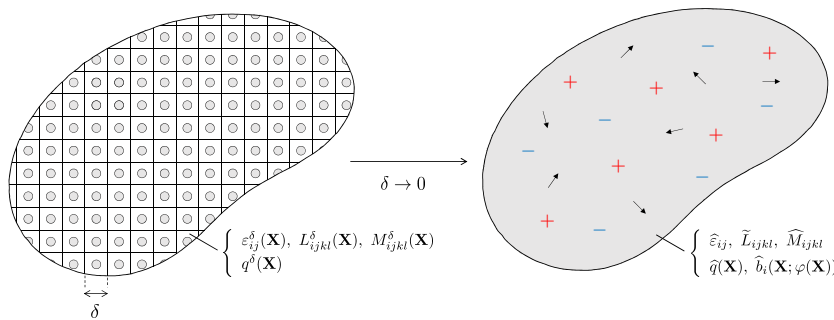


FIG. 2. Schematic illustrating that in the limit as  $\delta \rightarrow 0$  an elastic dielectric composite with heterogeneous properties  $\epsilon^\delta(\mathbf{X})$ ,  $\mathbf{L}^\delta(\mathbf{X})$ ,  $\mathbf{M}^\delta(\mathbf{X})$  containing a distribution of passive space charges—characterized by the density (10) with arbitrary but fixed functions  $\mathbf{f}(\mathbf{X})$  and  $\mathbf{g}(\delta^{-1}\mathbf{X})$ —reduces to an elastic dielectric with homogeneous effective properties  $\widehat{\epsilon}$ ,  $\tilde{\mathbf{L}}$ ,  $\widehat{\mathbf{M}}$ , effective space-charge density  $\widehat{q}(\mathbf{X})$ , and effective body-force density  $\widehat{\mathbf{b}}(\mathbf{X}; \varphi(\mathbf{X}))$ .

well as through the macroscopic electric potential  $\varphi(\mathbf{x})$ . Granted the boundedness (5)<sub>3</sub> of the local electrostriction tensor  $\mathbf{M}(\mathbf{y})$ , the regularity (11)<sub>1</sub> of the constitutive function  $\mathbf{f}(\mathbf{x})$ , the definitions (21) and (52)<sub>1</sub> of the functions  $\omega_i(\mathbf{y})$ ,  $\varpi_i(\mathbf{y})$ ,  $\chi_{ijk}(\mathbf{y})$ , and the fact that  $\text{Grad } \varphi \in H^1(\Omega; \mathbb{R}^N) \subset \mathcal{L}^4(\Omega; \mathbb{R}^N)$ , it follows that the effective body-force density (58) is of the divergence form

$$(64) \quad \widehat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x})) = \text{Div } \mathbf{B}(\mathbf{x}), \quad \mathbf{B} \in \mathcal{L}^2(\Omega; \mathbb{R}^{N \times N}).$$

iv. *Mathematical well-posedness.* Granted the properties (62)<sub>2,3</sub>, (63)<sub>2</sub>, (64) for  $\widetilde{\mathbf{L}}$ ,  $\widehat{\mathbf{M}}$ ,  $\widehat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x}))$ , the regularity result (31)<sub>2</sub> for  $\varphi(\mathbf{x})$ , the boundary condition  $\mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x}) \in H^{1/2}(\partial\Omega; \mathbb{R}^N)$  on  $\partial\Omega$ , and the smoothness of  $\partial\Omega$ , it follows from the Lax–Milgram theorem that the solution of the homogenized equation (55) for the macroscopic displacement field  $\mathbf{u}(\mathbf{x})$  exists, is unique, and

$$(65) \quad \mathbf{u} \in H^1(\Omega; \mathbb{R}^N).$$

v. *Computation of  $\widetilde{\mathbf{L}}$ ,  $\widehat{\mathbf{M}}$ , and  $\widehat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x}))$ .* Evaluation of the formulas (56) and (57) for the effective elasticity tensor  $\widetilde{\mathbf{L}}$  and the effective electrostriction tensor  $\widehat{\mathbf{M}}$  requires knowledge of the  $Y$ -periodic function  $\chi_{ipq} \in H_{\sharp}^1(Y)$  defined by the PDE (52)<sub>1</sub> and the  $Y$ -periodic function  $\omega_i \in H_{\sharp}^1(Y)$  defined by the PDE (21)<sub>1</sub>. These are linear elliptic PDEs that do not generally admit an analytical solution, but they can be readily solved numerically, for instance, by the finite element method. In addition to the solution of the homogenized equation (25) for the macroscopic electric potential  $\varphi(\mathbf{x})$ , evaluation of the formula (58) for the effective body-force density  $\widehat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x}))$  also requires the solutions for the functions  $\omega_i \in H_{\sharp}^1(Y)$  and  $\chi_{ijk} \in H_{\sharp}^1(Y)$ , as well as for the  $Y$ -periodic function  $\varpi_i \in H_{\sharp}^1(Y)$  defined by the linear elliptic PDE (21)<sub>2</sub>, whose gradients appear in the tensors  $\widehat{\mathbf{B}}^{(1)}$ ,  $\widehat{\mathbf{B}}^{(2)}$ , and  $\widehat{\mathbf{B}}^{(3)}$ .

vi. *The correction function  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$ .* By virtue of relation (53), having completely determined the function  $\varphi(\mathbf{x})$  from (25) and the function  $\mathbf{u}(\mathbf{x})$  from (55) allows one to determine (up to an additive function of  $\mathbf{x}$ ) the correction function  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$  in the expansion (13)<sub>2</sub>. Knowledge of  $\mathbf{u}^{(1)}(\mathbf{x}, \mathbf{y})$  allows one in turn to determine the leading-order term in the corresponding expansion for the gradient of the displacement field  $\mathbf{H}^\delta(\mathbf{X})$  in the limit as  $\delta \rightarrow 0$ :

$$(66) \quad H_{ij}^\delta(\mathbf{X}) = \frac{\partial u_i^\delta}{\partial X_j}(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k H_{ij}^{(k)}(\mathbf{x}, \mathbf{y}) = \frac{\partial u_i}{\partial x_j}(\mathbf{x}) + \frac{\partial u_i^{(1)}}{\partial y_j}(\mathbf{x}, \mathbf{y}) + O(\delta)$$

and, by the same token, the leading-order term of the expansion for the first Piola–Kirchhoff stress tensor  $\mathbf{S}^\delta(\mathbf{X})$ :

$$(67) \quad S_{ij}^\delta(\mathbf{X}) = L_{ijkl}^\delta(\mathbf{X}) H_{kl}^\delta(\mathbf{X}) + M_{ijkl}^\delta(\mathbf{X}) E_k^\delta(\mathbf{X}) E_l^\delta(\mathbf{X}) = \sum_{k=0}^{\infty} \delta^k S_{ij}^{(k)}(\mathbf{x}, \mathbf{y}) \\ = L_{ijkl}(\mathbf{y}) H_{kl}^{(0)}(\mathbf{x}, \mathbf{y}) + M_{ijkl}(\mathbf{y}) E_k^{(0)}(\mathbf{x}, \mathbf{y}) E_l^{(0)}(\mathbf{x}, \mathbf{y}) + O(\delta).$$

vii. *The macro-variables.* Similar to the identification of macro-variables in the homogenized equation (25) for the macroscopic electric potential  $\varphi(\mathbf{x})$ , a quick glance at (55) suffices to recognize not only  $\mathbf{u}(\mathbf{x})$  as the macro-variable for the displacement field, but also the macroscopic gradient of the displacement

$$(68) \quad H_{ij}(\mathbf{x}) \doteq \frac{\partial u_i}{\partial x_j}(\mathbf{x})$$

and the macroscopic stress

$$(69) \quad S_{ij}(\mathbf{x}) \doteq \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \widehat{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x})$$

as the corresponding macro-variables that complete the electroelastostatics characterization of the resulting effective elastic dielectric medium.

Akin to the standard macro-variable (34) that arises for the electric field, (68) is the standard macro-variable that emerges in the classical context of linear elasticity *without* rapidly oscillating source terms (see, e.g., Chapter 1 in [2]), in the sense that it corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (66) of the gradient of the displacement field  $\mathbf{H}^\delta(\mathbf{X})$ :

$$(70) \quad H_{ij}(\mathbf{x}) = \int_Y H_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y}.$$

The macro-variable (68) is then also in accord with the classical heuristic definition of macro-variables [13, 14].

By contrast, the macro-variable (69) differs from the classical result, since it corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (67) of the stress  $\mathbf{S}^\delta(\mathbf{X})$  plus additional contributions due to the presence of charges, specifically,

$$(71) \quad S_{ij}(\mathbf{x}) = \int_Y S_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y} + \widehat{B}_{ijkl}^{(1)} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) f_l(\mathbf{x}) + \widehat{B}_{ijkl}^{(2)} f_k(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) - \widehat{B}_{ijkl}^{(3)} f_k(\mathbf{x}) f_l(\mathbf{x}).$$

viii. *An alternative set of macro-variables.* By exploiting the divergence form of the effective body-force density (58) and rewriting the homogenized equation (55) as

$$(72) \quad \frac{\partial}{\partial x_j} \left[ \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \widehat{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) - \widehat{B}_{ijkl}^{(1)} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) f_l(\mathbf{x}) - \widehat{B}_{ijkl}^{(2)} f_k(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) + \widehat{B}_{ijkl}^{(3)} f_k(\mathbf{x}) f_l(\mathbf{x}) \right] = 0,$$

one can alternatively define the same macroscopic gradient of the displacement field

$$(73) \quad H_{ij}(\mathbf{x}) \doteq \frac{\partial u_i}{\partial x_j}(\mathbf{x})$$

as in remark vii above, but the different macro-variable

$$(74) \quad S_{ij}(\mathbf{x}) \doteq \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \widehat{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) - \widehat{B}_{ijkl}^{(1)} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) f_l(\mathbf{x}) - \widehat{B}_{ijkl}^{(2)} f_k(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) + \widehat{B}_{ijkl}^{(3)} f_k(\mathbf{x}) f_l(\mathbf{x})$$

for the macroscopic stress instead of (69). Contrary to the definition (69), the macro-variable (74) is consistent with the standard definition that emerges in the classical context of linear elasticity *without* rapidly oscillating source terms (see, e.g., Chapter 1 in [2]), in the sense that it corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (67) of the stress  $\mathbf{S}^\delta(\mathbf{X})$ :

$$(75) \quad S_{ij}(\mathbf{x}) = \int_Y S_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y}.$$



In regard to the above-identified alternative set of macro-variables, we emphasize that, in the alternative view (72) of the homogenized equation (55), the homogenized material is no longer a standard homogenous elastic dielectric with even electromechanical coupling that contains a distribution of space charges and body forces, but rather a source-free elastic dielectric with complicated electromechanical coupling which is neither even nor odd.

ix. *The absence of space charges.* In the absence of space charges when  $\mathbf{f}(\mathbf{x}) = \mathbf{0}$  and/or  $\mathbf{g}(\mathbf{y}) = \mathbf{0}$ , the effective space-charge density (26)<sub>2</sub> and the effective body-force density (58) vanish,

$$(76) \quad \tilde{q}(\mathbf{x}) = 0 \quad \text{and} \quad \hat{\mathbf{b}}(\mathbf{x}; \varphi(\mathbf{x})) = \mathbf{0},$$

and the homogenized equations (25) and (55), supplemented by the boundary conditions  $\varphi(\mathbf{x}) = \phi(\mathbf{x})$  and  $\mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x})$  on  $\partial\Omega$ , reduce to the homogenized equations originally obtained by Tian [37] and Tian et al. [38] via the two-scale convergence method [1].

**3. A class of active charges.** The preceding derivation of the homogenized equations (25) and (55) for the macroscopic electric field  $\varphi(\mathbf{x})$  and macroscopic displacement field  $\mathbf{u}(\mathbf{x})$  is valid for any choice—subject to the conditions (11)—of constitutive functions  $\mathbf{f}(\mathbf{x})$  and  $\mathbf{g}(\mathbf{y})$  defining the density of space charges (10) in the composite. These functions may be chosen not to be fixed or *passive* but to be *active* instead, in the sense that they may be selected to depend on  $\varphi^\delta(\mathbf{X})$  and/or on  $\mathbf{u}^\delta(\mathbf{X})$ . More generally, the functions  $\mathbf{f}(\mathbf{x})$  and  $\mathbf{g}(\mathbf{y})$  may be selected to have both passive as well as active components.

In this section, motivated by the work of Lopez-Pamies et al. [26], we work out the homogenized equations for elastic dielectric composites that contain a special class of active charges wherein the function  $\mathbf{g}(\mathbf{y})$  is arbitrary but fixed while the function  $\mathbf{f}(\mathbf{x})$  is set to be proportional to the macroscopic electric field:

$$(77) \quad f_i(\mathbf{x}) = -\frac{\partial\varphi}{\partial x_i}(\mathbf{x}).$$

From a mathematical point of view, we remark that this choice is valid provided that  $\varphi \in H^3(\Omega)$  since the function  $\mathbf{f}(\mathbf{x})$  was chosen from the outset to have the regularity (11)<sub>1</sub>. Accordingly, throughout this section, we shall assume that the boundary data  $\phi \in H^{5/2}(\partial\Omega)$ . As it will become clear further below, this will ensure that  $\varphi \in H^3(\Omega)$ ; cf. remark iv following the homogenized equation (25).

From a physical standpoint, roughly speaking, relation (77) corresponds to a microscopic distribution of space charges that scales in magnitude and aligns in direction with the electric field at the macroscopic material point  $\mathbf{x}$ . The precise details of the local alignment of the space charges are characterized by the specifics of the function  $\mathbf{g}(\mathbf{y})$ . At this point, it is important to emphasize that little is actually known about the constitutive behavior of active space charges in deformable solids from direct experimental measurements. Indeed, for the prominent case of dielectric elastomers filled with (semi)conducting or high-dielectric nanoparticles (see, e.g., [16, 28, 24]), space charges are expected to be active (i.e., locally mobile) in the regions of the elastomer immediately surrounding the nanoparticles [21, 30, 29], but direct measurements of the precise content and local mobility of the space charges contained therein have proved thus far difficult. The prescription (77) corresponds perhaps to the simplest physically plausible prototype that is consistent with the otherwise accessible macroscopic experimental measurements [25, 26, 20]. In this regard, it is also important to

remark that other classes of active space charges—such as those described by the local version  $\mathbf{f}(\mathbf{X}) = -\text{Grad } \varphi^\delta(\mathbf{X})$  of (77)—have been checked to lead to similar results to those that ensue from (77), and hence support the general physical implications presented here.

Granted the constitutive choice (77) for  $\mathbf{f}(\mathbf{x})$ , it is straightforward to deduce from (25) that the homogenized equation for the macroscopic electric field  $\varphi(\mathbf{x})$  is given by

$$(78) \quad \frac{\partial}{\partial x_i} \left[ -\tilde{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x}) \right] = 0$$

with

$$(79) \quad \tilde{\varepsilon}_{ij} = \hat{\varepsilon}_{ij} + \hat{\alpha}_{ij} = \int_Y \left\{ \varepsilon_{ik}(\mathbf{y}) \left( \delta_{jk} - \frac{\partial \omega_j}{\partial y_k}(\mathbf{y}) + \frac{\partial \varpi_j}{\partial y_k}(\mathbf{y}) \right) + y_i g_j(\mathbf{y}) \right\} d\mathbf{y}.$$

Similarly, it is straightforward to deduce from (55) that the homogenized equation for the macroscopic displacement field  $\mathbf{u}(\mathbf{x})$  is given by

$$(80) \quad \frac{\partial}{\partial x_j} \left[ \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \tilde{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}) \right] = 0,$$

where the macroscopic electric field  $\varphi(\mathbf{x})$  is defined implicitly by (78), it is recalled that  $\tilde{L}_{ijkl}$  is given by expression (56),

$$(81) \quad \begin{aligned} \tilde{M}_{ijkl} = & \hat{M}_{ijkl} + \hat{B}_{ijkl}^{(1)} + \hat{B}_{ijkl}^{(2)} + \hat{B}_{ijkl}^{(3)} = \int_Y M_{rspq}(\mathbf{y}) \left( \delta_{ri} \delta_{sj} + \frac{\partial \chi_{rij}}{\partial y_s}(\mathbf{y}) \right) \\ & \times \left( \delta_{pk} - \frac{\partial \omega_k}{\partial y_p}(\mathbf{y}) + \frac{\partial \varpi_k}{\partial y_p}(\mathbf{y}) \right) \left( \delta_{ql} - \frac{\partial \omega_l}{\partial y_q}(\mathbf{y}) + \frac{\partial \varpi_l}{\partial y_q}(\mathbf{y}) \right) d\mathbf{y}, \end{aligned}$$

and it is further recalled that  $\omega_i(\mathbf{y})$ ,  $\varpi_i(\mathbf{y})$ , and  $\chi_{ijk}(\mathbf{y})$  are the  $Y$ -periodic functions with zero average in  $Y$  defined by the PDEs (21) and (52)<sub>1</sub>.

Equations (78) and (80), together with the boundary conditions  $\varphi(\mathbf{x}) = \phi(\mathbf{x})$  and  $\mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x})$  on  $\partial\Omega$ , are the homogenized PDEs in  $\Omega$  for the macroscopic electric field  $\varphi(\mathbf{x})$  and macroscopic displacement field  $\mathbf{u}(\mathbf{x})$ . A number of remarks are in order:

i. *Physical interpretation of the homogenized equations (78) and (80) for  $\varphi(\mathbf{x})$  and  $\mathbf{u}(\mathbf{x})$ .* The one-way coupled system of PDEs (78) and (80) for the macroscopic electric field  $\varphi(\mathbf{x})$  and the macroscopic displacement field  $\mathbf{u}(\mathbf{x})$  constitute the governing equations for a *homogeneous* elastic dielectric medium, with constant effective permittivity tensor  $\tilde{\varepsilon}$ , constant effective elasticity tensor  $\tilde{\mathbf{L}}$ , and constant effective electrostriction tensor  $\tilde{\mathbf{M}}$ . Remarkably, in spite of the fact that the elastic dielectric composite contains a distribution of space charges at the length scale of the microstructure, neither an effective space-charge density nor an effective body-force density show up in the homogenized equations (78) and (80). Instead, the effect of the space charges shows up in the effective permittivity tensor  $\tilde{\varepsilon}$  and the effective electrostriction tensor  $\tilde{\mathbf{M}}$ ; this distinctive feature, which is in direct contrast to the result obtained for passive charges in the preceding section, is elaborated further in the next remarks. Figure 3 provides a schematic of the above-identified physical interpretation of the homogenized equations (78) and (80).

ii. *The effective permittivity, elasticity, and electrostriction tensors  $\tilde{\varepsilon}$ ,  $\tilde{\mathbf{L}}$ , and  $\tilde{\mathbf{M}}$ .* The effective elasticity tensor  $\tilde{\mathbf{L}}$  in the homogenized equation (80) is identical to the effective elasticity tensor in the homogenized equation (55) for the case of passive charges; its properties are outlined in remark ii of section 2.2. On the other hand,

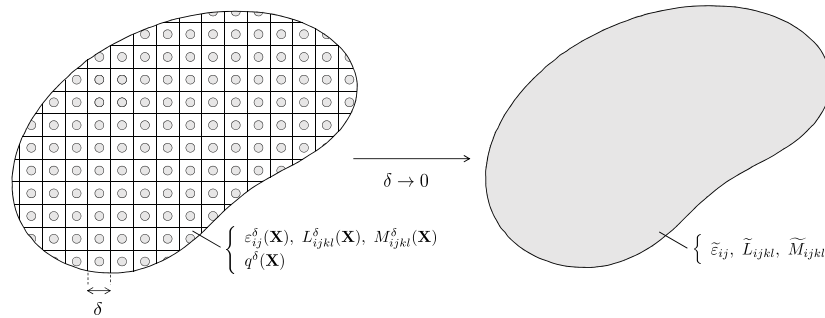


FIG. 3. Schematic illustrating that in the limit as  $\delta \rightarrow 0$  an elastic dielectric composite with heterogeneous properties  $\epsilon^{\delta}(\mathbf{X})$ ,  $\mathbf{L}^{\delta}(\mathbf{X})$ ,  $\mathbf{M}^{\delta}(\mathbf{X})$  containing a distribution of active space charges—characterized by the density (10) with the choice (77) for the function  $\mathbf{f}(\mathbf{X})$  and arbitrary but fixed function  $\mathbf{g}(\delta^{-1}\mathbf{X})$ —reduces to an elastic dielectric with homogeneous effective properties  $\tilde{\epsilon}$ ,  $\tilde{\mathbf{L}}$ ,  $\tilde{\mathbf{M}}$ .

the effective permittivity tensor (79) and the effective electrostriction tensor (81) that emerge in the homogenized equations (78) and (80) are different from their counterparts in (25) and (55). While they are independent of the choice of the domain  $\Omega$  occupied by the composite and the boundary conditions on  $\partial\Omega$ , the effective tensors (79) and (81) *do* depend strongly on the presence of space charges through the constitutive function  $\mathbf{g}(\mathbf{y})$ , which, as discussed in section 1, describes the distribution of space charges at the length scale of the microstructure.

More specifically, it follows from the regularity (5)<sub>1</sub> of the local permittivity tensor  $\epsilon(\mathbf{y})$ , the definitions (21) of the functions  $\omega_i(\mathbf{y})$  and  $\varpi_i(\mathbf{y})$ , and the boundedness (11)<sub>3</sub> of  $\mathbf{g}(\mathbf{y})$ , that the effective permittivity (79) is bounded,

$$(82) \quad \tilde{\epsilon}_{ij} \in \mathcal{L}^{\infty}(\Omega),$$

but, rather remarkably, it is *not* necessarily symmetric, *nor* positive definite for the cases when is symmetric; whether  $\tilde{\epsilon}$  is symmetric and, if so, positive definite, depends on the choice of constitutive function  $\mathbf{g}(\mathbf{y})$ . It further follows from the properties (4) and (5)<sub>3</sub> of the local electrostriction tensor  $\mathbf{M}(\mathbf{y})$ , together with the definitions (21) and (52)<sub>1</sub> of the functions  $\omega_i(\mathbf{y})$ ,  $\varpi_i(\mathbf{y})$ , and  $\chi_{ijk}(\mathbf{y})$ , that the effective electrostriction tensor (81) satisfies the standard properties

$$(83) \quad \tilde{M}_{ijkl} = \tilde{M}_{jikl} = \tilde{M}_{ijlk}, \quad \tilde{M}_{ijkl} \in \mathcal{L}^{\infty}(\Omega)$$

of a homogeneous elastic dielectric medium.

Here, it is important to recognize that, in spite of the boundedness (82) and (83)<sub>2</sub>, the components of the effective permittivity (79) and the effective electrostriction tensor (81) can be made to achieve arbitrarily large positive or negative values as, in essence, they are proportional to the constitutive function  $\mathbf{g}(\mathbf{y})$ . The physical implications of these features are far reaching. Indeed, these features confirm that judicious manipulation of space charges in deformable dielectric composites provides a promising path forward for the design of materials with exceptional electromechanical properties, including materials with unusually large permittivities and electrostriction coefficients and metamaterials featuring negative permittivity, cf. [34].

iii. *Mathematical well-posedness.* For choices of the constitutive function  $\mathbf{g}(\mathbf{y})$  that lead to effective permittivity tensors  $\tilde{\epsilon}$  that are symmetric and either positive or negative definite, the homogenized equation (78) for the macroscopic electric field

$\varphi(\mathbf{x})$  is nothing more than a standard second-order linear elliptic PDE with constant coefficient and hence, given the boundary condition  $\varphi(\mathbf{x}) = \phi(\mathbf{x}) \in H^{5/2}(\partial\Omega)$  on  $\partial\Omega$  and the smoothness of  $\partial\Omega$ , its solution exists, is unique, and possesses the following regularity properties:

$$(84) \quad \varphi \in H^3(\Omega) \quad \text{and} \quad \text{Grad } \varphi \in H^2(\Omega; \mathbb{R}^N) \subset \mathcal{L}^4(\Omega; \mathbb{R}^N).$$

In turn, granted the properties (62)<sub>2,3</sub> and (83)<sub>2</sub> of  $\tilde{\mathbf{L}}$  and  $\tilde{\mathbf{M}}$ , the regularity result (84)<sub>2</sub> for  $\varphi(\mathbf{x})$ , the boundary condition  $\mathbf{u}(\mathbf{x}) = \mathbf{v}(\mathbf{x}) \in H^{1/2}(\partial\Omega; \mathbb{R}^N)$  on  $\partial\Omega$ , and the smoothness of  $\partial\Omega$ , the Lax–Milgram theorem ensures existence and uniqueness of the solution of the homogenized equation (80) for the macroscopic displacement field  $\mathbf{u}(\mathbf{x})$ ; in particular,

$$(85) \quad \mathbf{u} \in H^1(\Omega; \mathbb{R}^N).$$

For choices of the constitutive function  $\mathbf{g}(\mathbf{y})$  that lead to effective permittivity tensors  $\tilde{\boldsymbol{\varepsilon}}$  that are not symmetric but satisfy the ellipticity condition  $\tilde{\varepsilon}_{ij}\xi_i\xi_j \geq \varepsilon_0\xi_k\xi_k \forall \boldsymbol{\xi} \in \mathbb{R}^N$ , solutions for the macroscopic fields  $\varphi(\mathbf{x})$  and  $\mathbf{u}(\mathbf{x})$  also exist, are unique, and possess the regularity (84) and (85). Finally, for choices of the constitutive function  $\mathbf{g}(\mathbf{y})$  that lead to effective permittivity tensors  $\tilde{\boldsymbol{\varepsilon}}$  that (are either symmetric or not symmetric but) do not satisfy the ellipticity condition  $\tilde{\varepsilon}_{ij}\xi_i\xi_j \geq \varepsilon_0\xi_k\xi_k \forall \boldsymbol{\xi} \in \mathbb{R}^N$ , the homogenized equation (78) is *not* elliptic and hence solutions for the macroscopic electric field  $\varphi(\mathbf{x})$  may *not* exist.

iv. *Computation of  $\tilde{\boldsymbol{\varepsilon}}$ ,  $\tilde{\mathbf{L}}$ , and  $\tilde{\mathbf{M}}$ .* Evaluation of the formulas (79) and (81) for the effective permittivity tensor  $\tilde{\boldsymbol{\varepsilon}}$  and effective electrostriction tensor  $\tilde{\mathbf{M}}$  requires knowledge of the  $Y$ -periodic functions  $\omega_i \in H^1_{\#}(Y)$ ,  $\varpi_i \in H^1_{\#}(Y)$  only through the linear combination  $\varpi_i(\mathbf{y}) - \omega_i(\mathbf{y})$ . Their evaluation amounts then to solving not the two boundary-value problems (21) for  $\omega_i(\mathbf{y})$  and  $\varpi_i(\mathbf{y})$ , but instead the single boundary-value problem

$$(86) \quad \begin{cases} \frac{\partial}{\partial y_i} \left[ \varepsilon_{ik}(\mathbf{y}) \left( \delta_{kj} + \frac{\partial \check{\omega}_j}{\partial y_k}(\mathbf{y}) \right) \right] = g_j(\mathbf{y}), \mathbf{y} \in Y & \text{for } \check{\omega}_i(\mathbf{y}) \doteq \varpi_i(\mathbf{y}) - \omega_i(\mathbf{y}). \\ \int_Y \check{\omega}_j(\mathbf{y}) d\mathbf{y} = 0 \end{cases}$$

Evaluation of the formula (56) for the effective elasticity tensor  $\tilde{\mathbf{L}}$  requires knowledge of the  $Y$ -periodic function  $\chi_{ijk} \in H^1_{\#}(Y)$  defined by the PDE (52)<sub>1</sub>. The two above-identified equations for  $\check{\omega}_i(\mathbf{y})$  and  $\chi_{ijk}(\mathbf{y})$  are linear elliptic PDEs that can be readily solved numerically, for instance, again, by the finite element method [35].

v. *The leading-order terms for the electric field  $\mathbf{E}^\delta(\mathbf{X})$ , the electric displacement field  $\mathbf{D}^\delta(\mathbf{X})$ , the gradient of the displacement field  $\mathbf{H}^\delta(\mathbf{X})$ , and the stress  $\mathbf{S}^\delta(\mathbf{X})$ .* Granted the constitutive choice (77) for the function  $\mathbf{f}(\mathbf{x})$ , it is a simple matter to deduce from (32) and (33) that the leading-order terms in the homogenization limit as  $\delta \rightarrow 0$  for the electric field  $\mathbf{E}^\delta(\mathbf{X})$  and the electric displacement field  $\mathbf{D}^\delta(\mathbf{X})$  specialize to

$$(87) \quad E_i^{(0)}(\mathbf{x}, \mathbf{y}) = - \left( \delta_{ik} - \frac{\partial \omega_k}{\partial y_i}(\mathbf{y}) + \frac{\partial \varpi_k}{\partial y_i}(\mathbf{y}) \right) \frac{\partial \varphi}{\partial x_k}(\mathbf{x})$$

and

$$(88) \quad D_i^{(0)}(\mathbf{x}, \mathbf{y}) = \varepsilon_{ij}(\mathbf{y}) E_j^{(0)}(\mathbf{x}, \mathbf{y}).$$

Similarly, with help of the notation  $\check{\chi}_{kpq}(\mathbf{y}) \doteq \check{\chi}_{kpq}^{(1)}(\mathbf{y}) - \check{\chi}_{kpq}^{(2)}(\mathbf{y}) - \check{\chi}_{kpq}^{(3)}(\mathbf{y}) + \check{\chi}_{kpq}^{(4)}(\mathbf{y})$ , it is a simple matter to deduce from (66) and (67) that the leading-order terms for the gradient of the displacement field  $\mathbf{H}^\delta(\mathbf{X})$  and the first Piola–Kirchhoff stress tensor  $\mathbf{S}^\delta(\mathbf{X})$  specialize to

$$(89) \quad H_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) = \frac{\partial u_i}{\partial x_j}(\mathbf{x}) + \frac{\partial \chi_{ipq}}{\partial y_j}(\mathbf{y}) \frac{\partial u_p}{\partial x_q}(\mathbf{x}) + \frac{\partial \check{\chi}_{ipq}}{\partial y_j}(\mathbf{y}) \frac{\partial \varphi}{\partial x_p}(\mathbf{x}) \frac{\partial \varphi}{\partial x_q}(\mathbf{x})$$

and

$$(90) \quad S_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) = L_{ijkl}(\mathbf{y}) H_{kl}^{(0)}(\mathbf{x}, \mathbf{y}) + M_{ijkl}(\mathbf{y}) E_k^{(0)}(\mathbf{x}, \mathbf{y}) E_l^{(0)}(\mathbf{x}, \mathbf{y}).$$

vi. *The macro-variables.* The macro-variables that emerge from the one-way coupled homogenized equations (78) and (80) for the macroscopic electric field and the macroscopic electric displacement field can be readily deduced to be given by

$$(91) \quad E_i(\mathbf{x}) \doteq -\frac{\partial \varphi}{\partial x_i}(\mathbf{x}) \quad \text{and} \quad D_i(\mathbf{x}) \doteq -\tilde{\varepsilon}_{ij} \frac{\partial \varphi}{\partial x_j}(\mathbf{x}),$$

while those that emerge for the macroscopic gradient of the displacement field and the macroscopic stress are given by

$$(92) \quad H_{ij}(\mathbf{x}) \doteq \frac{\partial u_i}{\partial x_j}(\mathbf{x}) \quad \text{and} \quad S_{ij}(\mathbf{x}) \doteq \tilde{L}_{ijkl} \frac{\partial u_k}{\partial x_l}(\mathbf{x}) + \tilde{M}_{ijkl} \frac{\partial \varphi}{\partial x_k}(\mathbf{x}) \frac{\partial \varphi}{\partial x_l}(\mathbf{x}).$$

The macro-variables (91)<sub>1</sub> and (92) for the electric field, the gradient of the deformation, and the stress agree with the classical definition,

$$(93) \quad E_i(\mathbf{x}) = \int_Y E_i^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y}, \quad H_{ij}(\mathbf{x}) = \int_Y H_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y}, \quad S_{ij}(\mathbf{x}) = \int_Y S_{ij}^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y},$$

while the macro-variable (91)<sub>2</sub> for the electric displacement field does *not*. Instead, relation (91)<sub>2</sub> corresponds to the average over the unit cell  $Y$  of the leading-order term in the asymptotic expansion (88) of the electric displacement field  $\mathbf{D}^\delta(\mathbf{X})$  plus an additional contribution due to the presence of charges, specifically,

$$(94) \quad D_i(\mathbf{x}) = \int_Y D_i^{(0)}(\mathbf{x}, \mathbf{y}) \, d\mathbf{y} - \left( \int_Y y_i g_j(\mathbf{y}) \, d\mathbf{y} \right) \frac{\partial \varphi}{\partial x_j}(\mathbf{x}).$$

As opposed to its counterpart (37) for the case of passive charges, relation (94) can be written as a surface integral:

$$(95) \quad D_i(\mathbf{x}) = \int_{\partial Y} y_i D_j^{(0)}(\mathbf{x}, \mathbf{y}) n_j \, d\mathbf{y}.$$

**4. Sample results.** The homogenized equations (25) and (55)—or, equivalently, (38) and (72)—provide a simple yet powerful tool to investigate the macroscopic elastic dielectric response of deformable dielectrics that, due to their fabrication process, contain from the outset a distribution of space charges in their “ground” state (i.e., in the absence of externally applied electric fields and mechanical forces). As mentioned during the setting of the problem, a prominent example of such a class of materials is electrets [18]. Similarly, the homogenized equations (78) and (80) provide a tool to investigate the macroscopic elastic dielectric response of deformable dielectrics that do not contain space charges in their ground state, but that, instead, develop an internal distribution of space charges when externally subjected to an electric field, for instance, by a charge injection process [21, 30]. Dielectric elastomers filled with (semi)conducting or high-dielectric nanoparticles have been recently identified

as a possible example of such a class of materials [21, 26, 20]. In this final section, with the compound purpose of demonstrating the use of the resulting homogenized equations and of illustrating the dominant effect that space charges can have on the macroscopic behavior of elastic dielectrics, we work out an example problem in  $N = 1$  space dimension (where all the pertinent calculations can be carried out analytically) with application to porous electrets [11, 5, 15].

**4.1. A porous electret with passive charges on the walls of the pores.**

We begin with the presentation of the results for the effective coefficients  $\widehat{\varepsilon} = \widehat{\varepsilon}_{11}$ ,  $\widehat{\alpha} = \widehat{\alpha}_{11}$ ,  $\widehat{\mathbf{L}} = \widehat{L}_{1111}$ ,  $\widehat{\mathbf{M}} = \widehat{M}_{1111}$ ,  $\widehat{\mathbf{B}}^{(1)} = \widehat{B}_{1111}^{(1)}$ ,  $\widehat{\mathbf{B}}^{(2)} = \widehat{B}_{1111}^{(2)}$ ,  $\widehat{\mathbf{B}}^{(3)} = \widehat{B}_{1111}^{(3)}$  in the homogenized equations (25), (55) and then turn to the corresponding macroscopic response under an externally applied uniaxial field of a porous electret made up of alternating layers of an elastic dielectric matrix and air-filled pores bonded through thin interphases that contain *passive* charges. From a physical point of view, these results are aimed at describing the enhanced piezoelectric-like response of porous electrets due to the presence of fixed charges on the walls of the enclosed pores.

*Microscopic description of the porous electret.* We take the matrix phase in the electret to be an ideal elastic dielectric with constant permittivity  $\varepsilon_m$  and elasticity modulus  $\mu_m$ . To account for their internal pressure [17, 27], we also take the air-filled pores to be ideal elastic dielectrics with constant permittivity  $\varepsilon_p$  and elasticity modulus  $\mu_p$  and, accordingly, write the sole component (recall that in this example  $N = 1$ ) of the local permittivity, elasticity, and electrostriction tensors (1) as the scalar functions

$$(96) \quad \varepsilon_{11}(y) = [1 - \theta_p(y)] \varepsilon_m + \theta_p(y) \varepsilon_p, \quad L_{1111}(y) = 2 [1 - \theta_p(y)] \mu_m + 2\theta_p(y) \mu_p, \\ M_{1111}(y) = \frac{1}{2} \varepsilon_{11}(y)$$

of the single space variable  $y$  along the Cartesian laboratory axis  $\mathbf{e}_1$  aligned with the unit cell  $Y = (0, 1)$ ; see Figure 4(a) for a schematic. In the above expressions,  $\theta_p(y)$  stands for the indicator function of the spatial regions occupied by the pores and is given by<sup>3</sup>

$$(97) \quad \theta_p(y) = \begin{cases} 1 & \text{if } \frac{1 - c_p}{2} < y < \frac{1 + c_p}{2}, \\ 0 & \text{otherwise} \end{cases}$$

with  $c_p = \int_Y \theta_p(y) dy$  denoting the volume fraction of pores in the electret. In addition, we model the distribution of passive charges through the choice of constant and piecewise constant constitutive functions

$$(98) \quad f_1(x) = 1 \text{ V/m} \quad \text{and} \quad g_1(y) = -[\theta_{i_1}(y) - \theta_{i_2}(y)]\beta$$

in (10). Here, the parameter  $\beta$  (of units F/m) corresponds to some measure of the charge content, while

$$(99) \quad \theta_{i_1}(y) = \begin{cases} 1 & \text{if } \frac{1 - c_p}{2} < y < \frac{1 - c_p + c_i}{2}, \\ 0 & \text{otherwise,} \end{cases} \quad \theta_{i_2}(y) = \begin{cases} 1 & \text{if } \frac{1 + c_p - c_i}{2} < y < \frac{1 + c_p}{2}, \\ 0 & \text{otherwise} \end{cases}$$

<sup>3</sup>Note that the piecewise constant permittivity (96)<sub>1</sub> does not fall within the realm of the regularity (5)<sub>1</sub> assumed at the outset. However, as already alluded to in section 1, the homogenization formulae worked out in sections 2 and 3 remain valid for the type of piecewise constant permittivity considered here.

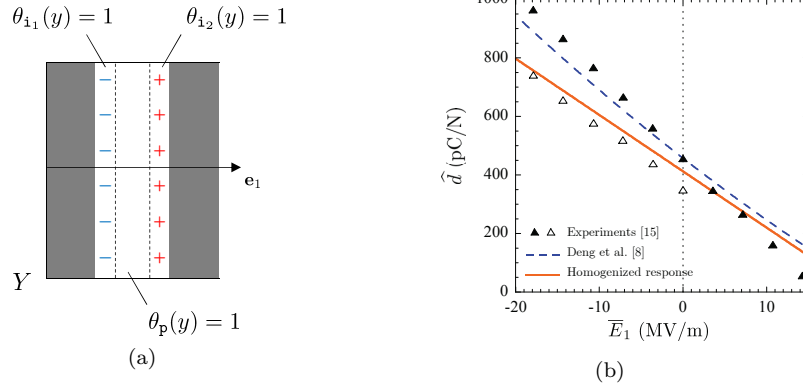


FIG. 4. (a) Schematic of the unit cell  $Y$  illustrating the layered distribution of air-filled pores and the surrounding interphases of space charges, as characterized by the indicator functions (97) and (99). (b) Comparison between the macroscopic “piezoelectric” coefficient  $\hat{d}$  (solid line) defined by (103), experimental data (triangles) of Hillenbrand and Sessler [15], and an earlier analytical result (dashed line) due to Deng et al. [8].

are the indicator functions of the two thin interphasial regions surrounding the pores where the charges are located; see Figure 4(a). In these last expressions,  $c_i = \int_Y \{\theta_{i_1}(y) + \theta_{i_2}(y)\} dy$  denotes the total volume fraction of the regions containing the charges.

The effective coefficients  $\hat{\varepsilon}_{11}$ ,  $\hat{\alpha}_{11}$ ,  $\tilde{L}_{1111}$ ,  $\widehat{M}_{1111}$ ,  $\widehat{B}_{1111}^{(1)}$ ,  $\widehat{B}_{1111}^{(2)}$ ,  $\widehat{B}_{1111}^{(3)}$ . Upon direct use of the local elastic dielectric properties (96) and constitutive functions (98) characterizing the distribution of charges, the ordinary differential equations that result from (21) and (52)<sub>1</sub> for the functions  $\omega_1(y)$ ,  $\varpi_1(y)$ , and  $\chi_{111}(y)$  can be readily solved in closed form. In turn, the integrals (26)<sub>1</sub>, (27), (56), (57), and (59)–(61) can be readily evaluated in closed form to render

$$(100) \quad \begin{aligned} \hat{\varepsilon}_{11} &= \frac{\varepsilon_m \varepsilon_p}{c_p \varepsilon_m + (1 - c_p) \varepsilon_p}, & \hat{\alpha}_{11} &= \frac{c_i (2c_p - c_i) \varepsilon_m \beta}{4[c_p \varepsilon_m + (1 - c_p) \varepsilon_p]}, & \tilde{L}_{1111} &= \frac{2\mu_m \mu_p}{c_p \mu_m + (1 - c_p) \mu_p}, \\ \widehat{M}_{1111} &= \frac{1}{4} \left( \frac{1 - c_p}{\varepsilon_m \mu_m} + \frac{c_p}{\varepsilon_p \mu_p} \right) \hat{\varepsilon}_{11}^2 \tilde{L}_{1111}, & \widehat{B}_{1111}^{(1)} &= \frac{(1 - c_p)(\mu_p - \mu_m)}{4\varepsilon_m \mu_m \mu_p} \hat{\varepsilon}_{11} \hat{\alpha}_{11} \tilde{L}_{1111}, \\ \widehat{B}_{1111}^{(2)} &= \widehat{B}_{1111}^{(1)}, & \widehat{B}_{1111}^{(3)} &= \left( \frac{1 - c_p}{4\varepsilon_m \mu_m} + \frac{(1 - c_p)^2 (3c_p - 2c_i) \varepsilon_p}{3(2c_p - c_i)^2 \varepsilon_m^2 \mu_p} \right. \\ & & & \left. + \frac{c_i (4c_p - 3c_i) [c_p \varepsilon_m + 2(1 - c_p) \varepsilon_p]}{12(2c_p - c_i)^2 \varepsilon_m \varepsilon_p \mu_p} \right) \hat{\alpha}_{11}^2 \tilde{L}_{1111}. \end{aligned}$$

For subsequent comparison with some experimental results of Hillenbrand and Sessler [15], we list in Table 1 the values taken by the effective coefficients (100) for the

TABLE 1  
Computed values of the seven effective coefficients  $\hat{\varepsilon}_{11}$ ,  $\hat{\alpha}_{11}$ ,  $\tilde{L}_{1111}$ ,  $\widehat{M}_{1111}$ ,  $\widehat{B}_{1111}^{(1)} = \widehat{B}_{1111}^{(2)}$ ,  $\widehat{B}_{1111}^{(3)}$  characterizing the macroscopic properties of the porous eletret with charges ( $\beta = 0.2$  F/m) on the walls of the enclosed pores.

$\hat{\varepsilon}_{11}$	$\hat{\alpha}_{11}$ (F/m)	$\tilde{L}_{1111}$ (MPa)	$\widehat{M}_{1111}$	$\widehat{B}_{1111}^{(1)}$ (F/m)	$\widehat{B}_{1111}^{(3)}$ (F/m)
$1.35 \varepsilon_0$	$7.35 \times 10^{-4}$	0.85	$0.91 \varepsilon_0$	$-1.72 \times 10^{-4}$	$4.04 \times 10^3$

choice of material parameters  $\epsilon_m = 2.35 \epsilon_0$ ,  $\mu_m = 1.0$  GPa,  $\epsilon_p = \epsilon_0$ ,  $\mu_p = 0.23$  MPa,  $\beta = 0.2$  F/m,  $c_p = 0.55$ , and  $c_i = 0.01$ ; recall that  $\epsilon_0 \approx 8.85 \times 10^{-12}$  F/m stands for the permittivity of vacuum. These values correspond to a polypropylene film with 55% porosity and overall Young’s modulus 0.84 MPa as in the experiments of Hillenbrand and Sessler [15].

*Macroscopic response of a thin film under a uniaxial electric field.* Now that the seven effective coefficients (100) have been determined, any boundary-value problem of interest may be investigated with help of the homogenized equations (25) and (55). Here, we consider a popular one in experiments wherein a thin film of thickness  $t$  made up of the porous electret is subjected to a uniaxial electric field across its thickness through the application of a voltage  $\Phi$ . In such a setup, neglecting fringe effects, the governing equations (25) and (55) are trivially satisfied and the macroscopic electric potential and macroscopic displacement field are given (up to an additive constant) by

$$(101) \quad \varphi(x) = -\bar{E}_1 x \quad \text{and} \quad u_1(x) = \bar{H}_{11} x$$

with

$$(102) \quad \bar{E}_1 = -\frac{\Phi}{t} \quad \text{and} \quad \bar{H}_{11} = -\frac{\widehat{M}_{1111}}{\widetilde{L}_{1111}} \bar{E}_1^2 - \frac{2\widehat{B}_{1111}^{(1)}}{\widetilde{L}_{1111}} \bar{E}_1 - \frac{\widehat{B}_{1111}^{(3)}}{\widetilde{L}_{1111}}.$$

A quantity of significant practical interest that can be readily extracted from the solution (101)–(102) is the macroscopic “piezoelectric” coefficient

$$(103) \quad \begin{aligned} \widehat{d} &\doteq \frac{\partial \bar{H}_{11}}{\partial \bar{E}_1} = -\frac{2\widehat{M}_{1111}}{\widetilde{L}_{1111}} \bar{E}_1 - \frac{2\widehat{B}_{1111}^{(1)}}{\widetilde{L}_{1111}} \\ &= -\frac{1}{2} \left( \frac{1 - c_p}{\epsilon_m \mu_m} + \frac{c_p}{\epsilon_p \mu_p} \right) \widehat{\epsilon}_{11}^2 \bar{E}_1 - \frac{(1 - c_p)(\mu_p - \mu_m)}{2\epsilon_m \mu_m \mu_p} \widehat{\epsilon}_{11} \widehat{\alpha}_{11}. \end{aligned}$$

For comparison with the experimental data (triangles) of Hillenbrand and Sessler [15] for a 71- $\mu\text{m}$ -thick polypropylene film with 55% porosity,<sup>4</sup> this coefficient is plotted (solid line) in Figure 4, as a function of the applied electric field  $\bar{E}_1$ , for the numerical values of the parameters listed in Table 1. For further comparisons, the earlier analytical result (dashed line) of Deng et al. [8] is also included in the figure.

**Appendix. Governing equations for elastic dielectric composites in the limit of small deformations and moderate electric fields in the presence of space charges.** Consider a deformable composite material with periodic microstructure of period  $\delta$  that occupies a bounded domain  $\Omega \subset \mathbb{R}^N$  ( $N = 1, 2, 3$ ), with smooth boundary  $\partial\Omega$  and closure  $\bar{\Omega} = \Omega \cup \partial\Omega$ , in its undeformed configuration. Material points are identified by their initial position vector  $\mathbf{X}$  in  $\Omega$  relative to some fixed point. Upon the application of mechanical and electrical stimuli, the position vector  $\mathbf{X}$  of a material point moves to a new position specified by  $\mathbf{x} = \mathbf{X} + \mathbf{u}^\delta(\mathbf{X})$ , where the displacement field  $\mathbf{u}^\delta(\mathbf{X})$  is loosely taken to possess sufficient regularity to warrant the mathematical well-posedness of the equations that follow. The associated deformation gradient is denoted by  $\mathbf{F}^\delta(\mathbf{X}) = \mathbf{I} + \text{Grad } \mathbf{u}^\delta(\mathbf{X})$ .

In the absence of magnetic fields, free currents, and body forces, and with no time dependence (see, e.g., Chapter 15 in [19]; see also [9, 25]), Maxwell’s and the momentum balance equations require that

<sup>4</sup>Note that the pores in the specimens of Hillenbrand and Sessler [15] were of oblate spheroidal shape, and *not* exactly layers as in the sample calculations presented here.



$$(104) \quad \text{Div } \mathbf{D}^\delta(\mathbf{X}) = q^\delta(\mathbf{X}), \quad \text{Curl } \mathbf{E}^\delta(\mathbf{X}) = 0, \quad \mathbf{X} \in \mathbb{R}^N$$

and

$$(105) \quad \text{Div } \mathbf{S}^\delta(\mathbf{X}) = 0, \quad \mathbf{S}^\delta \mathbf{F}^{\delta T} = \mathbf{F}^\delta \mathbf{S}^{\delta T}, \quad \mathbf{X} \in \Omega,$$

where  $\mathbf{D}^\delta(\mathbf{X})$ ,  $\mathbf{E}^\delta(\mathbf{X})$ ,  $\mathbf{S}^\delta(\mathbf{X})$  stand for the Lagrangian electric displacement field, the Lagrangian electric field, and the “total” first Piola–Kirchhoff stress tensor, while  $q^\delta(\mathbf{X})$  stands for the density of space charges per unit undeformed volume. For the specific case when the composite material is a (hyper)elastic dielectric with *even* electromechanical coupling, we further have the constitutive connections

$$(106) \quad \mathbf{D}^\delta(\mathbf{X}) = -\frac{\partial W^\delta}{\partial \mathbf{E}^\delta}(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta) \quad \text{and} \quad \mathbf{S}^\delta(\mathbf{X}) = \frac{\partial W^\delta}{\partial \mathbf{F}^\delta}(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta),$$

where the “total” free energy  $W^\delta(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta)$  is an objective function of the deformation gradient tensor  $\mathbf{F}^\delta$  and an even and objective function of the electric field  $\mathbf{E}^\delta$ , namely,  $W^\delta(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta) = W^\delta(\mathbf{X}, \mathbf{Q}\mathbf{F}^\delta, \mathbf{E}^\delta) = W^\delta(\mathbf{X}, \mathbf{F}^\delta, -\mathbf{E}^\delta)$  for all  $\mathbf{Q} \in \text{Orth}+$  and arbitrary  $\mathbf{X}$ ,  $\mathbf{F}^\delta$ , and  $\mathbf{E}^\delta$ .

Upon recognizing that the assumed objectivity of  $W^\delta(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta)$  implies the automatic satisfaction of the balance of angular momentum (105)<sub>2</sub> and that Faraday’s law (104)<sub>2</sub> is automatically satisfied by the introduction of an electric potential  $\varphi^\delta(\mathbf{X})$  such that  $\mathbf{E}^\delta(\mathbf{X}) = -\text{Grad } \varphi^\delta(\mathbf{X})$ , the equations governing the elastic dielectric response of the composite material reduce to the PDEs

$$(107) \quad \text{Div} \left[ -\frac{\partial W^\delta}{\partial \mathbf{E}^\delta}(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta) \right] = q^\delta(\mathbf{X}), \quad \mathbf{X} \in \mathbb{R}^N \quad \text{and} \quad \text{Div} \left[ \frac{\partial W^\delta}{\partial \mathbf{F}^\delta}(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta) \right] = \mathbf{0}, \quad \mathbf{X} \in \Omega.$$

*The classical limit of small deformations and moderate electric fields.* Now, let us define  $\zeta$  as a vanishingly small parameter and take the deformation measure  $\mathbf{H}^\delta(\mathbf{X}) = \mathbf{F}^\delta(\mathbf{X}) - \mathbf{I} = \text{Grad } \mathbf{u}^\delta(\mathbf{X})$  to be  $O(\zeta)$  and the electric field  $\mathbf{E}^\delta(\mathbf{X}) = -\text{Grad } \varphi^\delta(\mathbf{X})$  to be  $O(\zeta^{1/2})$ . Then, assuming that the composite material is stress free in the undeformed configuration  $\Omega$ , the asymptotic result

$$(108) \quad W^\delta(\mathbf{X}, \mathbf{F}^\delta, \mathbf{E}^\delta) = -\frac{1}{2} E_i^\delta \varepsilon_{ij}^\delta(\mathbf{X}) E_j^\delta + \frac{1}{2} H_{ij}^\delta L_{ijkl}^\delta(\mathbf{X}) H_{kl}^\delta \\ + H_{ij}^\delta M_{ijkl}^\delta(\mathbf{X}) E_k^\delta E_l^\delta - E_i^\delta E_j^\delta \mathcal{T}_{ijkl}^\delta(\mathbf{X}) E_k^\delta E_l^\delta + O(\zeta^3)$$

follows from a simple formal calculation (and the physically inconsequential choice that  $W^\delta(\mathbf{X}, \mathbf{I}, \mathbf{0}) = 0$ ). Here,  $\varepsilon_{ij}^\delta(\mathbf{X}) = -\partial^2 W^\delta(\mathbf{X}, \mathbf{I}, \mathbf{0}) / \partial E_i^\delta \partial E_j^\delta$  is the permittivity tensor,  $L_{ijkl}^\delta(\mathbf{X}) = \partial^2 W^\delta(\mathbf{X}, \mathbf{I}, \mathbf{0}) / \partial F_{ij}^\delta \partial F_{kl}^\delta$  is the elasticity tensor,  $M_{ijkl}^\delta(\mathbf{X}) = 1/2 \partial^3 W^\delta(\mathbf{X}, \mathbf{I}, \mathbf{0}) / \partial F_{ij}^\delta \partial E_k^\delta \partial E_l^\delta$  is the electrostriction tensor, and  $\mathcal{T}_{ijkl}^\delta(\mathbf{X}) = -1/24 \partial^4 W^\delta(\mathbf{X}, \mathbf{I}, \mathbf{0}) / \partial E_i^\delta \partial E_j^\delta \partial E_k^\delta \partial E_l^\delta$  is the permittivity tensor of second order. In turn, to leading order, the constitutive relations (106) reduce to (see also Chapter 2.25 in [36]; section 15 in [39]; Chapters 2 and 3 in [37]; [33, 35]):

$$(109) \quad D_i^\delta(\mathbf{X}) = \varepsilon_{ij}^\delta(\mathbf{X}) E_j^\delta + O(\zeta^{3/2}) \quad \text{and} \quad S_{ij}^\delta(\mathbf{X}) = L_{ijkl}^\delta(\mathbf{X}) H_{kl}^\delta + M_{ijkl}^\delta(\mathbf{X}) E_k^\delta E_l^\delta + O(\zeta^2).$$

By taking the space-charge density  $q^\delta(\mathbf{X})$  to be  $O(\zeta^{1/2})$ , and by restricting attention to Dirichlet boundary conditions, the one-way coupled boundary-value problems (8)–(9) in the main body of the text follow readily upon direct use of the asymptotic expressions (109) in the governing PDEs (107).

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