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A Theory of Flexoelectric Membranes and Effective Properties of Heterogeneous Membranes

Recent developments in flexoelectricity, especially in nanostructures, have lead to several interesting notions such as piezoelectric materials without using piezoelectric materials and enhanced energy harvesting at the nanoscale, among others. In the biological context also, membrane flexoelectricity has been hypothesized to play an important role, e.g., biological mechanotransduction and hearing mechanisms, among others. In this paper, we consider a heterogeneous flexoelectric membrane and derive the homogenized or renormalized flexoelectric, dielectric, and elastic response, therefore, relating the corresponding effective electromechanical properties to its microstructural details. Our work allows design of a microstructure to tailor flexoelectric response, and an illustrative example is given for biological membranes. [DOI: 10.1115/1.4023978]

1 Introduction

Piezoelectricity is perhaps the most widely known and exploited forms of electromechanical coupling. In a piezoelectric material, a uniform mechanical strain induces an electric field and vice versa. Piezoelectricity is preferentially used where precise control of mechanical motion is required, e.g., in scanning probe microscopes, and has now found wide applications: next-generation energy harvesters [1], artificial muscles [2], and sensors and actuators [3], among others. Crystallographic considerations restrict this technologically important property to noncentrosymmetric crystal systems [4] and indeed the latter is a necessary condition for a material to display piezoelectricity. However, a nonuniform strain field or presence of strain gradients can locally break inversion symmetry and induce polarization even in centrosymmetric crystals. This phenomenon is termed flexoelectrictiy [5–8], inspired by a similar effect in liquid crystals [9–11]. The reader is encouraged to refer to some recent review articles on flexoelectricity by Cross [12], Sharma et al. [13], and Tagantsev et al. [14].

Recently, flexoelectricity has attracted a fair amount of attention from both fundamental and applications points of view, leading to intensive experimental [12,15–22] and theoretical [13,23–31] activity in this topic. Lack of symmetry at surfaces and the capability to support large strain gradient in nanoscale structures enable unusual forms of piezoelectricity and flexoelectricity. For example: creating piezoelectric metamaterial from a nonpiezoelectric material has been investigated experimentally and computationally [28,32]. In fact, Chandratre and Sharma [33] recently showed that predicated on the phenomenon of flexoelectricity, graphene can be "coaxed" to behave like a piezoelectric material merely by creating holes of certain symmetry. The artificial piezoelectricity thus produced was found to be almost as strong as that of well-known piezoelectric substances such as quartz.

Several other works have appeared on elucidating flexoelectricity in two-dimensional structures [34]. Dumitrica et al. [31] and Kalinin and Meunier [30] showed that low dimensional systems such as graphene tend to exhibit electronic flexoelectricity, e.g., bending of nonpolar quantum systems leading to emergence of net dipole moments. Upon bending, redistribution of the electron gas in the normal direction results in the formation of a net dipole moment and, hence, flexoelectric coupling (Fig. 1). For large radii of curvatures and in the extreme case of closed seamless cylinder, the dipoles (formed) cancel out each other and the net polarization vanishes—which is why nonchiral (dielectric) carbon nanotubes have no dipole moment.

It is worthwhile to mention that investigating flexoelectricity effect in curved structures is also common in soft condensed materials such as liquid crystals and cellular membranes [35–40] pioneered by Meyer [9]. Synthetic and biological flexoelectric membranes are actuators that bend under the action of external electric fields, a phenomenon of interest to the development of emerging adaptive materials as well as biological mechanotransduction. Several works have explored biological implications of membrane flexoelectricity, e.g., mechanosensitivity, electromotility, and hearing systems [41–47].

Flexoelectricity in membranes is fundamentally different from three-dimensional materials (crystalline or otherwise). In this paper, we consider an important emerging problem that is unaddressed so far: What is the renormalized or effective flexoelectric, response of a heterogeneous two-dimensional structure? How do

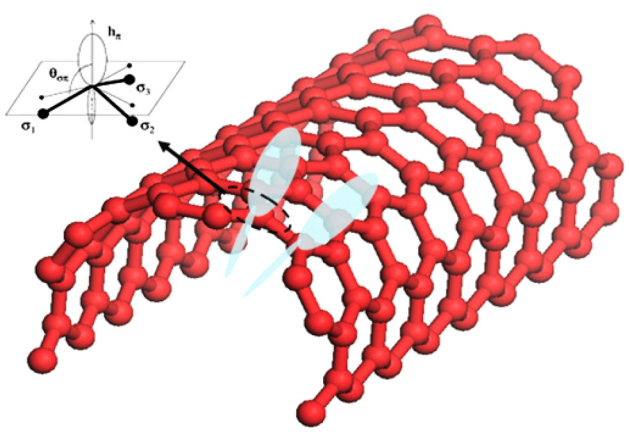

Fig. 1 Mechanism of flexoelectricity in 2D crystalline membranes such as graphene (adapted from Dumitrica et al. [31])

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Contributed by the Applied Mechanics Division of ASME for publication in the Journal of Applied Mechanics. Manuscript received January 5, 2013; final manuscript received February 14, 2013; published online August 22, 2013. Editor: Yonggang Huang.

the elastic and dielectric responses alter due to flexoelectricity? The answer to these questions may help interpret the behavior of complex biological membranes, in tailoring membranes such as graphene and boron-nitride sheets for various technological applications, and energy harvesting for stretchable electronics, among others. In Sec. 2, we present a simplified and linearized theory of flexoelectricity of two-dimensional deformable membranes. The homogenization problem is quite difficult even in the linear setting and, hence, we defer to future work the more complex case of nonlinear elastic membranes (see Ref. [48]). In Sec. 3, we present our homogenization approach and present specific results for heterogeneous membranes with circular holes. An illustrative example of our work is presented by considering a lipid bilayer membrane containing rigid protein inclusions.

Notation. In this paper we employ direct notation for brevity if possible. Tensors and vectors are denoted by bold symbols such as $\mathbf{L}$, $\boldsymbol{\xi}$, $\mathbf{M}$, etc., while scalars are denoted by lower case letters. When index notations are in use, the convention of summation over repeated index is followed. The inner (or dot) product between matrices $\mathbf{A}$ and $\mathbf{B}$ of the same size $m \times n$ is defined as $\mathbf{A} \cdot \mathbf{B} := \mathrm{Tr}(\mathbf{A}^T\mathbf{B}) = (\mathbf{A})_{ij}(\mathbf{B})_{ij}$, and the tensor product $\mathbf{A} \otimes \mathbf{B}$ is the fourth-order tensor such that for any $\mathbf{C} \in \mathbb{R}^{m\times n}$, $(\mathbf{A} \otimes \mathbf{B})\mathbf{C} = (\mathbf{B} \cdot \mathbf{C})\mathbf{A}$. For a second-order tensor field $\mathbf{M} : \mathbb{R}^2 \to \mathbb{R}^{2\times 2}$, in index form the operator $\nabla\nabla \cdot \mathbf{M}$ is equivalent to $(\mathbf{M})_{ij,ij}$.

2 A Theory for Flexoelectric Membranes

Let $\mathrm{U} \subset \mathbb{R}^2$ be an open bounded domain in xy-plane. Consider a thin dielectric membrane occupying $\mathrm{U} \times (-h/2, h/2) \subset \mathbb{R}^3$, where h is the thickness of the membrane. If the thickness $h \ll 1$ the thin membrane may be idealized as a two-dimensional body; the thermodynamic state is described by the out-of-plane displacement $w : \mathrm{U} \to \mathbb{R}$ and the out-of-plane polarization $P : \mathrm{U} \to \mathbb{R}$. The (linearized) curvature tensor of the membrane is defined as

$$\boldsymbol{\xi} = -\nabla\nabla w \tag{2.1}$$

The elasticity of the membrane will be modeled by the Helfrich–Canham curvature elasticity, which is in turn identical to the Kirchhoff–Love plate theory; the elastic contribution to the internal energy is given by

$$U_e[w] = \frac{1}{2}\int_{\mathrm{U}} \nabla\nabla w \cdot \mathbf{L}\nabla\nabla w \tag{2.2}$$

where $\mathbf{L} : \mathbb{R}^{2\times 2}_{\mathrm{sym}} \to \mathbb{R}^{2\times 2}_{\mathrm{sym}}$ is referred to as the "bending stiffness tensor." In particular, for isotropic materials,

$$\begin{aligned} &(\mathbf{L})_{piqj} = \mu_b(\delta_{pq}\delta_{ij} + \delta_{pj}\delta_{iq}) + \lambda_b\delta_{pq}\delta_{ij}, \quad \text{i.e.,} \\ &\frac{1}{2}\nabla\nabla w \cdot \mathbf{L}\nabla\nabla w = \mu_b|\nabla\nabla w|^2 + \frac{\lambda_b}{2}(\Delta w)^2 \end{aligned} \tag{2.3}$$

where μ_b, λ_b are the analogous Lamé constants for bending. In the biophysics community, the bending energy of a membrane is often written as

$$U_e[w] = \int_U \left[\frac{\kappa_b}{2}(\Delta w)^2 + \kappa_g \det(\nabla\nabla w)\right]$$

Comparing the above equation with Eq. (2.2) we find

$$\begin{cases} \mu_b = -\dfrac{\kappa_g}{2}, \\ \lambda_b = \kappa_b + \kappa_g, \end{cases} \quad \Leftrightarrow \quad \begin{cases} \kappa_b = 2\mu_b + \lambda_b, \\ \kappa_g = -2\mu_b \end{cases} \tag{2.4}$$

To model the flexoelectric effect, we postulate that the total internal energy of the isotropic membrane is given by

$$U[w, P] = \int_{\mathrm{U}} W(\nabla\nabla w, P) \tag{2.5}$$

where $W : \mathbb{R}^{2\times 2}_{\mathrm{sym}} \times \mathbb{R} \to \mathbb{R}$ is the total internal energy density function and given by a quadratic function

$$W(\boldsymbol{\xi}, P) = \frac{1}{2}\boldsymbol{\xi} \cdot \mathbf{L}\boldsymbol{\xi} + fP\mathrm{tr}\boldsymbol{\xi} + \frac{1}{2}aP^2 \tag{2.6}$$

Here, the tensor $\mathbf{L}$ and the constants f, a are material properties of the flexoelectric membrane and may in general depend on in-plane positions. In particular, the second term gives rise to the coupling between polarization and curvature (flexoelectric effects). We remark that the postulated internal energy density (2.6) may be validated from a well-grounded three-dimensional theory of flexoelectricity. Alternatively, we may regard the postulated form of internal energy density (2.6) for membranes as the linearized version of some more complete theory of flexoelectric membranes, including only the leading order terms for small bending and polarization. Such a postulation is equivalent to a constitutive law and necessary for completing a continuum theory. In this paper we do not attempt to rigorously justify either of the above viewpoints and merely take postulate (2.6) as our starting point. We note that the proposed energy functional has the basic ingredients of the flexoelectricity theory in three-dimensional materials: strain gradient (which in the present case is represented by curvature) leads to development of polarization and inhomogeneous electric fields will result in changes in curvature.

Let Γ_1 and Γ_2 be a disjoint subdivision of the boundary $\partial\mathrm{U}$ and $\Gamma_1 \neq \emptyset$. The following homogeneous boundary conditions may be applied on $\partial\mathrm{U}$:

(1) Clamped boundary conditions on Γ_1

$$w = 0, \quad \nabla w = 0 \quad \text{on} \quad \Gamma_1 \tag{2.7}$$

(2) Natural boundary conditions on Γ_2, i.e., there is no force or bending moment applied on Γ_2.

More general boundary conditions may also be considered, which will not be discussed below. Then under the application of an external electric field $E_z : \mathrm{U} \to \mathbb{R}$ and a mechanical body force $b_z : \mathrm{U} \to \mathbb{R}$, the total free energy of the membrane is given by

$$F[w, P] = \int_{\mathrm{U}} W(\nabla\nabla w, P) - \int_{\mathrm{U}} (PE_z + wb_z) \tag{2.8}$$

where the first integral is the internal energy of the flexoelectric membrane, and the second one is the potential energy arising from the interaction between the membrane and the external electric field and mechanical loading device. We remark that in the above formulation of free energy, we have neglected the nonlocal self-field energy associated with the electric field induced by polarization

$$\mathcal{E}^{\mathrm{self}}[w, P] = \frac{\epsilon_0}{2}\int_{\mathbb{R}^3} |\nabla\xi|^2, \quad \mathrm{div}[-\epsilon_0\nabla\xi + P\mathbf{e}_z\chi_B] = 0 \quad \text{in} \quad \mathbb{R}^3$$

where ϵ_0 is the electric permittivity of free space, $B = \mathrm{U} \times (-h/2, h/2)$ is the three-dimensional membrane body, and $\chi_B = 1$ in B ($=0$ otherwise). The above self-field energy $\mathcal{E}^{\mathrm{self}}[w, P]$ depends on the detailed geometry of U, favors depolarization state, and is difficult to handle. Nevertheless, we anticipate that the free energy (2.8) is sufficient for our purpose of modeling flexoelectric effects of membranes. One may also regard this nonlocal self-field energy is approximately included in the last term of Eq. (2.6) since both of them are quadratic, positive, and penalizing polarization.

In the equilibrium state, by the principle of minimum free energy the pair of (w, P) shall minimize the total free energy (2.8)

$$\min_{(w,P)\in\mathcal{S}} F[w,P] \tag{2.9}$$

where the admissible space for (w, P) is given by

$$\mathcal{S} := \left\{ (w,P) : \int_{\mathrm{U}} |\nabla\nabla w|^2 < +\infty, \int_{\mathrm{U}} |P|^2 < +\infty, w, \nabla w|_{\Gamma_1} = 0 \right\} \tag{2.10}$$

To find the Euler–Lagrange equations and boundary conditions associated with the above variational principle (2.19), we assume $(w,P) \in \mathcal{S}$ is the minimizer, and then for any perturbation $(w,P) \to (w,P) \pm \varepsilon(w_1, P_1)$ we have $(0 < \varepsilon \ll 1)$

$$F[w \pm \varepsilon w_1, P \pm \varepsilon P_1] \geq F[w,P] \tag{2.11}$$

which implies

$$\frac{d}{d\varepsilon} F[w + \varepsilon w_1, P + \varepsilon P_1]|_{\varepsilon=0} = 0 \tag{2.12}$$

Upon some tedious but standard calculations, the above equation implies

$$\begin{aligned} &\int_{\mathrm{U}} \{[\nabla\nabla\cdot(\mathbf{L}\nabla\nabla w) + \Delta(fP) - b_z]w_1 + [f\Delta w + aP - E_z]P_1\} \\ &+ \int_{\partial\mathrm{U}} \{\nabla w_1 \cdot [\mathbf{L}\nabla\nabla w + fP\mathbf{I}]\mathbf{n} - w_1 \mathbf{n}\cdot[\mathrm{div}(\mathbf{L}\nabla\nabla w) + \nabla(fP)]\} = 0 \end{aligned} \tag{2.13}$$

where $\mathbf{I} \in \mathbb{R}^{2\times 2}$ is the identity matrix and $\mathbf{n}$ is the unit outward normal on ∂U. Further, by the divergence theorem the last term on the left-hand side of Eq. (2.13) can be rewritten as ($\mathbf{t}$ is the tangential unit vector)

$$\begin{aligned} &\int_{\partial\mathrm{U}} \mathbf{n}\cdot(\mathbf{L}\nabla\nabla w + fP\mathbf{I})\mathbf{n}\partial_\nu w_1 \\ &\quad - \int_{\partial\mathrm{U}} w_1 \{\partial_\tau(\mathbf{t}\cdot(\mathbf{L}\nabla\nabla w + fP\mathbf{I})\mathbf{n}) + \mathbf{n}\cdot\mathrm{div}(\mathbf{L}\nabla\nabla w + fP\mathbf{I})\} \end{aligned} \tag{2.14}$$

where $(\partial_\nu, \partial_\tau) = (\mathbf{n}\cdot\nabla, \mathbf{t}\cdot\nabla)$ denote the normal and tangential derivatives on ∂U, respectively. Since (w_1, P_1) on U and $(\partial_\nu w_1, w_1)$ on ∂U can be chosen arbitrarily, Eqs. (2.13) and (2.14) imply the following Euler–Lagrange equations and boundary conditions associated with Eq. (2.9)

$$\begin{cases} \nabla\nabla\cdot(\mathbf{L}\nabla\nabla w) + \Delta(fP) - b_z = 0 & \text{on U}, \\ f\Delta w + aP - E_z = 0 & \text{on U}, \\ w = 0, \quad \nabla w = 0 & \text{on } \Gamma_1, \\ \mathbf{n}\cdot[\mathbf{L}\nabla\nabla w + fP\mathbf{I}]\mathbf{n} = 0, & \text{on } \Gamma_2, \\ \partial_\tau(\mathbf{t}\cdot(\mathbf{L}\nabla\nabla w + fP\mathbf{I})\mathbf{n}) + \mathbf{n}\cdot\mathrm{div}(\mathbf{L}\nabla\nabla w) + fP\mathbf{I}) = 0 & \text{on } \Gamma_2 \end{cases} \tag{2.15}$$

Using Eq. $(2.15)_2$ we eliminate P in Eq. $(2.15)_{1,4,5}$, and obtain

$$\begin{cases} \mathbf{M} := \tilde{\mathbf{L}}\nabla\nabla w, \quad \tilde{\mathbf{L}} := \mathbf{L} - \dfrac{f^2}{a}\mathbf{I}\otimes\mathbf{I} & \text{on U}, \\ \nabla\nabla\cdot\left(\mathbf{M} + \dfrac{f}{a}E_z\mathbf{I}\right) - b_z = 0 & \text{on U}, \\ w = 0, \quad \nabla w = 0 & \text{on } \Gamma_1, \\ \mathbf{n}\cdot\mathbf{M}\mathbf{n} = 0, \quad \partial_\tau(\mathbf{t}\cdot\mathbf{M}\mathbf{n}) + \mathbf{n}\cdot\mathrm{div}\mathbf{M} = 0 & \text{on } \Gamma_2 \end{cases} \tag{2.16}$$

We remark that the above Eq. (2.16) forms a standard elliptic boundary value problem for w; the existence, uniqueness, and stability has been thoroughly investigated; see, e.g., Evans [49].

In the absence of external electric field E_z and mechanical load b_z, a trivial solution to the above problem is clearly given by $(w, P) = (0, 0)$. The stability and uniqueness of this trivial state $(w, P) = (0, 0)$ requires that

$$\frac{1}{2}\boldsymbol{\xi}\cdot\mathbf{L}\boldsymbol{\xi} + f\,\mathrm{tr}(\boldsymbol{\xi})P + \frac{1}{2}aP^2 > 0 \quad \forall \text{ nonzero } \boldsymbol{\xi} \in \mathbb{R}^{2\times 2}_{\mathrm{sym}}\ P \in \mathbb{R} \tag{2.17}$$

For isotropic membranes with $\mathbf{L}$ specified by Eq. (2.3), the requirement (2.17) is equivalent to

$$\mu_b > 0, \quad \mu_b + \lambda_b > 0, \quad a > 0 \quad \& \quad \mu_b + \lambda_b > \frac{f^2}{a} \tag{2.18}$$

or in terms of κ_b, κ_g (cf. Eq. (2.4))

$$\kappa_b > 0, \quad -2\kappa_b < \kappa_g < 0, \quad a > 0 \quad \& \quad \kappa_b + \frac{\kappa_g}{2} > \frac{f^2}{a} \tag{2.19}$$

Moreover, it is worthwhile to present relations between the material constants in Eq. (2.6) and some more familiar quantities. First of all, if the self-field energy is neglected, from Toupin [50] we can show that

$$a = \frac{1}{\varepsilon_0 \chi_e h}$$

where ε_0 is the electric permittivity of the free space, χ_e is the electric susceptibility, and h is the thickness of the membrane. Including the self-field energy in the term $(1/2)aP^2$ would increase the constant a. Petrov [51], in the biological context, and Kalinin and Meunier [30], for a crystalline membrane, have shown that the net macroscopic electromechanical polarization developed in a bent membrane may be phenomenologically be considered to be linearly proportional to its curvature

$$P = -\gamma H \tag{2.20}$$

where $H = \mathrm{tr}(\boldsymbol{\xi})$ is (twice of) the mean curvature with the dimension of 1/m, P is the polarization per unit area with the dimension of C/m, and hence, γ is the flexoelectric constant and has the dimension of C, which is quantitatively different from the flexoelectric coefficient in our expressions. In our theory, for a homogenous membrane and in the absence of external electric field, Eq. $(2.15)_2$ implies that

$$P = -\frac{f}{a}\mathrm{tr}\boldsymbol{\xi} = -\frac{f}{a}H \tag{2.21}$$

Comparing with Eq. (2.20), we find

$$\gamma = \frac{f}{a} \tag{2.22}$$

Therefore, our definition of flexoelectric constant f has the dimension of Nm/C.

3 Homogenization of Heterogeneous Flexoelectric Membranes

We now consider a heterogeneous flexoelectric membrane with material properties are given by

$$\mathbf{L}^{(\varepsilon)}(\mathbf{x}) = \mathbf{L}_p\left(\frac{\mathbf{x}}{\varepsilon}\right), \quad f^{(\varepsilon)}(\mathbf{x}) = f_p\left(\frac{\mathbf{x}}{\varepsilon}\right), \quad a^{(\varepsilon)}(\mathbf{x}) = a_p\left(\frac{\mathbf{x}}{\varepsilon}\right) \tag{3.1}$$

where $\varepsilon \ll 1$ characterizes the length scale of the microscopic variations of the material properties, and without loss of generality, assume the material constants $\mathbf{L}_p : \mathbb{R}^2 \to \mathbb{R}^{2\times2\times2\times2}$ and $f_p, a_p : \mathbb{R}^2 \to \mathbb{R}$ are periodic functions with unit cell $Y = (0, 1)^2$. Since the material properties are highly oscillating, we anticipate the solutions to Eqs. (2.15) or (2.16), denoted by $(w^{(\varepsilon)}, P^{(\varepsilon)})$, are highly oscillating as well. To find the coarse-grained effective properties of the heterogeneous flexoelectric membrane, we employ the formal two-scale expansion method, assuming that

$$(w^{(\varepsilon)}(\mathbf{x}), P^{(\varepsilon)}(\mathbf{x})) = \sum_{k=0}^{\infty} \varepsilon^k (w_k(\mathbf{x}, \mathbf{y}), P_k(\mathbf{x}, \mathbf{y})) \tag{3.2}$$

where $\mathbf{y} = \mathbf{x}/\varepsilon$ is the "fast" variable for capturing the microscopic oscillations, (in contrast, $\mathbf{x}$-dependence characterizes macroscopic or coarse-grained behavior) $\mathbf{y} \mapsto (w_k(\mathbf{x}, \mathbf{y}), P_k(\mathbf{x}, \mathbf{y}))$ are periodic functions of period Y, and

$$\frac{1}{|Y|}\int_Y (w_k(\mathbf{x}, \mathbf{y}), P_k(\mathbf{x}, \mathbf{y}))d\mathbf{y} = (0, 0) \quad \forall k \geq 1 \tag{3.3}$$

From Eq. (3.2), direct calculations show that

$$\nabla\nabla w^{(\varepsilon)}(\mathbf{x}) = \sum_{k=0}^{\infty} \left[\frac{1}{\varepsilon^2} \nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_k(\mathbf{x}, \mathbf{y}) + \frac{2}{\varepsilon} \nabla_{\mathbf{y}}\nabla_{\mathbf{x}} w_k(\mathbf{x}, \mathbf{y}) + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}} w_k(\mathbf{x}, \mathbf{y}) \right] \tag{3.4}$$

where subscript $\mathbf{x}$ or $\mathbf{y}$ indicates that the derivatives are taken with respect to the first or the second variables of w_k. Collecting terms according to the orders of ε we rewrite Eq. (3.4) as

$$\begin{aligned} \nabla\nabla w^{(\varepsilon)}(\mathbf{x}) = {} & \frac{1}{\varepsilon^2} \nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_0(\mathbf{x}, \mathbf{y}) + \frac{1}{\varepsilon} [2\nabla_{\mathbf{y}}\nabla_{\mathbf{x}} w_0(\mathbf{x}, \mathbf{y}) \\ & + \nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_1(\mathbf{x}, \mathbf{y})] + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}} w_0(\mathbf{x}, \mathbf{y}) \\ & + 2\nabla_{\mathbf{y}}\nabla_{\mathbf{x}} w_1(\mathbf{x}, \mathbf{y}) + \nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_2(\mathbf{x}, \mathbf{y}) + o(1) \end{aligned} \tag{3.5}$$

For ease of notation, we introduce

$$\tilde{\mathbf{L}}^{(\varepsilon)} = \mathbf{L}^{(\varepsilon)} - \frac{(f^{(\varepsilon)})^2}{a^{(\varepsilon)}} \mathbf{I} \otimes \mathbf{I}, \quad \tilde{\mathbf{L}}_p = \mathbf{L}_p - \frac{f_p^2}{a_p} \mathbf{I} \otimes \mathbf{I} \tag{3.6}$$

Then, by Eq. $(2.16)_1$ we see that the original oscillating solution $w^{(\varepsilon)}$ satisfies

$$\nabla\nabla \cdot (\tilde{\mathbf{L}}^{(\varepsilon)} \nabla\nabla w^{(\varepsilon)}) + \Delta\left(\frac{f^{(\varepsilon)}}{a^{(\varepsilon)}} E_z\right) - b_z = 0 \quad \text{on U} \tag{3.7}$$

Inserting Eq. (3.2) into Eq. (3.7) and collecting terms associated with ε^k for $k \leq -2$, we obtain a cascade of equations since a Laurent series of ε vanishes on a neighborhood of origin if and only if all coefficients are zero. The leading order is ε^{-4} and implies

$$\nabla_{\mathbf{y}}\nabla_{\mathbf{y}} \cdot (\tilde{\mathbf{L}}_p(\mathbf{y}) \nabla\nabla w_0(\mathbf{x}, \mathbf{y})) = 0 \quad \forall (\mathbf{x}, \mathbf{y}) \in \mathrm{U} \times Y \tag{3.8}$$

The above equation concerns a function of $\mathbf{y}$-variables while $\mathbf{x}$ can be regarded as a parameter. By Eq. (2.17) and standard theory of elliptic equations, it can be shown that Eq. (3.8) admits a unique periodic solution within an additive constant. This implies that w_0 shall be independent of $\mathbf{y}$ and will be denoted by

$$w_0 = w_0(\mathbf{x}) \tag{3.9}$$

The next order is ε^{-3} and yields

$$\nabla_{\mathbf{y}}\nabla_{\mathbf{y}} \cdot (\tilde{\mathbf{L}}_p(\mathbf{y}) \nabla\nabla w_1(\mathbf{x}, \mathbf{y})) = 0 \quad \forall (\mathbf{x}, \mathbf{y}) \in \mathrm{U} \times Y \tag{3.10}$$

which, by similar argument and Eq. (3.3), implies

$$w_1 = w_1(\mathbf{x}) = 0 \tag{3.11}$$

Collecting terms associated with ε^{-2} we obtain

$$\begin{aligned} & \nabla_{\mathbf{y}}\nabla_{\mathbf{y}} \cdot [\tilde{\mathbf{L}}_p(\mathbf{y})(\nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_2(\mathbf{x}, \mathbf{y}) + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}} w_0(\mathbf{x}))] \\ & + \Delta_{\mathbf{y}}\left(\frac{f_p(\mathbf{y})}{a_p(\mathbf{y})} E_z\right) = 0 \quad \forall (\mathbf{x}, \mathbf{y}) \in \mathrm{U} \times Y \end{aligned} \tag{3.12}$$

One may continue the above calculations for higher order terms such as ε^{-1}, ε^0, etc., which, presumably, would enable us to identify the homogenized effective flexoelectric properties of the membrane and the equations governing the macroscopic quantities. However, these calculations would inevitably involve higher order terms such as $w_3(\mathbf{x}, \mathbf{y})$ and $w_4(\mathbf{x}, \mathbf{y})$ in the expansion (3.2) and, hence, be tedious and obsolete. It is, therefore, favorable to switch to the variational formulation (2.9). Inserting Eq. (3.2) into Eq. (2.8) and recalling the identity that for $\varepsilon \ll 1$ and smooth function $f(\mathbf{x}, \mathbf{y})$, which is periodic with unit cell Y with respect to variable $\mathbf{y}$,

$$\int_{\mathrm{U}} f\left(\mathbf{x}, \frac{\mathbf{x}}{\varepsilon}\right) dx = \int_{\mathrm{U}} \fint_Y f(\mathbf{x}, \mathbf{y}) d\mathbf{y} d\mathbf{x} + o(1) \tag{3.13}$$

where $\fint_V = 1/\mathrm{volume}(V) \int_V$ denote the average value of the integrand over the domain V. By Eqs. (3.5), (3.9), and (3.11) we find that in terms of w_0, w_2 and P_0, the total free energy is given by

$$F[w^{(\varepsilon)}, P^{(\varepsilon)}] = F^{2S}[w_0, \bar{P}_0; w_2, P_0'] + o(1) \tag{3.14}$$

where $(\bar{P}_0(\mathrm{x}) = \fint_Y P_0(\mathbf{x}, \mathbf{y}) d\mathbf{y}$ and $P_0'(\mathrm{x}, \mathrm{y}) = P_0(\mathbf{x}, \mathbf{y}) - \bar{P}_0(\mathbf{x}))$

$$\begin{aligned} F^{2S}[w_0, \bar{P}_0; w_2, P_0'] = {} & \int_{\mathrm{U}} \fint_Y \left\{ \frac{1}{2} (\nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_2 + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}} w_0) \right. \\ & \cdot \mathbf{L}_p (\nabla_{\mathbf{y}}\nabla_{\mathbf{y}} w_2 + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}} w_0) + f_p (P_0' + \bar{P}_0) \\ & \left. \times (\Delta_{\mathbf{y}} w_2 + \Delta_{\mathbf{x}} w_0) + \frac{a_p}{2} |P_0' + \bar{P}_0|^2 \right\} \\ & - \int_{\mathrm{U}} (\bar{P}_0 E_z + w_0 b_z) \end{aligned} \tag{3.15}$$

Here, the leading order free energy functional is expressed in terms of both macroscopic ($\mathbf{x}$ only-dependent) state $(w_0, \bar{P}_0)$ and microscopic ($\mathbf{y}$-dependent) state $(w_2, \bar{P}_0')$. Neglecting the higher order terms in Eq. (3.14), from the variational principle (2.9) we obtain the two-scale variational problem

$$\min_{(w_0, \bar{P}_0) \in \mathcal{S}} \min_{(w_2, P_0')} F^{2S}[w_0, \bar{P}_0; w_2, P_0'] \tag{3.16}$$

The above variational problem determines both the macroscopic state $(w_0, \bar{P}_0)$ and microscopic state $(w_2, \bar{P}_0')$ of the heterogeneous flexoelectric membrane. We remark that by definition (3.3) the microscopic state (w_2, P_0') shall satisfy that, in addition to the periodicity, integrability, and differentiability conditions,

$$\int_Y w_2(\mathbf{x}, \mathbf{y}) d\mathbf{y} = 0, \quad \int_Y P_0'(\mathbf{x}, \mathbf{y}) d\mathbf{y} = 0 \quad \forall \mathbf{x} \in \mathrm{U}$$

Focusing on the inner minimization problem in Eq. (3.16) first, we introduce the concept of *effective internal energy density*

$W^e : \mathbb{R}^{2\times2}_{\text{sym}} \times \mathbb{R} \to \mathbb{R}$ as a quadratic form such that for any $(\boldsymbol{\xi}, P) \in \mathbb{R}^{2\times2}_{\text{sym}} \times \mathbb{R}$,

$$W^e(\boldsymbol{\xi}, P) = \min_{(w',P')\in\mathcal{S}'} F_p[w', P'; \boldsymbol{\xi}, P] =: \frac{1}{2}\boldsymbol{\xi}\cdot\mathbf{L}^e\boldsymbol{\xi} + \mathbf{F}^e\cdot\boldsymbol{\xi}P + \frac{1}{2}a^eP^2 \tag{3.17}$$

where

$$F_p[w', P'; \xi, P] := \int_Y \left[\frac{1}{2}\left(\nabla_y\nabla_y w' + \boldsymbol{\xi}\right)\cdot\mathbf{L}_p\left(\nabla_y\nabla_y w' + \boldsymbol{\xi}\right) + f_p(P'+P)(\Delta_y w' + \text{tr}(\boldsymbol{\xi})) + \frac{a_p}{2}|P'+P|^2\right],$$

$$\mathcal{S}' := \left\{(w', P') : \int_Y w' = 0, \int_Y P' = 0, \text{ periodic with period } Y\right\} \tag{3.18}$$

Then it is straightforward to show that the outer minimization problem in Eq. (3.16) is equivalent to

$$\min_{(w_0,\bar{P}_0)\in\mathcal{S}} F^e[w_0, \bar{P}_0],$$
$$F^e[w_0, \bar{P}_0] := \int_U W^e(\nabla\nabla w_0, P_0) - \int_U (\bar{P}_0 E_z + w_0 b_z)$$

Comparing the above variational problem with the original variational problem (2.9) and (2.8), we henceforth justify our definition of effective energy density and associated effective material properties in Eq. (3.17).

We remark that Eq. (3.17) is reminiscent of the classic variational definitions of the effective properties of, e.g., elastic composites or conductive composites, and it completely determines the effective material properties $\mathbf{L}^e, \mathbf{F}^e$ and a^e. To explicitly solve for the minimizer (w', P') of the variational problem in Eq. (3.17), it is useful to find the associated Euler–Lagrange equation. By argument in analogy with Eqs. (2.11)–(2.13) and restricted to any subdomain $D \subset Y$ on which (w', P') are smooth, we have that for any $(w'_1, P'_1) \in \mathcal{S}'$,

$$\int_D \{[\nabla\cdot[\mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi})] + \Delta[f_p(P'+P)]]w'_1 + [f_p(\Delta w' + \text{tr}\boldsymbol{\xi}) + a_p(P'+P)]P'_1\} + \int_{\partial D}\nabla w'_1\cdot[\mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi}) + f_p(P'+P)\mathbf{I}]\mathbf{n} - \int_{\partial D} w'_1\mathbf{n}\cdot[\text{div}[\mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi})] + \nabla[f_p(P'+P)]] = 0 \tag{3.19}$$

where $\mathbf{n}$ is the unit outward normal on ∂D. Here and subsequently, we drop the subscript $_y$ associated with differential operators without the danger of confusion. Further, the last two terms in Eq. (3.19) can be rewritten as ($\mathbf{t}$ is the tangential unit vector)

$$\int_{\partial D}\partial_\nu w'_1\mathbf{n}\cdot\mathbf{M}\mathbf{n} - \int_{\partial D} w'_1\{\mathbf{n}\cdot\text{div}\mathbf{M} + \partial_\tau(\mathbf{t}\cdot\mathbf{M}\mathbf{n})\} \tag{3.20}$$

where

$$\mathbf{M} = \mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi}) + f_p(P'+P)\mathbf{I} \tag{3.21}$$

is introduced for brevity. Physically we may identify the above tensor as the "bending moment tensor."

If $D = Y$ (or in weak sense), Eq. (3.19) holds for any $(w'_1, P'_1) \in \mathcal{S}'$ only if the following Euler–Lagrange equations holds:

$$\begin{cases}\nabla\nabla\cdot\mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi}) + \Delta[f_p(P'+P)] = \lambda' & \text{on } Y,\\ f_p(\Delta w' + \text{tr}\boldsymbol{\xi}) + a_p(P'+P) = \lambda & \text{on } Y\end{cases} \tag{3.22}$$

where the constants $\lambda, \lambda' \in \mathbb{R}$ are the Lagrange multipliers arising from the constraints defined in Eq. (3.18). Integrating Eq. $(3.22)_1$ over Y implies $\lambda' = 0$; integrating $1/a_p$ $(3.22)_2$ over Y implies

$$\lambda\int_Y\frac{1}{a_p} = P + \int_Y\frac{f_p}{a_p}(\Delta w' + \text{tr}\boldsymbol{\xi}) \tag{3.23}$$

which can be used to determine the Lagrange multiplier λ. Eliminating $(P'+P)$-term in Eq. $(3.22)_1$ by Eq. $(3.22)_2$ we obtain

$$\nabla\nabla\cdot\mathbf{M} = 0, \quad \mathbf{M} = \tilde{\mathbf{L}}_p(\nabla\nabla w' + \boldsymbol{\xi}) + \frac{f_p}{a_p}\lambda\mathbf{I} = 0 \quad \text{on } Y \tag{3.24}$$

Taking Eq. (3.22) into account, by Eq. $(3.18)_1$ we find the minimum of the energy functional defined by Eq. (3.17) can be rewritten as

$$\begin{aligned}W^e(\boldsymbol{\xi}, P) &= \int_Y\left[\frac{1}{2}(\nabla\nabla w' + \boldsymbol{\xi})\cdot\mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi}) - \frac{f_p^2}{2a_p}(\Delta w' + \text{tr}\boldsymbol{\xi})^2 + \frac{\lambda f_p}{2a_p}(\Delta w' + \text{tr}\boldsymbol{\xi}) + \frac{1}{2}(P'+P)[f_p(\Delta_y w' + \text{tr}\boldsymbol{\xi}) + a_p(P'+P)]\right]\\ &= \int_Y\left[\frac{1}{2}(\nabla\nabla w' + \boldsymbol{\xi})\cdot\tilde{\mathbf{L}}_p(\nabla\nabla w' + \boldsymbol{\xi}) + \frac{\lambda^2}{2}\int_Y\frac{1}{a_p}\right.\\ &=: \frac{1}{2}\boldsymbol{\xi}\cdot\mathbf{L}^e\boldsymbol{\xi} + \mathbf{F}^e\cdot\boldsymbol{\xi}P + \frac{1}{2}a^eP^2\end{aligned} \tag{3.25}$$

For a heterogeneous membrane with microstructure as illustrated in Fig. 2(*a*), in general (w', P') cannot be smooth over the entire domain. Therefore, the partial different Eqs. (3.22) and (3.24) hold restricted to each phase. Across the interfaces between two phases, by Eq. (3.19) and corresponding to Eq. $(2.16)_{3,4}$ we infer the following interfacial conditions shall hold

$$\begin{cases}[\![w]\!] = 0, & [\![\nabla w]\!] = 0,\\ \mathbf{n}\cdot[\![\mathbf{M}]\!]\mathbf{n} = 0, & [\![\mathbf{n}\cdot\text{div}\mathbf{M} + \partial_\tau(\mathbf{t}\cdot\mathbf{M}\mathbf{n})]\!] = 0\end{cases} \tag{3.26}$$

where $[\![\cdot]\!]$ denote the jump across the interface. For specified heterogeneity of the membrane, one can solve Eqs. (3.24) and (3.26) for w' and determine the effective properties by Eq. (3.17). Examples of solutions will be presented in the next section.

4 Effective Flexoelectric Properties of Two-Phase Membranes

We now consider two-phase periodic flexoelectric membranes. As illustrated in Fig. 2, the microstructure of the composite

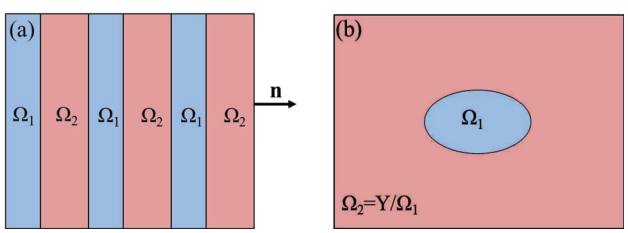

Fig. 2 A representative volume element of two-phase heterogeneous membrane: (*a*) a simple laminate; (*b*) inclusions embedded in a continuous matrix

membrane is described by a periodic piecewise constant material property tensor

$$\tilde{\mathbf{L}}_p(\mathbf{y}) = \tilde{\mathbf{L}}(\mathbf{y}) = \tilde{\mathbf{L}}^{(i)}, \quad f(\mathbf{y}) = f^{(i)}, \quad a(\mathbf{y}) = a^{(i)} \quad \text{if } \mathbf{y} \in \Omega_i \tag{4.1}$$

where Ω_i ($i = 1, 2$) is the region occupied by the ith phase and forms a subdivision of the unit cell Y. Here and subsequently, we drop the subscript $_p$ associated with material properties without the danger of confusion. Denote by $\theta_i = |\Omega_i|/|Y|$ the volume fraction of the ith phase. Our goad is to solve Eqs. (3.24) and (3.26) for w', which, by Eq. (4.1), is rewritten below

$$\begin{cases} \nabla\nabla \cdot \mathbf{M} = 0 & \text{on } \Omega_1 \cup \Omega_2, \\ [\![w']\!] = 0, \quad [\![\nabla w']\!] = 0, & \text{on } \partial\Omega_1, \\ \mathbf{n} \cdot [\![\mathbf{M}]\!]\mathbf{n} = 0, \quad [\![\mathbf{n} \cdot \mathrm{div}\mathbf{M} + \partial_\tau(\mathbf{t} \cdot \mathbf{M}\mathbf{n})]\!] = 0 & \text{on } \partial\Omega_1, \\ \text{periodic boundary conditions} & \text{on } \partial Y \end{cases} \tag{4.2}$$

where

$$\mathbf{M} = \begin{cases} \tilde{\mathbf{L}}^{(1)}(\nabla\nabla w' + \boldsymbol{\xi}) + \dfrac{f^{(1)}}{a^{(1)}}\lambda\mathbf{I} & \text{on } \Omega_1, \\ \tilde{\mathbf{L}}^{(2)}(\nabla\nabla w' + \boldsymbol{\xi}) + \dfrac{f^{(2)}}{a^{(2)}}\lambda\mathbf{I} & \text{on } \Omega_2 \end{cases} \tag{4.3}$$

Below we present solutions to Eqs. (4.2) and (4.3) for microstructures of simple laminates (Fig. 1(*b*)) and elliptic inclusions (Fig. 1(*a*)) in the dilute limit. The closed-form formula of effective properties is also obtained for periodic E-inclusions in finite volume fractions. These solutions give useful insights on engineering flexoelectric membranes for a variety of applications.

4.1 Simple Laminates. For a simple laminate as sketched in Fig. 2, we anticipate the curvature tensor $\nabla\nabla w'$ is piecewise constant for any applied average curvature $\boldsymbol{\xi}$, assuming that $\nabla\nabla w' = \boldsymbol{\xi}_i \in \mathbb{R}^{2\times 2}_{\mathrm{sym}}$ on Ω_i ($i = 1, 2$). Clearly, being a second gradient of a continuously differentiable scalar field w', across the interface $\boldsymbol{\xi}_1$ and $\boldsymbol{\xi}_2$ shall satisfy ($\mathbf{n}$ is the unit normal of the interface)

$$\boldsymbol{\xi}_1 - \boldsymbol{\xi}_2 = \rho\mathbf{n} \otimes \mathbf{n} \tag{4.4}$$

for some constant $\rho \in \mathbb{R}$. Clearly, Eq. $(4.2)_1$ is trivially satisfied inside Ω_1 and Ω_2, so is Eq. $(4.2)_2$. The third of Eq. (4.2) and the periodicity of w' require that

$$\begin{aligned} &\mathbf{n} \cdot \left[\tilde{\mathbf{L}}^{(1)}(\boldsymbol{\xi}_1 + \boldsymbol{\xi}) + \frac{f^{(1)}}{a^{(1)}}\lambda\mathbf{I} - \tilde{\mathbf{L}}^{(2)}(\boldsymbol{\xi}_2 + \boldsymbol{\xi}) - \frac{f^{(2)}}{a^{(2)}}\lambda\mathbf{I}\right]\mathbf{n} = 0, \\ &\theta_1\boldsymbol{\xi}_1 + \theta_2\boldsymbol{\xi}_2 = 0 \end{aligned} \tag{4.5}$$

Solving Eqs. (4.4) and (4.5) for $\boldsymbol{\xi}_1$ and $\boldsymbol{\xi}_2$, we obtain

$$\begin{aligned} &\boldsymbol{\xi}_1 = \theta_2\rho\mathbf{n} \otimes \mathbf{n} && \boldsymbol{\xi}_2 = -\theta_1\rho\mathbf{n} \otimes \mathbf{n}, && \Delta\tilde{\mathbf{L}} = \tilde{\mathbf{L}}_2 - \tilde{\mathbf{L}}_1, \\ &\Delta\boldsymbol{\beta} = \Delta\tilde{\mathbf{L}}(\mathbf{n} \otimes \mathbf{n}), && \rho = \frac{\boldsymbol{\xi} \cdot \Delta\boldsymbol{\beta} + \lambda_\Delta\gamma}{\tilde{D}_{\mathbf{n}}}, \\ &{}_\Delta\gamma = \gamma^{(2)} - \gamma^{(1)}, && \gamma^{(i)} = \frac{f^{(i)}}{a^{(i)}} \quad (i = 1, 2), \\ &\tilde{D}_{\mathbf{n}} = D(\theta_1\tilde{\mathbf{L}}_2 + \theta_2\tilde{\mathbf{L}}_1, \mathbf{n}), && D(\mathbf{L}, \mathbf{n}) := (\mathbf{n} \otimes \mathbf{n}) \cdot \mathbf{L}(\mathbf{n} \otimes \mathbf{n}) \end{aligned} \tag{4.6}$$

Further, for brevity we introduce

$$a^h = \left(\fint_Y \frac{1}{a}\right)^{-1}, \quad \gamma^a = \fint_Y \frac{f}{a} \tag{4.7}$$

where superscript ^{h} (^{a}) indicates the harmonic (arithmetic) mean. Inserting Eq. (4.6) into Eq. (3.23), we find that

$$\begin{aligned} &\lambda = a^h(\lambda_P P + \boldsymbol{\lambda}_H \cdot \boldsymbol{\xi}), \quad \lambda_P = \frac{\tilde{D}_{\mathbf{n}}}{\tilde{D}_{\mathbf{n}} + \theta_1\theta_2 a^h_\Delta\gamma^2}, \\ &\boldsymbol{\lambda}_H = \frac{\tilde{D}_{\mathbf{n}}\gamma^a\mathbf{I} - \theta_1\theta_2\Delta\gamma\Delta\,\boldsymbol{\beta}}{\tilde{D}_{\mathbf{n}} + \theta_1\theta_2 a^h_\Delta\gamma^2} \end{aligned} \tag{4.8}$$

and, hence,

$$\begin{aligned} &\rho = \rho_P P + \rho_H \cdot \boldsymbol{\xi}, \quad \rho_P = \frac{a^h_\Delta\gamma}{\tilde{D}_{\mathbf{n}} + \theta_1\theta_2 a^h_\Delta\gamma^2}, \\ &\rho_H = \frac{\Delta\,\boldsymbol{\beta} + a^h_\Delta\gamma\gamma^a\mathbf{I}}{\tilde{D}_{\mathbf{n}} + \theta_1\theta_2 a^h_\Delta\gamma^2} \end{aligned} \tag{4.9}$$

Inserting Eqs. (4.6), (4.8), and (4.9) into Eq. (3.25), we obtain

$$\begin{aligned} &\frac{1}{2}\boldsymbol{\xi} \cdot \mathbf{L}^e\boldsymbol{\xi} + \mathbf{F}^e \cdot \boldsymbol{\xi}P + \frac{1}{2}a^e|P|^2 \\ &= \frac{\lambda^2}{2a^h} + \int_Y \left[\frac{1}{2}(\nabla\nabla w' + \boldsymbol{\xi}) \cdot \mathbf{L}_p(\nabla\nabla w' + \boldsymbol{\xi})\right. \\ &= \frac{1}{2}\boldsymbol{\xi} \cdot (\theta_1\mathbf{L}_1 + \theta_2\tilde{\mathbf{L}}_2)\boldsymbol{\xi} - \theta_1\theta_2\rho\boldsymbol{\xi} \cdot \Delta\,\boldsymbol{\beta} + \frac{\theta_1\theta_2}{2}\rho^2\tilde{D}_{\mathbf{n}} + \frac{\lambda^2}{2a^h} \end{aligned}$$

Therefore, the effective material properties are given by

$$\begin{cases} \mathbf{L}^e = \theta_1\tilde{\mathbf{L}}_1 + \theta_2\tilde{\mathbf{L}}_2 + \theta_1\theta_2[\tilde{D}_{\mathbf{n}}\rho_H \otimes \rho_H \\ \qquad - (\Delta\,\boldsymbol{\beta} \otimes \rho_H + \rho_H \otimes \Delta\boldsymbol{\beta})] + a^h\boldsymbol{\lambda}_H \otimes \boldsymbol{\lambda}_H, \\ \mathbf{F}^e = \theta_1\theta_2\rho_P(\tilde{D}_{\mathbf{n}}\rho_H - \Delta\boldsymbol{\beta}) + a^h\lambda_P\boldsymbol{\lambda}_H \\ \qquad = a^h\lambda_P\boldsymbol{\lambda}_H(1 + \theta_1\theta_2 a^h_\Delta\gamma^2/\tilde{D}_{\mathbf{n}}) = a^h\boldsymbol{\lambda}_H, \\ a^e = a^h\lambda_P^2 + \theta_1\theta_2\rho_P^2\tilde{D}_{\mathbf{n}} = a^h\lambda_P \end{cases} \tag{4.10}$$

4.2 Elliptic Inclusions in the Dilute Limit. We now consider particulate composite membrane with particles of one phase embedded in another continuous phase. In the dilute limit, the interactions between particles are negligible; the overall heterogeneous membrane is well modeled by a single inclusion Ω_1 embedded in an infinite matrix $\Omega_2 = \mathbb{R}^2\backslash\Omega_1$. In this single inclusion model, the problem of interest can still be formulated as Eqs. (4.2) and (4.3) upon replacing the unit cell Y by $\mathbb{R}^2$ and periodic boundary conditions on w' by the decay condition

$$|\nabla\nabla w'(\mathbf{y})| \to 0 \quad \text{as} \quad |\mathbf{y}| \to +\infty \tag{4.11}$$

Closed-form solutions to Eqs. (4.2) and (4.3) for heterogeneous materials as specified by Eq. (4.1) are rare. In the context of classic elasticity, an exception is when the inclusion Ω_1 is an ellipsoid. As will be shown below, this exception persists for present setting of flexoelectricity since the critical *uniformity property* holds for the corresponding homogeneous problem (cf., Eqs. (4.12) and (4.13)). To see this, following the classic work of Eshelby [52,53] we first consider the *homogeneous* inclusion problem

$$\nabla\nabla \cdot [\tilde{\mathbf{L}}^{(2)}\nabla\nabla w' + \mathbf{M}^*\chi_{\Omega_1}] = 0 \quad \text{on } \mathbb{R}^2 \tag{4.12}$$

where $\mathbf{M}^* \in \mathbb{R}^{2\times 2}_{\text{sym}}$ is analogous to "eigenstress" in the context of Eshelby [52], and $\chi_{\Omega_1} = 1$ on Ω_1; $= 0$ otherwise. By the same argument of Eshelby [52] (also see Ref. [54]), it can be shown that a solution to Eqs. (4.12) and (4.11) satisfies that for any $\mathbf{x} \in \Omega_1$,

$$\nabla\nabla w'(\mathbf{x}) = \frac{-1}{(2\pi)^2}\int_{\mathbb{R}^2} \frac{\hat{\mathbf{k}}\otimes\hat{\mathbf{k}}(\hat{\mathbf{k}}\cdot\mathbf{M}^*\hat{\mathbf{k}})}{D(\tilde{\mathbf{L}}^{(2)},\hat{\mathbf{k}})}\int_{\Omega_1}\exp(-i\mathbf{k}\cdot(\mathbf{x}'-\mathbf{x}))d\mathbf{x}'d\mathbf{k} = -\mathbf{R}\mathbf{M}^* \tag{4.13}$$

where $\hat{\mathbf{k}} = \mathbf{k}/|\mathbf{k}|$ is a unit vector, $\mathbf{R}: \mathbb{R}^{2\times 2}_{\text{sym}} \to \mathbb{R}^{2\times 2}_{\text{sym}}$ is recognized as the *Eshelby tensor* and given by [55,56]

$$(\mathbf{R})_{piqj} = \int_{S^1} \frac{(\hat{\mathbf{k}})_p(\hat{\mathbf{k}})_i(\hat{\mathbf{k}})_q(\hat{\mathbf{k}})_j \det(\mathbf{A})}{D(\tilde{\mathbf{L}}^{(2)},\hat{\mathbf{k}})|\mathbf{A}\hat{\mathbf{k}}|^2}d\hat{\mathbf{k}}$$

where $\mathbf{A} \in \mathbb{R}^{2\times 2}_{\text{sym}}$ is such that $\Omega_1 = \{\mathbf{A}\mathbf{x} : \mathbf{x} \in B_2\}$, $B_2 \subset \mathbb{R}^2$ is a unit ball, $D(\mathbf{L},\hat{\mathbf{k}})$ is as defined in Eq. (4.6) and $S^1 = \{\mathbf{x} \in \mathbb{R}^2 : |\mathbf{x}| = 1\}$ is the unit sphere.

The remarkable uniformity property of $\nabla\nabla w'$ on Ω_1, i.e., Eq. (4.13), entails closed-form solutions to the *inhomogeneous* problem (4.2) by solutions to the homogeneous problem (4.12). To see this, we first observe that a solution to Eq. (4.12) automatically satisfies Eq. $(4.2)_1$ on the matrix Ω_2 since they are the same equations and on the inclusion Ω_1 since $\nabla\nabla w'$ is uniform on Ω_1 by Eq. (4.13). Further, solutions to Eq. (4.12) necessarily satisfy the following interfacial conditions on $\partial\Omega_1$

$$\begin{cases} \mathbf{n}\cdot[\tilde{\mathbf{L}}^{(2)}\nabla\nabla w' + \mathbf{M}^*\chi_{\Omega_1}]\mathbf{n} = 0 & \text{on } \partial\Omega_1, \\ \Big[\Big[\mathbf{n}\cdot\text{div}[\tilde{\mathbf{L}}^{(2)}\nabla\nabla w' + \mathbf{M}^*\chi_{\Omega_1}] \\ \quad + \partial_\tau\{\mathbf{t}\cdot[\tilde{\mathbf{L}}^{(2)}\nabla\nabla w' + \mathbf{M}^*\chi_{\Omega_1}]\mathbf{n}\}\Big]\Big] = 0 & \text{on } \partial\Omega_1 \end{cases}$$

which can be rewritten as

$$\begin{cases} \mathbf{n}\cdot[\mathbf{L}^{(2)}\nabla\nabla w']\mathbf{n}|_{\partial\Omega_1^+} = \mathbf{n}\cdot[\mathbf{L}^{(2)}\nabla\nabla w']\mathbf{n}|_{\partial\Omega_1^-} + \mathbf{n}\cdot\mathbf{M}^*\mathbf{n}, \\ \{\mathbf{n}\cdot\text{div}[\mathbf{L}^{(2)}\nabla\nabla w'] + \partial_\tau[\mathbf{t}\cdot(\mathbf{L}^{(2)}\nabla\nabla w')\mathbf{n}]\}|_{\partial\Omega_1^+} \\ = \{\mathbf{n}\cdot\text{div}[\mathbf{L}^{(2)}\nabla\nabla w'] + \partial_\tau[\mathbf{t}\cdot(\mathbf{L}^{(2)}\nabla\nabla w')\mathbf{n}]\}|_{\partial\Omega_1^-} + \partial_\tau(\mathbf{t}\cdot\mathbf{M}^*\mathbf{n}) \end{cases} \tag{4.14}$$

Inserting Eq. (4.3) into Eqs. $(4.2)_2$ and $(4.2)_3$, they can be rewritten as

$$\begin{cases} \mathbf{n}\cdot[\mathbf{L}^{(2)}\nabla\nabla w']\mathbf{n}|_{\partial\Omega_1^+} = \mathbf{n}\cdot[\mathbf{L}^{(1)}\nabla\nabla w']\mathbf{n}|_{\partial\Omega_1^-} - \mathbf{n}\cdot(\Delta\mathbf{L}\boldsymbol{\xi})\mathbf{n} - {}_\Delta\gamma\lambda, \\ \mathbf{n}\cdot\text{div}[\mathbf{L}^{(2)}\nabla\nabla w']|_{\partial\Omega_1^+} + \partial_\tau\{\mathbf{t}\cdot[\mathbf{L}^{(2)}\nabla\nabla w']\mathbf{n}\}|_{\partial\Omega_1^+} \\ \quad = \{\mathbf{n}\cdot\text{div}[\mathbf{L}^{(1)}\nabla\nabla w'] + \partial_\tau[\mathbf{t}\cdot(\mathbf{L}^{(1)}\nabla\nabla w')\mathbf{n}]\}|_{\partial\Omega_1^-} \\ -\partial_\tau[\mathbf{t}\cdot(\Delta\tilde{\mathbf{L}}\boldsymbol{\xi})\mathbf{n}] \end{cases} \tag{4.15}$$

Further, we notice that $\text{div}[\mathbf{L}^{(1)}\nabla\nabla w']|_{\partial\Omega_1^-} = 0$ for a solution to Eq. (4.12) according to Eq. (4.13). Comparing Eq. (4.15) with Eq. (4.14) we conclude that if the "equivalent eigenstress" $\mathbf{M}^*$ is such that

$$\mathbf{M}^* = -[\Delta\tilde{\mathbf{L}}(\nabla\nabla w' + \boldsymbol{\xi}) + {}_\Delta\gamma\lambda\mathbf{I}]|_{\partial\Omega_1^-} = \Delta\mathbf{L}\tilde{\mathbf{R}}\mathbf{M}^* - \Delta\tilde{\mathbf{L}}\boldsymbol{\xi} - {}_\Delta\gamma\lambda\mathbf{I} \tag{4.16}$$

then Eq. (4.15) is satisfied by a solution satisfying Eqs. (4.14) and (4.13).

The above relation for equivalence between two problems can be obtained by a more direct argument. To see this, let us first rewrite the inhomogeneous problem Eqs. (4.2) and (4.3) in a compact form as

$$\nabla\nabla\cdot[\tilde{\mathbf{L}}^{(2)}\nabla\nabla w' - [\Delta\tilde{\mathbf{L}}(\nabla\nabla w' + \boldsymbol{\xi}) + {}_\Delta\gamma\lambda\mathbf{I}]\chi_{\Omega_1}] = 0 \quad \text{on} \quad \mathbb{R}^2 \tag{4.17}$$

Comparing with Eq. (4.12), we see that the above equation has the same "source" term if Eqs. (4.13) and (4.16) hold, and hence, their solutions coincide.

Knowing $\nabla\nabla w'$ in one of the phase such as Eq. (4.13) is sufficient to determine the effective properties of the composite membrane. In subsequent calculations we take the unit cell Y as finite; the physical assumption is that though Y is finite, since $\theta_1 \ll 1$ (dilute limit), solutions to the inhomogeneous problem (4.2) and (4.3) restricted to the inclusion Ω_1 are well approximated by the single inclusion model where Y is are assumed to be infinite (i.e., Eq. (4.17) with decay condition (4.11)). In other words, in the dilute limit we assume that a solution to the inhomogeneous problem (4.2) and (4.3) for elliptic Ω_1 still satisfies Eq. (4.13) for finite unit cell Y. Therefore, by Eqs. (3.23) and (4.13) we find that

$$\begin{aligned} \lambda &= a^h\left(P + \gamma^a\text{tr}\boldsymbol{\xi} - {}_\Delta\gamma\fint_Y \chi_{\Omega_1}\Delta w'\right) \\ &= a^h[P + \gamma^a\text{tr}\boldsymbol{\xi} + \theta_1{}_\Delta\gamma\text{tr}(\mathbf{R}\mathbf{M}^*)] \end{aligned} \tag{4.18}$$

Inserting the above equation into Eq. (4.16) we obtain that

$$\begin{aligned} &[\Delta\tilde{\mathbf{L}}\mathbf{R} - \mathbf{II} - \theta_1 a^h{}_\Delta\gamma^2\mathbf{I}\otimes(\mathbf{R}\mathbf{I})]\mathbf{M}^* \\ &\quad = [\Delta\tilde{\mathbf{L}} + {}_\Delta\gamma a^h\gamma^a\mathbf{I}\otimes\mathbf{I}]\boldsymbol{\xi} + {}_\Delta\gamma a^h P\mathbf{I} \end{aligned}$$

Assume that the tensor $\Delta\tilde{\mathbf{L}}\mathbf{R} - \mathbf{II} - \theta_1 a^h_\Delta\gamma^2\mathbf{I}\otimes(\mathbf{R}\mathbf{I}) : \mathbb{R}^{2\times 2}_{\text{sym}} \to \mathbb{R}^{2\times 2}_{\text{sym}}$ is invertible, we have

$$\begin{aligned} \mathbf{R}\mathbf{M}^* &= \mathbf{N}((\Delta\tilde{\mathbf{L}} + {}_\Delta\gamma a^h\gamma^a\mathbf{I}\otimes\mathbf{I})\boldsymbol{\xi} + {}_\Delta\gamma a^h P\mathbf{I}), \\ \mathbf{N} &= [\Delta\tilde{\mathbf{L}} - \mathbf{R}^{-1} - \theta_1 a^h_\Delta\gamma^2\mathbf{I}\otimes\mathbf{I}]^{-1} \end{aligned} \tag{4.19}$$

where the tensor $\mathbf{N}: \mathbb{R}^{2\times 2} \to \mathbb{R}^{2\times 2}$ is introduced for brevity. Therefore, by Eq. (4.18) we have

$$\begin{aligned} &\lambda = a^h(\lambda_P P + \boldsymbol{\lambda}_H\cdot\boldsymbol{\xi}), \\ &\lambda_P = 1 + \theta_1 a^h_\Delta\gamma^2\mathbf{I}\cdot\mathbf{N}\mathbf{I}, \quad \boldsymbol{\lambda}_H = \theta_1\Delta\gamma\Delta\tilde{\mathbf{L}}(\mathbf{N}(\mathbf{I})) + \gamma^a\lambda_P\mathbf{I} \end{aligned} \tag{4.20}$$

To determine the effective properties, we need to evaluate the integral in Eq. (3.25). To this end, we first notice that upon multiplying Eq. $(4.2)_1$ by w', by the divergence theorem we have (cf. Eq. (4.1))

$$\fint_Y \nabla\nabla w'\cdot[\tilde{\mathbf{L}}\nabla\nabla w' - (\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} + {}_\Delta\gamma\lambda\mathbf{I})\chi_{\Omega_1}] = 0$$

and hence, (cf. Eq. (4.13))

$$\begin{aligned} \fint_Y \nabla\nabla w'\cdot\tilde{\mathbf{L}}\nabla\nabla w' &= \fint_Y \nabla\nabla w'\cdot(\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} + {}_\Delta\gamma\lambda\mathbf{I})\chi_{\Omega_1} \\ &= -\theta_1(\mathbf{R}\mathbf{M}^*)\cdot(\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} + {}_\Delta\gamma\lambda\mathbf{I}) \end{aligned} \tag{4.21}$$

Therefore, the effective internal energy density (cf., Eq. (3.25))

$$\begin{aligned} W^e(\boldsymbol{\xi},P) &= \fint_Y \left[\frac{1}{2}\nabla\nabla w' \cdot \tilde{\mathbf{L}}\nabla\nabla w' + \boldsymbol{\xi}\cdot\tilde{\mathbf{L}}\nabla\nabla w' + \frac{1}{2}\boldsymbol{\xi}\cdot\mathbf{L}\boldsymbol{\xi}\right] + \frac{\lambda^2}{2a^h} \\ &= -\frac{\theta_1}{2}(\mathbf{R}\mathbf{M}^*)\cdot(\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} + {}_\Delta\gamma\lambda\mathbf{I}) + \theta_1\boldsymbol{\xi}\cdot\Delta\tilde{\mathbf{L}}\mathbf{R}\mathbf{M}^* \\ &\quad + \frac{\lambda^2}{2a^h} + \frac{1}{2}\boldsymbol{\xi}\cdot\left(\fint_Y \tilde{\mathbf{L}}\right)\boldsymbol{\xi} \\ &= \frac{\theta_1}{2}(\mathbf{R}\mathbf{M}^*)\cdot\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} - \frac{\lambda}{2}\left(\frac{\lambda}{a^h} - P - \gamma^a \mathrm{tr}\boldsymbol{\xi}\right) + \frac{\lambda^2}{2a^h} \\ &\quad + \frac{1}{2}\boldsymbol{\xi}\cdot\left(\fint_Y \tilde{\mathbf{L}}\right)\boldsymbol{\xi} \\ &=: \frac{1}{2}\boldsymbol{\xi}\cdot\mathbf{L}^e\boldsymbol{\xi} + \mathbf{F}^e\cdot\boldsymbol{\xi}P + \frac{1}{2}a^e|P|^2 \end{aligned} \tag{4.22}$$

and the material properties are given by (see Eq. (4.20) for values of $\lambda_P, \boldsymbol{\lambda}_H$)

$$\begin{cases} \mathbf{L}^e = \theta_1\left[\Delta\tilde{\mathbf{L}}\mathbf{N}\Delta\tilde{\mathbf{L}} + \dfrac{a^h}{2}{}_\Delta\gamma\gamma^a(\Delta\tilde{\mathbf{L}}\mathbf{N}\mathbf{I}\otimes\mathbf{I} + \mathbf{I}\otimes\Delta\tilde{\mathbf{L}}\mathbf{N}\mathbf{I})\right] \\ \qquad + \dfrac{a^h\gamma^a}{2}(\boldsymbol{\lambda}_H\otimes\mathbf{I} + \mathbf{I}\otimes\boldsymbol{\lambda}_H) + \theta_1\tilde{\mathbf{L}}_1 + \theta_2\tilde{\mathbf{L}}_2, \\ \mathbf{F}^e = \dfrac{a^h}{2}(\boldsymbol{\lambda}_H - \gamma^a\lambda_P\mathbf{I}) + a^h\left(\dfrac{1}{2}\boldsymbol{\lambda}_H + \dfrac{\lambda_P\gamma^a}{2}\mathbf{I}\right) = a^h\boldsymbol{\lambda}_H, \\ a^e = a^h\lambda_P \end{cases} \tag{4.23}$$

We remark that the formula (4.10) and (4.23) for simple laminates and dilute elliptic inclusions are applicable for general anisotropic constituent phases. In subsequent sections, we shall restrict ourselves to isotropic constituent phases. In this case, the factor appears in the denominator of integrand in Eq. (4.13), by Eqs. (4.6), (2.3), and (3.6), is given by

$$D(\tilde{\mathbf{L}}^{(2)},\hat{\mathbf{k}}) = 2\mu_b^{(2)} + \lambda_b^{(2)} - \frac{|f^{(2)}|^2}{a^{(2)}} =: k_b^{(2)} \tag{4.24}$$

which is independent of $\hat{\mathbf{k}}$. This property of isotropic materials implies relations between system of equations such as Eq. (4.12) and simple scalar potential problems that can be conveniently explored by Fourier analysis [57] or Green's functions [52]. Equation (4.24) is also critical for showing the uniformity property (4.13) can be generalized to periodic E-inclusions for dilatational $\mathbf{M}^* = m^*\mathbf{I}$.

4.3 Periodic E-inclusions. The closed-form predictions (4.23) are applicable to the dilute limit. For finite volume fractions, closed-form solutions to the inhomogeneous problem (4.2) and (4.3) can be obtained for periodic E-inclusions using similar argument as for ellipses in the dilute limit. This approach has been explored to obtain closed-form predictions of effective properties in physical contexts such as conductivity, elasticity, magnetoelectricity, and thermoelectricity. Below we derive the closed-form formula of flexoelectric properties for a two-phase membrane of periodic E-inclusions.

First of all, we recall that a periodic E-inclusion in a finite unit cell $Y \subset \mathbb{R}^2$ is defined as a domain $\Omega_1 \subset Y$ such that a solution to the potential problem [57,58]

$$\begin{cases} \nabla^2\phi = \theta_1 - \chi_{\Omega_1} & \text{on } Y, \\ \text{periodic boundary conditions} & \text{on } \partial Y \end{cases} \tag{4.25}$$

satisfies the overdetermined condition

$$\nabla\nabla\phi = -(1-\theta_1)\mathbf{Q} \quad \text{on } \Omega_1 \tag{4.26}$$

where $\theta_1 = |\Omega_1|/|Y|$ is the volume fraction of the inclusion, and $\mathbf{Q} \in \mathbb{Q} := \{\mathbf{M} \in \mathbb{R}^{2\times2}_{\mathrm{sym}} : \mathbf{M} \text{ is nonnegative, } \mathrm{tr}\mathbf{M} = 1\}$ is referred to as the shape matrix of the E-inclusions. By Fourier analysis and Eq. (4.25), Eq. (4.26) implies that for any $\mathbf{x} \in \Omega_1$,

$$\begin{aligned} \nabla\nabla\phi &= -\sum_{\mathbf{k}\in\mathcal{K}\setminus\{0\}} \hat{\mathbf{k}}\otimes\hat{\mathbf{k}}\int_{\Omega_1}\exp(-i\mathbf{k}\cdot(\mathbf{x}'-\mathbf{x}))d\mathbf{x}' \\ &= -(1-\theta_1)\mathbf{Q} \end{aligned} \tag{4.27}$$

where $\mathcal{K}$ is the reciprocal lattice associated with the (primitive) unit cell Y ([59] chap. 4). We remark that the overdetermined condition (4.26) places strong restrictions on the shape of Ω_1; the existence of periodic E-inclusions can be proved by the theory of variational inequalities [57]. The reader is referred to Refs. [57,58] for examples of periodic E-inclusions with volume fractions, shape matrices, unit cells, and in two and three dimensions. A simply connected periodic E-inclusion in two dimensions is also called Vigdergauz microstructures [60]. The terminology "*E*-inclusion" arises from the associations with the "Eshelby," "ellipsoid," and "extremal" properties of such geometries.

To solve the inhomogeneous flexoelectricity problem (4.2) and (4.3) for periodic E-inclusions, we notice that, by Fourier analysis, a solution to the homogeneous problem

$$\begin{cases} \nabla\nabla\cdot[\tilde{\mathbf{L}}^{(2)}\nabla\nabla w' + \mathbf{M}^*\chi_{\Omega_1}] = 0 & \text{on } Y, \\ \text{periodic boundary conditions} & \text{on } \partial Y \end{cases} \tag{4.28}$$

satisfies that for any $\mathbf{x} \in Y$,

$$\nabla\nabla w'(\mathbf{x}) = -\sum_{\mathbf{k}\in\mathcal{K}\setminus\{0\}} \frac{\hat{\mathbf{k}}\otimes\hat{\mathbf{k}}(\hat{\mathbf{k}}\cdot\mathbf{M}^*\hat{\mathbf{k}})}{D(\tilde{\mathbf{L}}^{(2)},\hat{\mathbf{k}})}\int_{\Omega_1}\exp(-i\mathbf{k}\cdot(\mathbf{x}'-\mathbf{x}))d\mathbf{x}'d\mathbf{k} \tag{4.29}$$

If the material is isotropic, by Eq. (4.24) and comparing with Eq. (4.27) we conclude that if $\mathbf{M}^* = m^*\mathbf{I}$ for some $m^* \in \mathbb{R}$ and Ω_1 is a periodic E-inclusion with volume fraction θ_1 and shape matrix $\mathbf{Q}$, then

$$\nabla\nabla w'(\mathbf{x}) = -\frac{(1-\theta_1)m^*}{k_b^{(2)}}\mathbf{Q} \quad \forall\mathbf{x}\in\Omega_1 \tag{4.30}$$

In analogy with Eqs. (4.14)–(4.17), it can be shown that the solution to the inhomogeneous problem (4.2) and (4.3) coincides with that of the homogeneous problem (4.28) if the following algebraic relation holds:

$$\begin{aligned} m^*\mathbf{I} &= -[\Delta\tilde{\mathbf{L}}(\nabla\nabla w' + \boldsymbol{\xi}) + {}_\Delta\gamma\lambda\mathbf{I}]|_{\partial\Omega_1^-} \\ &= \frac{\theta_2 m^*}{k_b^{(2)}}\Delta\tilde{\mathbf{L}}\mathbf{Q} - \Delta\tilde{\mathbf{L}}\boldsymbol{\xi} - {}_\Delta\gamma\lambda\mathbf{I} \end{aligned} \tag{4.31}$$

In particular, if the shape matrix as $\mathbf{Q} = \mathbf{I}/2$, then

$$\boldsymbol{\xi} = \frac{1}{2}\left[-\frac{m^* + {}_\Delta\gamma\lambda}{\Delta\kappa} + \frac{\theta_2 m^*}{k_b^{(2)}}\right]\mathbf{I} =: \frac{H}{2}\mathbf{I} \Rightarrow m^* = \frac{\Delta\kappa k_b^{(2)}H + {}_\Delta\gamma k_b^{(2)}\lambda}{\theta_2\Delta\kappa - k_b^{(2)}} \tag{4.32}$$

where H is the average applied mean curvature

$$\Delta\kappa = \kappa^{(2)} - \kappa^{(1)}, \quad \kappa^{(i)} = \mu_b^{(i)} + \lambda_b^{(i)} - \frac{|f^{(i)}|^2}{a^{(i)}}, \quad \frac{1}{2}\tilde{\mathbf{L}}^{(i)}\mathbf{I} = \kappa^{(i)}\mathbf{I} \tag{4.33}$$

By Eqs. (3.23) and (4.31) we have

$$\lambda = a^h\left(P + \gamma^a \mathrm{tr}\boldsymbol{\xi} + \theta_1\theta_2\frac{{}_\Delta\gamma m^*}{k_b^{(2)}}\right) \tag{4.34}$$

By Eqs. (4.32) and (4.34) we can express m^* and λ in terms of H and P as

$$\lambda = a^h(\lambda_P P + \lambda_H H), \quad \lambda_P = \frac{\theta_2\Delta\kappa - k_b^{(2)}}{\theta_2\Delta\kappa - k_b^{(2)} - \theta_1\theta_2 a_\Delta^h\gamma^2},$$
$$\lambda_H = \frac{\gamma^a(\theta_2\Delta\kappa - k_b^{(2)}) + \theta_1\theta_2\Delta\kappa_\Delta\gamma}{\theta_2\Delta\kappa - k_b^{(2)} - \theta_1\theta_2 a_\Delta^h\gamma^2}, \quad \theta_1\theta_2\frac{{}_\Delta\gamma m^*}{k_b^{(2)}} = \frac{\lambda}{a^h} - P - \gamma^a H \tag{4.35}$$

Therefore, by Eqs. (4.30) and (4.33), we find that the effective internal energy density is given by (cf., Eqs. (3.25), (4.21), (4.22) and recall that $\boldsymbol{\xi} = (H/2)\mathbf{I}$, $\mathbf{Q} = \mathbf{I}/2$)

$$\begin{aligned}
W^e(\boldsymbol{\xi}, P) &= \fint_Y \left[\frac{1}{2}\nabla\nabla w' \cdot (\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} + {}_\Delta\gamma\lambda\mathbf{I})\chi_{\Omega_1} - \boldsymbol{\xi}\cdot\Delta\tilde{\mathbf{L}}\nabla\nabla w'\chi_{\Omega_1}\right. \\
&\quad \left. + \frac{1}{2}\boldsymbol{\xi}\cdot\tilde{\mathbf{L}}\boldsymbol{\xi}\right] + \frac{\lambda^2}{2a^h} \\
&= -\frac{\theta_1}{2}(\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} - {}_\Delta\gamma\lambda\mathbf{I})\cdot\fint_{\Omega_1}\nabla\nabla w' + \frac{\lambda^2}{2a^h} + \frac{1}{2}\boldsymbol{\xi}\cdot\left(\fint_Y \mathbf{L}\right)\boldsymbol{\xi} \\
&= \frac{\theta_1\theta_2 m^*}{4k_b^{(2)}}(\Delta\tilde{\mathbf{L}}\boldsymbol{\xi} - {}_\Delta\gamma\lambda\mathbf{I})\cdot\mathbf{I} + \frac{\lambda^2}{2a^h} + \frac{1}{2}\boldsymbol{\xi}\cdot\left(\fint_Y \tilde{\mathbf{L}}\right)\boldsymbol{\xi} \\
&= \frac{\theta_1\theta_2 m^*}{2k_b^{(2)}}(\Delta\kappa H - {}_\Delta\gamma\lambda) + \frac{\lambda^2}{2a^h} + \frac{1}{2}\boldsymbol{\xi}\cdot\left(\fint_Y \tilde{\mathbf{L}}\right)\boldsymbol{\xi} \\
&= \frac{\Delta\kappa}{2{}_\Delta\gamma}\left(\frac{\lambda}{a^h} - P - \gamma^a H\right)H + \frac{1}{2}\lambda(P + \gamma^a H) + \frac{1}{2}\boldsymbol{\xi}\cdot\left(\fint_Y \mathbf{L}\right)\boldsymbol{\xi} \\
&=: \frac{1}{2}\boldsymbol{\xi}\cdot\mathbf{L}^e\boldsymbol{\xi} + \mathbf{F}^e\cdot\boldsymbol{\xi}P + \frac{1}{2}a^e|P|^2
\end{aligned}$$

Eliminating m^* and λ by Eq. (4.35), we conclude that

$$\begin{cases}
\mathbf{I}\cdot\mathbf{L}^e\mathbf{I}/4 = \dfrac{\Delta\kappa}{{}_\Delta\gamma}(\lambda_H - \gamma^a) + a^h\gamma^a\lambda_H + \mathbf{I}\cdot(\theta_1\tilde{\mathbf{L}}_1 + \theta_2\tilde{\mathbf{L}}_2)\mathbf{I}/4, \\
\mathbf{F}^e\cdot\mathbf{I}/2 = \dfrac{\Delta\kappa}{2{}_\Delta\gamma}(\lambda_P - 1) + \dfrac{a^h}{2}(\lambda_H + \gamma^a\lambda_P) = a^h\lambda_H, \\
a^e = a^h\lambda_P
\end{cases} \tag{4.36}$$

In addition, we assume that the heterogeneous membrane is effectively isotropic in the sense that effective internal energy density is given by

$$W^e(\boldsymbol{\xi}, P) = \mu_b^e\boldsymbol{\xi}\cdot\boldsymbol{\xi} + \frac{\lambda_b^e}{2}(\mathrm{tr}\boldsymbol{\xi})^2 + f^e P\mathrm{tr}\boldsymbol{\xi} + \frac{1}{2}a^e|P|^2$$

Then by Eq. (4.36) we have (see Eqs. (3.6), (4.6), (4.7), (4.24), (4.33), and (4.35) for definitions of constants in terms of the original material constants)

$$\begin{aligned}
\mu_b^e + \lambda_b^e &= \frac{\Delta\kappa}{{}_\Delta\gamma}(\lambda_H - \gamma^a) + a^h\gamma^a\lambda_H + \mu_b^a + \lambda_b^a, \\
f^e &= a^h\lambda_H, \quad a^e = a^h\lambda_P
\end{aligned} \tag{4.37}$$

where $\mu_b^a = \theta_1\mu_b^{(1)} + \theta_2\mu_b^{(2)}$ and $\lambda_b^a = \theta_1\lambda_b^{(1)} + \lambda_2\mu_b^{(2)}$ are the arithmetic average of μ_b and λ_b on the unit cell, respectively. To compare with experiments, it is more convenient

$$\gamma^e = \frac{f^e}{a^e} = \frac{\lambda_H}{\lambda_P} = \gamma^a - \theta_1\theta_2\frac{\Delta\kappa_\Delta\gamma}{k_b^{(2)} - \theta_2\Delta\kappa} \tag{4.38}$$

5 Results and Discussion

We have obtained the closed-form predictions of the effective elastic-flexoelectric properties of heterogeneous membranes. These expressions can now be conveniently used to make an assessment of the effect of inhomogeneities on the effective flexoelectric coefficient. As example, we consider two-phase membranes of protein inclusions (phase 1) in lipid bilayer (phase 2). Representative material properties are given below [42]

$$\begin{cases}
\mu_b^{(1)} = \mu_b^{(2)} = 0, \\
\lambda_b^{(2)} = 6.9\times10^{-23}\,\mathrm{N\cdot m}, \quad \lambda_b^{(1)} = 5.52\times10^{-22}\,\mathrm{N\cdot m}, \\
a^{(2)} = 2.26\times10^{19}\,\dfrac{\mathrm{N\cdot m}}{C^2}, \quad a^{(1)} = 7.53\times10^{18}\,\dfrac{\mathrm{N\cdot m}}{C^2}, \\
\gamma^{(2)} = \dfrac{f^{(2)}}{a^{(2)}} = 4.43\times10^{-22}C, \quad \gamma^{(1)} = \dfrac{f^{(1)}}{a^{(1)}} = 4.43\times10^{-22}C
\end{cases} \tag{5.1}$$

Since experiments directly measure polarization per unit area (as a function of curvature), i.e., the constant γ defined by Eq. (2.22), below we present the effective constant $\gamma^e = f^e/a^e$ and how it depends on the shapes and volume fractions of the

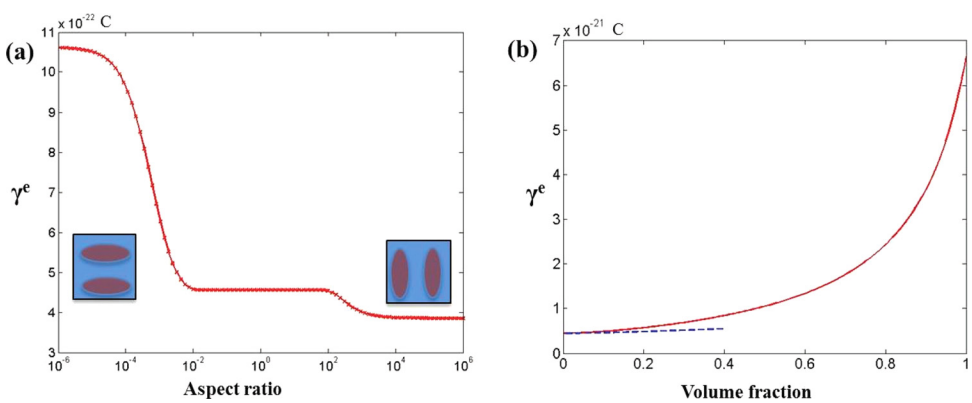

Fig. 3 Effective flexoelectric of protein inclusions in lipid bilayer: (*a*) the effective constant $\gamma^e = f^e/a^e$ as a function of aspect ratio of protein ellipsoid. The volume fraction of protein is assumed to be 0.1, and (*b*) the effective constant $\gamma^e = f^e/a^e$ as a function of volume fraction of inclusion. The solid curve is predicted by Eq. (4.38); the dashed curve is calculated by Eq. (5.2) assuming $a_x/a_y = 1$.

inclusions. In the dilute limit, by Eq. (4.23) we have that for bending in $\mathbf{e}_x$-direction

$$\gamma^e := \frac{\mathbf{e}_x \cdot \mathbf{F}^e \mathbf{e}_x}{a^e} = \frac{\mathbf{e}_x \cdot \boldsymbol{\lambda}_H \mathbf{e}_x}{\lambda_P} \tag{5.2}$$

which depends on the aspect ratio of the ellipse a_x/a_y. Figure 3(*a*) show the functional dependence of γ^e on the aspect ratio a_x/a_y at the fixed volume fraction $\theta_2 = 0.1$. We remark that $a_x/a_y \ll 1$ ($a_x/a_y \gg 1$) implies that the microstructure is essentially laminates parallel to $\mathbf{e}_x$ ($\mathbf{e}_y$).

The solid curve in Fig. 3(*b*) show the functional dependence of γ^e on the volume fraction predicted by Eq. (4.38); the dashed curve is calculated by Eq. (5.2) assuming $a_x/a_y = 1$.

We have developed a general framework to estimate the effective elastic, dielectric, and flexoelectric properties of heterogeneous membranes. Our results are analytical due to the approximations made (dilute limit) and the simplified microstructures considered (circular inhomogeneities and laminate). However, the presented framework can be solved numerically to consider more complex microstructures and to "design" flexoelectricity. There is strong evidence of the importance of flexoelectricity in two-dimensional structures such as graphene and soft-lipid bilayers and the presented work can serve as the starting point for further explorations. In particular we note the rather strong effect of inclusion shape on the flexoelectric response. Given this outcome, specifically introducing inhomogeneities that are polar may provide avenues to significantly enhance flexoelectric response for both artificial and natural membranes.

Several challenges remain. We have stayed strictly within the linearized regime. For solid membranes, out of plane deformation modes are coupled to the in-plane behavior. Homogenization of nonlinear membranes is nontrivial and presents both a challenging problem and opportunity for future work.

Acknowledgment

L.L. acknowledges the support of the National Science Foundation (NSF) under Grant No. CMMI-1238835 and AFOSR (YIP-12). P.S. gratefully acknowledges support from NSF IMI center IIMEC 0844082 and NSF CMMI NIRT Grant 0708096.

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