# The Calculus of Boundary Variations and the Dielectric Boundary Force in the Poisson–Boltzmann Theory for Molecular Solvation

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**Abstract**

In a continuum model of the solvation of charged molecules in an aqueous solvent, the classical Poisson–Boltzmann (PB) theory for the electrostatics of an ionic solution is generalized to include the solute point charges and the dielectric boundary that separates the high-dielectric solvent from the low-dielectric solutes. With such a setting, we construct an effective electrostatic free-energy functional of ionic concentrations. The functional admits a unique minimizer whose corresponding electrostatic potential is the unique solution to the boundary-value problem of the nonlinear dielectric-boundary PB equation. The negative first variation of this minimum free energy with respect to variations of the dielectric boundary defines the normal component of the dielectric boundary force. Together with the solute-solvent interfacial tension and van der Waals interaction forces, such boundary force drives an underlying charged molecular system to a stable equilibrium, as described by a variational implicit-solvent model. We develop an $L^2$-theory for boundary variations and derive an explicit formula of the dielectric boundary force. Our results agree with a molecular-level prediction that the electrostatic force points from the high-dielectric aqueous solvent to the low-dielectric charged molecules. Our method of analysis is general as it does not rely on any variational principles.

**Keywords:** Molecular solvation, electrostatic free energy, dielectric boundary force, nonlinear Poisson–Boltzmann equation, the calculus of boundary variations.

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# 1 Introduction

Charged molecules such as proteins polarize the surrounding aqueous solvent (i.e., water or salted water), generating a strong electrostatic force [15, 32, 46]. In a class of implicit-solvent (i.e., continuum-solvent) models, electrostatic interactions in a charged molecular system are described by the Poisson–Boltzmann (PB) theory [3, 9, 16, 18, 37, 49, 52]. Key in such a description is the dielectric boundary (i.e., the solute-solvent interface) that separates the high-dielectric solvent from the low-dielectric solutes (i.e., charged molecules). The dielectric boundary force—the macroscopic electrostatic force exerted on the boundary—plays a critical role in the molecular conformational dynamics [21, 22, 29, 53, 57]. Here, we present a detailed mathematical study of this force within the PB framework.

Briefly, the classical PB theory provides a continuum description of electrostatic interactions in an ionic solution through the nonlinear PB equation [2, 8, 19, 25, 28]

$$\nabla \cdot \varepsilon \nabla \psi - B'(\psi) = -\rho \qquad \text{in } \Omega_0, \tag{1.1}$$

where $\Omega_0 \subseteq \mathbb{R}^3$ is the region of the ionic solution, $\varepsilon$ is the dielectric coefficient, $\rho : \Omega_0 \to \mathbb{R}$ is the density of fixed charges, and $\psi : \Omega_0 \to \mathbb{R}$ is the electrostatic potential. In (1.1), the function $B : \mathbb{R} \to \mathbb{R}$ is defined by

$$B(s) = \beta^{-1} \sum_{j=1}^{M} c_j^\infty \left( e^{-\beta q_j s} - 1 \right) \qquad \forall s \in \mathbb{R}, \tag{1.2}$$

where $\beta = (k_{\mathrm{B}} T)^{-1}$ with $k_{\mathrm{B}}$ the Boltzmann constant and $T$ the temperature, $M$ is the total number of ionic species, $c_j^\infty$ is the bulk ionic concentration of the $j$th ionic species, and $q_j = z_j e$ is the charge of an ion of the $j$th species with $z_j$ the valence of such an ion and $e$ the elementary charge. The PB equation (1.1) is a combination of Poisson's equation

$$\nabla \cdot \varepsilon \nabla \psi = -\left( \rho + \sum_{j=1}^{M} q_j c_j \right) \qquad \text{in } \Omega_0,$$

where $c_j : \Omega_0 \to [0, \infty)$ is the ionic concentration of the $j$th ionic species, and the Boltzmann distributions for the equilibrium ionic concentrations

$$c_j(x) = c_j^\infty e^{-\beta q_j \psi(x)}, \qquad x \in \Omega_0, \ j = 1, \ldots, M.$$

In modeling charged molecules in an aqueous solvent with an implicit-solvent, the PB theory is generalized to include the point charges of molecules and the dielectric boundary [9, 16, 18, 37, 49, 52]. To be more specific, let us assume that the entire solvation system occupies a region $\Omega \subseteq \mathbb{R}^3$. It is the union of three disjoint parts: the region of solutes (i.e., charged molecules) $\Omega_-$; the region of solvent $\Omega_+$; and the solute-solvent interface or dielectric boundary $\Gamma$, which is a closed surface with possibly multiple components, that separates $\Omega_-$ and $\Omega_+$; cf. Figure 1. We denote by $n$ the unit normal to the boundary $\Gamma$ pointing from $\Omega_-$

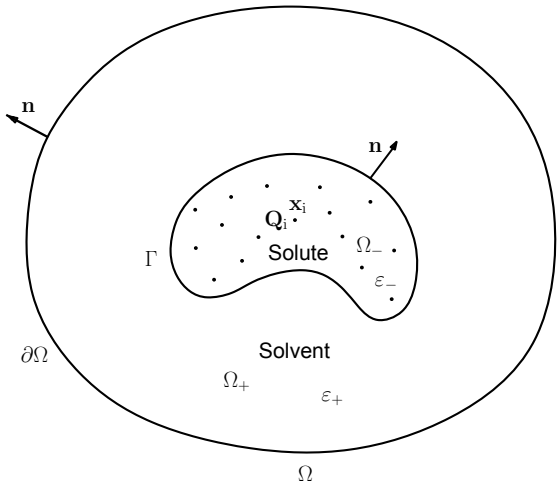


Figure 1: A schematic description of a solvation system with an implicit solvent.

to $\Omega_+$ and also the exterior unit normal to $\partial\Omega$, the boundary of $\Omega$. The solute region $\Omega_-$ contains all the solute atoms that are located at $x_1, \ldots, x_N$ and that carry partial charges $Q_1, \ldots, Q_N$, respectively, with $N \geq 1$ a given integer. The solvent region $\Omega_+$ is the region of ionic solution, same as $\Omega_0$ in (1.1). As above, we assume that there are $M$ species of ions in the solvent with the valence $z_j$, charge $q_j = z_j e$, bulk concentration $c_j^\infty$, and concentration $c_j : \Omega_+ \to [0, \infty)$ for the $j$ ionic species ($j = 1, \ldots, M$). The dielectric coefficients in the solute region $\Omega_-$ and solvent region $\Omega_+$ are denoted by $\varepsilon_-$ and $\varepsilon_+$, respectively. Typically, $\varepsilon_- = 1$ and $\varepsilon_+ = 76 \sim 80$ in the unit of vacuum permittivity. The density of fixed charges is now $\rho = \sum_{i=1}^N Q_i \delta_{x_i}$, where $\delta_{x_i}$ is the Dirac delta function at $x_i$.

Our study consists of three parts. First, we introduce the electrostatic free-energy functional of the ionic concentrations $c = (c_1, \ldots, c_M)$ in the solvent region $\Omega_+$ [9, 26, 37, 48]

$$F_\Gamma[c] = \frac{1}{2} \sum_{i=1}^N Q_i (\psi - \hat{\phi}_{\mathrm{C}})(x_i) + \frac{1}{2} \int_{\Omega_+} \left( \sum_{j=1}^M q_j c_j \right) \psi \, dx$$
$$+ \beta^{-1} \sum_{j=1}^M \int_{\Omega_+} \left\{ c_j \left[ \log(\Lambda^3 c_j) - 1 \right] + c_j^\infty \right\} dx - \sum_{j=1}^M \int_{\Omega_+} \mu_j c_j \, dx, \tag{1.3}$$

where $\Lambda$ is the thermal de Broglie wavelength, $\mu_j$ is the chemical potential for ions of the $j$th species, and $c_j^\infty = \Lambda^{-3} e^{\beta \mu_j}$ ($j = 1, \ldots, M$). In (1.3), $\psi : \Omega \to \mathbb{R}$ is the electrostatic potential. It is the unique weak solution to the boundary-value problem (BVP) of Poisson's equation

$$\nabla \cdot \varepsilon_\Gamma \nabla \psi = - \left( \sum_{i=1}^N Q_i \delta_{x_i} + \chi_+ \sum_{j=1}^M q_j c_j \right) \quad \text{in } \Omega \quad \text{and} \quad \psi = \phi_\infty \quad \text{on } \partial\Omega, \tag{1.4}$$

where the dielectric coefficient $\varepsilon_\Gamma : \Omega \to \mathbb{R}$ is defined by

$$\varepsilon_\Gamma(x) = \begin{cases} \varepsilon_- & \text{if } x \in \Omega_-, \\ \varepsilon_+ & \text{if } x \in \Omega_+, \end{cases} \tag{1.5}$$

$\chi_+ = \chi_{\Omega_+}$ is the characteristic function of $\Omega_+$, and $\phi_\infty$ is a given function on the boundary $\partial\Omega$. The function $\hat{\phi}_{\rm C}$ in (1.3) is the Coulomb potential arising from the point charges $Q_i$ at $x_i$ $(i = 1, \ldots, N)$ in the medium with the dielectric coefficient $\varepsilon_-$, serving as a reference field. It is given by

$$\hat{\phi}_{\rm C}(x) = \sum_{i=1}^{N} \frac{Q_i}{4\pi\varepsilon_-|x - x_i|} \qquad \forall x \in \mathbb{R}^3 \setminus \{x_1, \ldots, x_N\}. \tag{1.6}$$

We note that the ionic concentrations $c_1, \ldots, c_M$ are only defined on the solvent region $\Omega_+$. Implicitly, this assumes that no mobile ions in the solvent are allowed to cross the boundary $\Gamma$ and enter into the solute region $\Omega_-$, an approximation made in a continuum model. The conservation of mass for the mobile ions in the solvent region is enforced through the chemical potentials $\mu_1, \ldots, \mu_M$ that are independent of the solute atomic positions $x_i$ and partial charges $Q_i$ $(i = 1, \ldots, N)$. We also note that in the electrostatic free energy $F_\Gamma[c]$ (cf. (1.3)), the bulk ionic concentrations $c_j^\infty$ and the chemical potentials $\mu_j$, which are related by $c_j^\infty = \Lambda^{-3} e^{\beta\mu_j}$, both appear in integrals over the solvent region $\Omega_+$. Since $\Gamma$ is part of the boundary of $\Omega_+$, variations of the dielectric boundary $\Gamma$ will depend on the chemical potentials [9].

We prove that the functional $F_\Gamma[c]$ has a unique minimizer $c_\Gamma = (c_{\Gamma,1}, \ldots, c_{\Gamma,M})$ in a class of admissible concentrations, and derive the equilibrium conditions $\delta_{c_j} F_\Gamma[c_\Gamma] = 0$ $(j = 1, \ldots, M)$, which lead to the Boltzmann distributions $c_{\Gamma,j} = c_{\Gamma,j}(\psi_\Gamma)$ $(j = 1, \ldots, M)$, where $\psi_\Gamma$ is the corresponding electrostatic potential. We also prove that $\psi_\Gamma$ is the unique solution to the BVP of the nonlinear dielectric-boundary PB equation

$$\nabla \cdot \varepsilon_\Gamma \nabla\psi - \chi_+ B'\left(\psi - \frac{\phi_{\Gamma,\infty}}{2}\right) = -\sum_{i=1}^{N} Q_i \delta_{x_i} \quad \text{in } \Omega \quad \text{and} \quad \psi = \phi_\infty \quad \text{on } \partial\Omega, \tag{1.7}$$

where $B$ is given in (1.2) and $\phi_{\Gamma,\infty} : \Omega \to \mathbb{R}$ is the unique weak solution to the BVP

$$\nabla \cdot \varepsilon_\Gamma \nabla\phi_{\Gamma,\infty} = 0 \quad \text{in } \Omega \quad \text{and} \quad \phi_{\Gamma,\infty} = \phi_\infty \quad \text{on } \partial\Omega; \tag{1.8}$$

cf. Theorem 2.1 and Theorem 2.2. We denote the minimum free energy by

$$E[\Gamma] = \min F_\Gamma[\cdot] = F_\Gamma[c_\Gamma],$$

which depends solely on the dielectric boundary $\Gamma$. We construct a strictly concave functional $G_\Gamma$ of all admissible electrostatic potentials $\psi$ such that the unique solution $\psi_\Gamma$ to the BVP of the dielectric-boundary PE equation is a solution to the Euler–Lagrange equation for $G_\Gamma$, and hence the unique maximizer of $G_\Gamma$. Moreover,

$$E[\Gamma] = \max G_\Gamma[\cdot] = G_\Gamma[\psi_\Gamma];$$

cf. Lemma 3.1 and Theorem 3.1.

Second, we define the (normal component of the) dielectric boundary force to be $-\delta_\Gamma E[\Gamma]$, the negative first variation of the functional $E[\Gamma]$ with respect to the variation of boundary

$\Gamma$. The boundary variation is defined via a smooth vector field. Specifically, let $V : \mathbb{R}^3 \to \mathbb{R}^3$ be a smooth map vanishing outside a small neighborhood of the dielectric boundary $\Gamma$. Let $x = x(t, X)$ be the solution map of the dynamical system defined by [5, 20, 33, 50].

$$\frac{dx(t,X)}{dt} = V(x(t,X)) \quad \forall\, t \in \mathbb{R} \qquad \text{and} \qquad x(0,X) = X \quad \forall X \in \mathbb{R}^3.$$

Such solution maps define a family of transformations $T_t : \mathbb{R}^3 \to \mathbb{R}^3$ ($t \in \mathbb{R}$) by $T_t(X) = x(t, X)$ for any $X \in \mathbb{R}^3$. The variational derivative of the functional $E[\Gamma]$ in the direction of $V : \mathbb{R}^3 \to \mathbb{R}^3$ is defined to be

$$\delta_{\Gamma,V} E[\Gamma] = \frac{d}{dt} E[\Gamma_t(V)]\bigg|_{t=0},$$

if it exists, where $\Gamma_t(V) = \{x(t, X) : X \in \Gamma\}$.

We prove that $\delta_{\Gamma,V} E[\Gamma]$ exists, and is an integral over $\Gamma$ of the product of $V \cdot n$ and some function that is independent of $V$, where $n$ is the unit normal along $\Gamma$, pointing from $\Omega_-$ to $\Omega_+$. This function on $\Gamma$ is identified as the variational derivative (i.e., shape derivative) of $E[\Gamma]$ and is denoted by $\delta_\Gamma E[\Gamma]$. We obtain an explicit formula for $\delta_\Gamma E[\Gamma]$. If the boundary value $\phi_\infty = 0$ on $\Gamma$, then

$$\delta_\Gamma E[\Gamma] = -\frac{1}{2}\left(\frac{1}{\varepsilon_+} - \frac{1}{\varepsilon_-}\right) |\varepsilon_\Gamma \partial_n \psi_\Gamma|^2 + \frac{1}{2}(\varepsilon_+ - \varepsilon_-) \, |\nabla_\Gamma \psi_\Gamma|^2 + B\left(\psi_\Gamma\right), \tag{1.9}$$

where $\psi_\Gamma$ is the unique solution to (1.7), $\varepsilon_\Gamma \partial_n \psi_\Gamma$ is the common value from both sides of $\Gamma$, and $\nabla_\Gamma = (I - n \otimes n)\nabla$ (with $I$ the $3 \times 3$ identity matrix) is the tangential derivative along $\Gamma$. Additional terms arise from a general, inhomogeneous boundary value $\phi_\infty$; cf. Theorem 3.2.

Finally, to describe the electrostatic free energy with point charges and to prove our theorems, we introduce various auxiliary functions that are weak solutions to the BVP of the operator $-\Delta$ or $-\nabla \cdot \varepsilon_\Gamma \nabla$, with or without the point charges $\sum_{i=1}^N Q_i \delta_{x_i}$ and with homogeneous or inhomogeneous Dirichlet boundary conditions. We prove several lemmas, Lemmas 4.1–4.4, showing the continuity and differentiability of those functions with respect to boundary variations. Lemma 4.2 states that the "$\Gamma$-derivative" of the function $\phi_{\Gamma,\infty}$ which is defined in (1.8) is the unique weak solution $\zeta_{\Gamma,V} \in H_0^1(\Omega)$ to the elliptic problem $-\nabla \cdot \varepsilon_\Gamma \nabla \zeta_{\Gamma,V} = f$ in $\Omega$, where $f$ depends on $\phi_{\Gamma,V}$ and $V$. Moreover,

$$\frac{\phi_{\Gamma_t(V),\infty} \circ T_t - \phi_{\Gamma,\infty}}{t} \to \zeta_{\Gamma,V} \quad \text{in } H^1(\Omega) \quad \text{as } t \to 0.$$

(We use the standard notation of Sobolev spaces, such as $H^1(\Omega)$ and $H_0^1(\Omega)$, and other function spaces; cf. [1, 24, 27].) Lemma 4.3 and Lemma 4.4 generalize the result to other $\Gamma$-dependent functions, including the electrostatic potential $\psi_\Gamma$ that is the unique solution to the BVP of the nonlinear dielectric-boundary PB equation (1.7).

We now make several remarks on our results. A nonzero Dirichlet boundary value in (1.4) leads to an extra term $\phi_{\Gamma,\infty}/2$ in the Boltzmann distribution and hence in the PB equation

(1.7). If there are surface charges on the boundary $\partial\Omega$, then one can also use the Neumann boundary condition for the electrostatic potential on $\partial\Omega$. In that case, the electrostatic energy includes a boundary integral term involving the surface charge density; cf. [39, 44].

If we use the homogeneous Dirichlet boundary condition $\phi_\infty = 0$ for the electrostatic potential, then the dielectric boundary force points from the high-dielectric solvent region $\Omega_+$ to the low-dielectric solute region $\Omega_-$; cf. (1.9). Such a macroscopic prediction is consistent with a microscopic picture of molecular forces that charged molecules polarize the surrounding aqueous solvent, which is otherwise electrically neutral, generating an additional electric field that attracts the solvent to the solutes [15]. Since the force points to $\Omega_-$, one expects that no bounded region $\Omega_-$ will minimize the sum of the electrostatic energy and the surface energy [45]. If a small, high-dielectric solvent region is surrounded by the low-dielectric solute molecules (such as a cluster of water molecules buried in a protein), then the competition between the solute-solvent interfacial tension force and the dielectric boundary force results an equilibrium solute-solvent interface, which is however unstable with long-wave perturbations [10, 41]. This may possibly explain why water molecules in proteins are metastable [55, 56]. It remains open to confirm if the dielectric boundary force still points from the high-dielectric solvent region to the low-dielectric solute region for a general inhomogeneous Dirichlet boundary value $\phi_\infty$.

In [6, 7, 54], the authors use the Maxwell stress tensor to define and derive the dielectric boundary force given an electrostatic potential that is determined by the dielectric-boundary PB equation. The shape derivative approach seems first introduced in [38] to define and derive the dielectric boundary force. However, approximations of point charges by smooth functions are made there, and the derivation of the boundary force relies on the underlying variational principle that the electrostatic potential extremizes the PB free-energy functional. This approach is applied to the electrostatic force acting on membranes [47]. Here, we use the direct calculations to derive the boundary force, which is a more general approach.

Our study is closely related to the development of a variational implicit-solvent model (VISM) for biomolecules [21, 22] (cf. also [11–13, 53, 57, 58]). Central in the VISM is an effective free-energy functional of all possible dielectric boundaries that consists mainly of the surface energy of solute molecules, solute-solvent van der Waals interaction energy, and continuum electrostatic free energy. Minimization of the free-energy functional with respect to the dielectric boundary yields optimal solute-solvent interfaces, as well as the solvation free energy. In [40], the authors use the matched asymptotic analysis to derive the sharp-interface limit of a phase-field VISM [51]. In [17], the authors prove the convergence of the free energy and force in the phase-field VISM to their sharp-interface counterparts.

In Section 2, we study the BVP of the nonlinear dielectric-boundary PB equation, and the electrostatic free-energy functionals of ionic concentrations and electrostatic potentials, respectively. In Section 3, we reformulate the minimum electrostatic free energy, define the dielectric boundary force, and present the main formula for such force. In Section 4, we prove several lemmas on the calculus of boundary variations. Finally, in Section 5, we prove the main theorem (Theorem 3.2) of the dielectric boundary force.

# 2 The Poisson–Boltzmann Equation and Free-Energy Functional

## 2.1 Assumptions and Auxiliary Functions

Unless otherwise stated, we assume the following throughout the rest of the paper:

A1. The set $\Omega \subset \mathbb{R}^3$ is non-empty, bounded, open, and connected. The sets $\Omega_- \subset \mathbb{R}^3$ and $\Omega_+ \subset \mathbb{R}^3$ are non-empty, bounded, and open, and satisfy that $\overline{\Omega_-} \subset \Omega$ and $\Omega_+ = \Omega \backslash \overline{\Omega_-}$. The interface $\Gamma = \partial\Omega_- = \overline{\Omega_-} \cap \overline{\Omega_+}$ and the boundary $\partial\Omega$ are of the class $C^3$ and $C^2$, respectively. The unit normal vector at the boundary $\Gamma$ exterior to $\Omega_-$ and that at $\partial\Omega$ exterior to $\Omega$ are both denoted by $n$. The $N$ points $x_1, \ldots, x_N$ for some integer $N \geq 1$ belong to $\Omega_-$; cf. Figure 1. Moreover, there exists a constant $s_0 > 0$ such that

$$\operatorname{dist}(\Gamma, \partial\Omega) \geq s_0; \tag{2.1}$$

A2. All the integer $M \geq 2$, and real numbers $\beta > 0$, $\Lambda > 0$, $Q_i \in \mathbb{R}$ $(1 \leq i \leq N)$, $q_j \neq 0$ and $\mu_j \in \mathbb{R}$ $(1 \leq j \leq M)$, and $\varepsilon_- > 0$ and $\varepsilon_+ > 0$ are given. Moreover, $\varepsilon_- \neq \varepsilon_+$. The parameter $c_j^\infty$ is defined by $c_j^\infty = \Lambda^{-3} e^{\beta\mu_j}$ $(j = 1, \ldots, M)$. The parameters $q_j$ and $c_j^\infty$ $(1 \leq j \leq M)$ satisfy the condition of charge neutrality

$$\sum_{j=1}^{M} q_j c_j^\infty = 0; \tag{2.2}$$

A3. The functions $B : \mathbb{R} \to \mathbb{R}$ and $\varepsilon_\Gamma \in L^\infty(\Omega)$ are defined in (1.2) and (1.5), respectively. The boundary data $\phi_\infty$ is the trace of a given function, also denoted by $\phi_\infty$, in $C^2(\overline{\Omega})$.

Note that $B \in C^\infty(\mathbb{R})$ is strictly convex and $B'(0) = 0$ by the charge neutrality (2.2). Hence, $B(s) > B(0) = 0$ for all $s \neq 0$. The charge neutrality (2.2) implies that there exist some $q_j > 0$ and some $q_k < 0$. Hence, $B(\pm\infty) = \infty$ and $B'(\pm\infty) = \pm\infty$.

We now introduce several auxiliary functions to treat the point-charge singularities, the dielectric discontinuity $\Gamma$, and the inhomogeneous boundary data $\phi_\infty$ on $\partial\Omega$. There are two basic such functions that will directly enter into our results (e.g., the expression of electrostatic free energy and that of the related dielectric boundary force). They are:

- $\hat{\phi}_{\mathrm{C}}$: the Coulomb field defined in (1.6), which is the unique weak solution to $-\varepsilon_- \Delta \hat{\phi}_{\mathrm{C}} = \sum_{i=1}^{N} Q_i \delta_{x_i}$ in $\mathbb{R}^3$ and $\hat{\phi}_{\mathrm{C}} = 0$ at $\infty$;
- $\phi_{\Gamma,\infty}$ : defined as the unique weak solution to the BVP (1.8).

The function $\phi_{\Gamma,\infty}$ is determined by $\phi_{\Gamma,\infty} \in H^1(\Omega)$, $\phi_{\Gamma,\infty} = \phi_\infty$ on $\partial\Omega$, and

$$\int_\Omega \varepsilon_\Gamma \nabla\phi_{\Gamma,\infty} \cdot \nabla\eta \, dx = 0 \qquad \forall \eta \in H_0^1(\Omega). \tag{2.3}$$

By the regularity theory, we have

$$\phi_{\Gamma,\infty} \in W^{1,\infty}(\Omega) \quad \text{and} \quad \phi_{\Gamma,\infty}|_{\Omega_s} \in C^\infty(\Omega_s) \cap H^2(\Omega_s) \quad \text{for } s = -, +. \tag{2.4}$$

Moreover, there exists a constant $C = C(\Omega, \varepsilon_+, \varepsilon_-, \phi_\infty) > 0$, independent of $\Gamma$, such that

$$\|\phi_{\Gamma,\infty}\|_{H^1(\Omega)} + \|\phi_{\Gamma,\infty}\|_{L^\infty(\Omega)} \le C. \tag{2.5}$$

See [42] (Corollary 1.3) for the $W^{1,\infty}$-regularity, which implies that $\phi_{\Gamma,\infty} \in C(\overline{\Omega})$. The $H^1$-estimate in (2.5) is standard; it can be derived simply by setting $\eta = \phi_{\Gamma,\infty} - \phi_\infty$ in (2.3). See [27] (Theorem 8.29) for the global Hölder estimate which implies the global $L^\infty$ estimate in (2.5). (Note that the global $W^{1,\infty}$-estimate is established in [42] but the constant $C$ may depend on the smoothness of $\Gamma$.) For the piecewise $H^2$-regularity, see [35] (Section 16 of Chapter 3) and [30, 31]. By (2.3), we have

$$\Delta\phi_{\Gamma,\infty} = 0 \qquad \text{in } \Omega_- \cup \Omega_+. \tag{2.6}$$

This implies the piecewise $C^\infty$-regularity in (2.4). By the fact that $\phi_{\Gamma,\infty} \in C(\overline{\Omega}) \cap H^1(\Omega)$ and by routine calculations using (2.3) and the Divergence Theorem, we have [37]

$$[\![\phi_{\Gamma,\infty}]\!]_\Gamma = 0 \qquad \text{and} \qquad [\![\varepsilon_\Gamma \partial_n \phi_{\Gamma,\infty}]\!]_\Gamma = 0, \tag{2.7}$$

where the jump $[\![\cdot]\!]_\Gamma$ is defined for any function $u$ on $\Omega$ that has the trace on $\Gamma$ by

$$[\![u]\!]_\Gamma = u|_{\Omega_+} - u|_{\Omega_-} \qquad \text{on } \Gamma. \tag{2.8}$$

We now introduce some more auxiliary functions as weak solutions to BVPs; these functions are only used in proofs of some lemmas or theorems:

$$\hat{\phi}_0: \qquad -\varepsilon_-\Delta\hat{\phi}_0 = \sum_{i=1}^N Q_i\delta_{x_i} \quad \text{in } \Omega \qquad \text{and} \qquad \hat{\phi}_0 = 0 \quad \text{on } \partial\Omega; \tag{2.9}$$

$$\hat{\phi}_\infty: \qquad -\varepsilon_-\Delta\hat{\phi}_\infty = \sum_{i=1}^N Q_i\delta_{x_i} \quad \text{in } \Omega \qquad \text{and} \qquad \hat{\phi}_\infty = \phi_\infty \quad \text{on } \partial\Omega; \tag{2.10}$$

$$\hat{\phi}_{\Gamma,\infty}: \qquad -\nabla\cdot\varepsilon_\Gamma\nabla\hat{\phi}_{\Gamma,\infty} = \sum_{i=1}^N Q_i\delta_{x_i} \quad \text{in } \Omega \qquad \text{and} \qquad \hat{\phi}_{\Gamma,\infty} = \phi_\infty \quad \text{on } \partial\Omega. \tag{2.11}$$

Note that a hat means the right-hand side is given by $\sum_{i=1}^N Q_i\delta_{x_i}$. A subscript $\Gamma$ corresponds to $-\nabla\cdot\varepsilon_\Gamma\nabla$, while no subscript $\Gamma$ referring to $-\varepsilon_-\Delta$. A subscript $\infty$ or 0 corresponds to boundary value $\phi_\infty$ or 0, respectively.

The function $\hat{\phi} = \hat{\phi}_{\rm C}$, or $\hat{\phi}_0$, or $\hat{\phi}_\infty$ satisfies that $\hat{\phi} \in \hat{\phi}_{\rm C} + H^1(\Omega)$. It is uniquely determined by its boundary value on $\partial\Omega$, and

$$\int_\Omega \varepsilon_-\nabla\hat{\phi}\cdot\nabla\eta\,dx = \sum_{i=1}^N Q_i\eta(x_i) \qquad \forall\eta \in C_{\rm c}^1(\Omega), \tag{2.12}$$

where $C_{\rm c}^1(\Omega)$ denotes the class of $C^1(\Omega)$-functions that are compactly supported in $\Omega$. Clearly, we can modify the value of $\hat{\phi}$ on a set of zero Lebesgue measure, if necessary, so that $\hat{\phi}$ is a

$C^\infty$-function in $\Omega\setminus\{x_1,\dots,x_N\}$. Moreover, $\Delta\hat\phi = 0$ in $\Omega\setminus\{x_1,\dots,x_N\}$ and $\Delta(\hat\phi-\hat\phi_{\rm C}) = 0$ in $\Omega$. Since $\partial\Omega$ is $C^2$ and $\phi_\infty\in C^2(\overline\Omega)$, we have $\hat\phi-\hat\phi_{\rm C}\in H^2(\Omega)$; cf. Chapter 8 in [27]. Therefore, $\hat\phi\in\hat\phi_{\rm C}+H^2(\Omega)\cap C^\infty(\Omega)\subset W^{1,1}(\Omega)$.

We remark that $\eta\in C^1_{\rm c}(\Omega)$ in (2.12) can be replaced by $\eta\in H^1_0(\Omega)$ with $\eta|_{\Omega_-}\in C^1(\Omega_-)$. To see this, we first note that (2.12) holds true if $\hat\phi$ is replaced by $\hat\phi_{\rm C}$ (cf. (1.6)). Thus,

$$\int_\Omega \varepsilon_-\nabla(\hat\phi-\hat\phi_{\rm C})\cdot\nabla\eta\,dx = 0 \qquad \forall\eta\in H^1_0(\Omega),$$

as $\hat\phi-\hat\phi_{\rm C}\in H^1(\Omega)$ and $C^1_{\rm c}(\Omega)$ is dense in $H^1(\Omega)$. If $\eta\in H^1_0(\Omega)$ also satisfies $\eta|_{\Omega_-}\in C^1(\Omega_-)$, then $\nabla\hat\phi_{\rm C}\cdot\nabla\eta$, hence $\nabla\hat\phi\cdot\nabla\eta$, is integrable in $\Omega$. Moreover,

$$\int_\Omega \varepsilon_-\nabla\hat\phi\cdot\nabla\eta\,dx = \int_\Omega \varepsilon_-\nabla\hat\phi_{\rm C}\cdot\nabla\eta\,dx = \sum_{i=1}^N Q_i\eta(x_i),$$

where the second equality follows from straight forward calculations using (1.6).

The function $\hat\phi_{\Gamma,\infty}$ defined in (2.11) belongs to $\hat\phi_{\rm C}+H^1(\Omega)$. It is uniquely determined by its boundary value on $\partial\Omega$ and

$$\int_\Omega \varepsilon_\Gamma\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\eta\,dx = \sum_{i=1}^N Q_i\eta(x_i) \qquad \forall\eta\in C^1_{\rm c}(\Omega); \tag{2.13}$$

cf. [23, 43]. If $\hat\phi=\hat\phi_{\rm C}$ (the Coulomb field), or $\hat\phi_0$ defined in (2.9) , or $\hat\phi_\infty$ defined in (2.10), then (2.13) is equivalent to

$$\begin{aligned}\int_\Omega \varepsilon_\Gamma\nabla(\hat\phi_{\Gamma,\infty}-\hat\phi)\cdot\nabla\eta\,dx &= -(\varepsilon_+-\varepsilon_-)\int_{\Omega_+}\nabla\hat\phi\cdot\nabla\eta\,dx\\ &= (\varepsilon_+-\varepsilon_-)\int_\Gamma \partial_n\hat\phi\,\eta\,dS \qquad \forall\eta\in H^1_0(\Omega),\end{aligned} \tag{2.14}$$

where the unit normal $n$ at $\Gamma$ points from $\Omega_-$ to $\Omega_+$. If $\eta\in H^1_0(\Omega)$ satisfies $\eta|_{\Omega_-}\in C^1(\Omega_-)$, then it follows from (2.14) and (2.13) that

$$\int_\Omega \varepsilon_\Gamma\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\eta\,dx = \int_\Omega \varepsilon_-\nabla\hat\phi\cdot\nabla\eta\,dx = \sum_{i=1}^N Q_i\eta(x_i).$$

Therefore, we can replace $\eta\in C^1_{\rm c}(\Omega)$ in (2.13) by $\eta\in H^1_0(\Omega)$ that satisfies $\eta|_{\Omega_-}\in C^1(\Omega_-)$.

By (2.13) and (2.14), we have

$$\Delta(\hat\phi_{\Gamma,\infty}-\hat\phi) = 0 \quad\text{in } \Omega_- \quad\text{and}\quad \Delta\hat\phi_{\Gamma,\infty} = 0 \quad\text{in } (\Omega_-\setminus\{x_1,\dots,x_N\})\cup\Omega_+, \tag{2.15}$$

$$[\![\hat\phi_{\Gamma,\infty}]\!]_\Gamma = 0 \qquad\text{and}\qquad [\![\varepsilon_\Gamma\partial_n\hat\phi_{\Gamma,\infty}]\!]_\Gamma = 0 \qquad\text{on } \Gamma. \tag{2.16}$$

Moreover, it follows from the elliptic regularity theory [23, 27, 30, 31, 35, 42] that

$$\hat{\phi}_{\Gamma,\infty} - \hat{\phi} \in W^{1,\infty}(\Omega) \quad \text{and} \quad (\hat{\phi}_{\Gamma,\infty} - \hat{\phi})|_{\Omega_s} \in C^\infty(\Omega_s) \cap H^2(\Omega_s) \quad s = +, -, \tag{2.17}$$

$$\|\hat{\phi}_{\Gamma,\infty} - \hat{\phi}\|_{H^1(\Omega)} + \|\hat{\phi}_{\Gamma,\infty} - \hat{\phi}\|_{L^\infty(\Omega)} \leq C, \tag{2.18}$$

where the constant $C > 0$ does not depend on $\Gamma$. These results (2.17) and (2.18) follow from the same arguments used above with $\eta \in C_c^1(\Omega)$ so chosen that supp $(\eta)$ is in a neighborhood of $\Gamma$ that excludes the singularities $x_i$ $(i = 1, \ldots, N)$.

We end this subsection by defining some linear operator and a class of functions. For any $g \in H^{-1}(\Omega)$, let $L_\Gamma g \in H_0^1(\Omega)$ be the unique weak solution (defined using test functions in $H_0^1(\Omega)$) to the BVP

$$\nabla \cdot \varepsilon_\Gamma \nabla L_\Gamma g = -g \quad \text{in } \Omega \quad \text{and} \quad L_\Gamma g = 0 \quad \text{on } \partial\Omega. \tag{2.19}$$

This defines a linear, continuous, and self-adjoint operator $L_\Gamma : H^{-1}(\Omega) \to H_0^1(\Omega)$. The map

$$g \mapsto \|g\|_{L_\Gamma} := \sqrt{\langle g, L_\Gamma g \rangle_{H^{-1}(\Omega), H_0^1(\Omega)}} = \left[ \int_\Omega \varepsilon_\Gamma |\nabla (L_\Gamma g)|^2 dx \right]^{1/2} \tag{2.20}$$

defines a norm on $H^{-1}(\Omega)$ which is equivalent to the $H^{-1}(\Omega)$-norm.

Let $g \in L^1(\Omega)$ and assume that

$$\sup \left\{ \int_\Omega gu \, dx : u \in H_0^1(\Omega) \cap L^\infty(\Omega) \text{ and } \|u\|_{H^1(\Omega)} = 1 \right\} < \infty. \tag{2.21}$$

Define $T_g : H_0^1(\Omega) \cap L^\infty(\Omega) \to \mathbb{R}$ by

$$T_g[u] = \int_\Omega gu \, dx \qquad \forall u \in H_0^1(\Omega) \cap L^\infty(\Omega).$$

It follows from (2.21) that $T_g$ is a bounded (with respect to the $H_0^1(\Omega)$-norm) linear functional on $H_0^1(\Omega) \cap L^\infty(\Omega)$, a subspace of $H_0^1(\Omega)$. Since this subspace is dense in $H_0^1(\Omega)$, we can extend $T_g$ uniquely to a bounded linear functional, still denoted by $T_g$, on the entire space $H_0^1(\Omega)$, i.e., $T_g \in H^{-1}(\Omega)$. For convenience, we shall write $g \in L^1(\Omega) \cap H^{-1}(\Omega)$ to mean that $g \in L^1(\Omega)$, (2.21) holds true, and $g$ is identified as $T_g \in H^{-1}(\Omega)$.

## 2.2 The Poisson–Boltzmann Equation

**Definition 2.1.** *A function $\psi \in \hat{\phi}_C + H^1(\Omega)$ is a weak solution to the BVP of the dielectric-boundary PB equation* (1.7)*, if $\psi = \phi_\infty$ on $\partial\Omega$, $\chi_+ B'(\psi - \phi_{\Gamma,\infty}/2) \in L^1(\Omega) \cap H^{-1}(\Omega)$, and*

$$\int_\Omega \left[ \varepsilon_\Gamma \nabla\psi \cdot \nabla\eta + \chi_+ B' \left( \psi - \frac{\phi_{\Gamma,\infty}}{2} \right) \eta \right] dx = \sum_{i=1}^N Q_i \eta(x_i) \qquad \forall \eta \in C_c^1(\Omega). \tag{2.22}$$

Note that we can replace $\eta \in C_c^1(\Omega)$ in (2.22) by $\eta \in H_0^1(\Omega)$ that satisfies $\eta|_{\Omega_-} \in C^1(\Omega_-)$; cf. the remark below (2.14). The theorem below provides the existence and uniqueness of the solution to the BVP of the dielectric-boundary PB equation, and an equivalent formulation of such a BVP. These results are essentially proved in [38]. Here we sketch the proof and add some points that are not included in the previous proof due to some differences between the current and previous statements. Note that $\hat{\phi}_C + H^1(\Omega) = \hat{\phi}_{\Gamma,\infty} + H^1(\Omega)$. So, we can replace $\hat{\phi}_C$ by $\hat{\phi}_{\Gamma,\infty}$ in the above definition.

**Theorem 2.1.** (1) *There exists a unique weak solution $\psi_\Gamma \in \hat{\phi}_{\Gamma,\infty} + H_0^1(\Omega)$ of the BVP of the dielectric-boundary PB equation* (1.7)*. Moreover, $\psi_\Gamma - \hat{\phi}_{\Gamma,\infty} \in C(\overline{\Omega}) \cap W^{1,\infty}(\Omega)$, $(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})|_{\Omega_-} \in C^\infty(\Omega_-) \cap H^2(\Omega_-)$, and $\psi_\Gamma|_{\Omega_+} \in C^\infty(\Omega_+) \cap H^2(\Omega_+)$. Further, there exists a constant $C > 0$ independent of $\Gamma$ such that*

$$\|\psi_\Gamma - \hat{\phi}_{\Gamma,\infty}\|_{H^1(\Omega)} + \|\psi_\Gamma - \hat{\phi}_{\Gamma,\infty}\|_{L^\infty(\Omega)} \le C. \tag{2.23}$$

(2) *A function $\psi \in \hat{\phi}_{\Gamma,\infty} + H^1(\Omega)$ with $\chi_+ B'(\psi - \phi_{\Gamma,\infty}/2) \in L^1(\Omega) \cap H^{-1}(\Omega)$ is the weak solution to the BVP of the dielectric-boundary PB equation* (1.7) *if and only if it is the unique solution to the following problem:*

$$\begin{cases} \Delta(\psi - \hat{\phi}_C) = 0 & \text{in } \Omega_-, \\ \varepsilon_+ \Delta\psi - B'\left(\psi - \dfrac{\phi_{\Gamma,\infty}}{2}\right) = 0 & \text{in } \Omega_+, \\ [\![\psi]\!]_\Gamma = 0 \quad \text{and} \quad [\![\varepsilon_\Gamma \partial_n \psi]\!]_\Gamma = 0 & \text{on } \Gamma, \\ \psi = \psi_\infty & \text{on } \partial\Omega. \end{cases} \tag{2.24}$$

*Proof.* (1) With $u = \psi - \hat{\phi}_{\Gamma,\infty}$ and by (2.13) and (2.22), it is equivalent to show that there exists a unique $u_\Gamma \in H_0^1(\Omega)$ such that $\chi_+ B'(u_\Gamma + \hat{\phi}_{\Gamma,\infty} - \phi_{\Gamma,\infty}/2) \in L^1(\Omega) \cap H^{-1}(\Omega)$, and

$$\int_\Omega \left[\varepsilon_\Gamma \nabla u_\Gamma \cdot \nabla\eta + \chi_+ B'\left(u_\Gamma + \hat{\phi}_{\Gamma,\infty} - \frac{\phi_{\Gamma,\infty}}{2}\right)\eta\right] dx = 0 \qquad \forall \eta \in H_0^1(\Omega). \tag{2.25}$$

Define

$$I[u] = \int_\Omega \left[\frac{\varepsilon_\Gamma}{2}|\nabla u|^2 + \chi_+ B\left(u + \hat{\phi}_{\Gamma,\infty} - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dx \qquad \forall u \in H_0^1(\Omega).$$

Since $B \ge 0$ and $B$ is convex, we can use the direct method in the calculus of variations to obtain a unique minimizer $u_\Gamma \in H_0^1(\Omega)$ of the functional $I : H_0^1(\Omega) \to [0, \infty]$. Moreover, comparing the values $I[u_\Gamma]$ and $I[u_{\Gamma,\lambda}]$ for any constant $\lambda > 0$ large enough, where $u_{\Gamma,\lambda} = u_\Gamma$ if $|u_\Gamma| \le \lambda$ and $u_{\Gamma,\lambda} = \lambda \operatorname{sign}(u_\Gamma)$ otherwise, we have by the convexity of $B$ that $u_\Gamma = u_{\Gamma,\lambda}$ a.e. $\Omega$ for some $\lambda$ independent on $\Gamma$. Hence, $u_\Gamma \in L^\infty(\Omega)$, and $\|u_\Gamma\|_{L^\infty(\Omega)} \le C$ for some constant $C > 0$ independent of $\Gamma$; cf. [38]. This allows the use of the Lebesgue Dominated Convergence Theorem in the routine calculations of $(d/dt)|_{t=0} I[u_\Gamma + t\eta] = 0$ for any $\eta \in C_c^1(\Omega)$ to obtain the equation in (2.25). Since $C_c^1(\Omega)$ is dense in $H_0^1(\Omega)$, (2.25) holds true. The convexity of $B$ now implies that $u_\Gamma$ is the unique solution as desired.

The regularity of the solution $\psi_\Gamma$ follows from the elliptic regularity theory [14, 23, 27, 30, 31, 35, 42], with the same argument above for the regularity of the function $\hat{\phi}_{\Gamma,\infty}$; cf. (2.17) and (2.18). Note that the piecewise $C^\infty$ smoothness follows from a usual bootstrapping method.

(2) This part of the proof is the same as that given in [37]. □

## 2.3 Electrostatic Free-Energy Functional of Ionic Concentrations

We define

$$\mathcal{X} = \left\{ (c_1, \ldots, c_M) \in L^1(\Omega, \mathbb{R}^M) : c_j = 0 \text{ a.e. } \Omega_- \text{ for } j = 1, \ldots, M \text{ and } \sum_{j=1}^{M} q_j c_j \in H^{-1}(\Omega) \right\},$$

$$\mathcal{X}_+ = \left\{ (c_1, \ldots, c_M) \in \mathcal{X} : c_j \geq 0 \text{ a.e. } \Omega_+ \text{ for } j = 1, \ldots, M \right\}.$$

Here, for any $g \in L^1(\Omega)$, we define $g \in L^1(\Omega) \cap H^{-1}(\Omega)$ by (2.21). The space $\mathcal{X}$ is a Banach space equipped with the norm $\|c\|_{\mathcal{X}} = \sum_{j=1}^{M} \|c_j\|_{L^1(\Omega)} + \| \sum_{j=1}^{M} q_j c_j \|_{H^{-1}(\Omega)}$. Moreover, $\mathcal{X}_+$ is a convex and closed subset of $\mathcal{X}$. For any $c = (c_1, \ldots, c_M) \in \mathcal{X}$, standard arguments (cf. [23, 24, 27, 43]) imply that there exists a unique weak solution $\psi$ to the BVP (1.4), defined by $\psi \in \hat{\phi}_{\mathrm{C}} + H^1(\Omega)$, $\psi = \phi_\infty$ on $\partial\Omega$, and

$$\int_\Omega \varepsilon_\Gamma \nabla\psi \cdot \nabla\eta \, dx = \sum_{i=1}^{N} Q_i \eta(x_i) + \int_{\Omega_+} \left( \sum_{j=1}^{M} q_j c_j \right) \eta \, dx \qquad \forall \eta \in C_{\mathrm{c}}^1(\Omega), \tag{2.26}$$

Equivalently, if $\hat{\phi} \in \hat{\phi}_{\mathrm{C}} + H^1(\Omega)$ satisfies (2.12), then

$$\int_\Omega \varepsilon_\Gamma \nabla(\psi - \hat{\phi}) \cdot \nabla\eta \, dx = \int_{\Omega_+} \left[ (\varepsilon_- - \varepsilon_+) \nabla\hat{\phi} \cdot \nabla\eta + \left( \sum_{j=1}^{M} q_j c_j \right) \eta \right] dx \qquad \forall \eta \in H_0^1(\Omega).$$

Clearly, $\psi - \hat{\phi}$ is harmonic in $\Omega_-$. Moreover, it follows from the definition of $\hat{\phi}_{\Gamma,\infty}$ (cf. (2.13)) and $L_\Gamma$ (cf. (2.19)) that

$$\psi = \hat{\phi}_{\Gamma,\infty} + L_\Gamma \left( \sum_{j=1}^{M} q_j c_j \right). \tag{2.27}$$

Since the function $s \mapsto s \log s$ $(s \geq 0)$ is bounded below and $\Omega$ is bounded, $F_\Gamma[c] > -\infty$ for any $c \in \mathcal{X}_+$, where $F_\Gamma[c]$ is defined in (1.3).

**Theorem 2.2.** *Let $\psi_\Gamma$ be the unique weak solution to the BVP of the dielectric-boundary PB equation* (1.7). *For each $j \in \{1, \ldots, M\}$, define $c_{\Gamma,j} : \Omega \to [0, \infty)$ by*

$$c_{\Gamma,j}(x) = \begin{cases} 0 & \text{if } x \in \overline{\Omega_-}, \\ c_j^\infty e^{-\beta q_j [\psi_\Gamma(x) - \phi_{\Gamma,\infty}(x)/2]} & \text{if } x \in \Omega_+. \end{cases} \tag{2.28}$$

*Then* $c_\Gamma := (c_{\Gamma,1}, \ldots, c_{\Gamma,M}) \in \mathcal{X}_+$ *and* $\psi_\Gamma$ *is the electrostatic potential corresponding to* $c_\Gamma$*, i.e., the unique weak solution to* (1.4) *with* $c_j$ *replaced by* $c_{\Gamma,j}$ $(j = 1, \ldots, M)$*. Moreover,* $c_\Gamma$ *is the unique minimizer of the functional* $F_\Gamma : \mathcal{X}_+ \to (-\infty, \infty]$ *defined in* (1.3)*, and*

$$F_\Gamma[c_\Gamma] = \frac{1}{2}\sum_{i=1}^{N} Q_i(\psi_\Gamma - \hat{\phi}_{\mathrm{C}})(x_i) + \int_{\Omega_+} \left[\frac{1}{2}\left(\psi_\Gamma - \phi_{\Gamma,\infty}\right) B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) - B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dx. \tag{2.29}$$

*Proof.* By the properties of $\psi_\Gamma$ (cf. Theorem 2.1) and $\phi_{\Gamma,\infty}$ (cf. (2.5)), we have $c_\Gamma \in \mathcal{X}_+$. If we replace $c_j$ in (1.4) by $c_{\Gamma,j}$ defined in (2.28) and note the definition of $B$ in (1.2), we get exactly the PB equation (1.7). Therefore, the unique solution $\psi_\Gamma$ to the BVP of the PB equation (1.7) is also the unique solution to the BVP of Poisson's equation (1.4) corresponding to $c_\Gamma$.

We now prove that $c_\Gamma$ is the unique minimizer of $F_\Gamma : \mathcal{X}_+ \to (-\infty, \infty]$. To do so, we first re-write the functional $F_\Gamma$. Let $c = (c_1, \ldots, c_M) \in \mathcal{X}_+$ and let $\psi \in \hat{\phi}_{\mathrm{C}} + H^1(\Omega)$ be the corresponding electrostatic potential, i.e., the weak solution to (1.4) defined in (2.26). Denote $f = \sum_{j=1}^{M} q_j c_j$. Since $f = 0$ a.e. in $\Omega_-$, we have by the definition of $L_\Gamma$ (cf. (2.19)) that $L_\Gamma f$ is harmonic in $\Omega_-$. Moreover,

$$\sum_{i=1}^{N} Q_i \left(L_\Gamma f\right)(x_i) = \int_{\Omega_+} (\hat{\phi}_{\Gamma,\infty} - \phi_{\Gamma,\infty}) f \, dx;$$

cf. Lemma 3.2 in [37] (where $L/(4\pi)$ and $G/(4\pi)$ are our $L_\Gamma$ and $\hat{\phi}_{\Gamma,\infty} - \phi_{\Gamma,\infty}$ here, respectively). This, together with (2.27) and the fact that all $\psi - \hat{\phi}_{\mathrm{C}}$, $\hat{\phi}_{\Gamma,\infty} - \hat{\phi}$, and $L_\Gamma f$ are harmonic in $\Omega_-$, implies that

$$\begin{aligned}\sum_{i=1}^{N} Q_i(\psi - \hat{\phi}_{\mathrm{C}})(x_i) &= \sum_{i=1}^{N} Q_i(L_\Gamma f)(x_i) + \sum_{i=1}^{N} Q_i(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_{\mathrm{C}})(x_i) \\ &= \int_{\Omega_+} (\hat{\phi}_{\Gamma,\infty} - \phi_{\Gamma,\infty}) \left(\sum_{j=1}^{M} q_j c_j\right) dx + \sum_{i=1}^{N} Q_i(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_{\mathrm{C}})(x_i).\end{aligned}$$

With this and (2.27), we can rewrite $F_\Gamma[c]$ (1.3) as

$$F_\Gamma[c] = \int_{\Omega_+} \left[\frac{1}{2}\left(\sum_{j=1}^{M} q_j c_j\right) L_\Gamma\left(\sum_{j=1}^{M} q_j c_j\right) + \sum_{j=1}^{M} \left(\beta^{-1} c_j \log c_j + \alpha_j c_j\right)\right] dx + E_{0,\Gamma}, \tag{2.30}$$

where all $\alpha_j = \alpha_j(x)$ $(j = 1, \ldots, M)$ and $E_{0,\Gamma}$ are independent of $c$, given by

$$\alpha_j(x) = q_j\left[\hat{\phi}_{\Gamma,\infty}(x) - \frac{1}{2}\phi_{\Gamma,\infty}(x)\right] + \beta^{-1}\left(3\log\Lambda - 1\right) - \mu_j \quad \forall x \in \Omega,\ j = 1, \ldots, M, \tag{2.31}$$

$$E_{0,\Gamma} = \frac{1}{2}\sum_{i=1}^{N} Q_i(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_{\rm C})(x_i) + \beta^{-1}|\Omega_+|\sum_{j=1}^{M} c_j^{\infty}.$$

Here and below, we denote by $|A|$ the Lebesgue measure of $A$ when no confusion arises.

We now compare $F_\Gamma[c]$ and $F_\Gamma[c_\Gamma]$. By Taylor's expansion, we have for any $s, t \in (0,\infty)$ that

$$s\log s - t\log t = (1+\log t)(s-t) + \frac{1}{2r}(s-t)^2 \geq (1+\log t)(s-t),$$

where $r$ is in between $s$ and $t$. Consequently, by (2.30) and the fact that $L_\Gamma$ is self-adjoint, we have

$$\begin{aligned} F_\Gamma[c] - F_\Gamma[c_\Gamma] = &\int_\Omega \frac{1}{2}\left(\sum_{j=1}^{M} q_j(c_j - c_{\Gamma,j})\right) L_\Gamma\left(\sum_{j=1}^{M} q_j(c_j - c_{\Gamma,j})\right) dx \\ &+ \int_\Omega \left(\sum_{j=1}^{M} q_j(c_j - c_{\Gamma,j})\right) L_\Gamma\left(\sum_{k=1}^{M} q_k c_{\Gamma,k}\right) dx \\ &+ \beta^{-1}\sum_{j=1}^{M}\int_\Omega (c_j\log c_j - c_{\Gamma,j}\log c_{\Gamma,j})\, dx + \sum_{j=1}^{M}\int_\Omega (c_j - c_{\Gamma,j})\alpha_j\, dx \\ \geq &\sum_{j=1}^{M}\int_\Omega (c_j - c_{\Gamma,j})\left[q_j L_\Gamma\left(\sum_{k=1}^{M} q_k c_{\Gamma,k}\right) + \beta^{-1}(1+\log c_{\Gamma,j}) + \alpha_j\right] dx. \end{aligned}$$

It follows from the fact that $c_j^\infty = \Lambda^{-3}e^{\beta\mu_j}$ (cf. the assumption (A2)), (2.27), (2.28), and (2.31) that the quantity inside the brackets in the above integral vanishes. Thus, $F[c] \geq F[c_\Gamma]$. Hence, $c_\Gamma$ is a minimizer of $F_\Gamma : \mathcal{X}_+ \to (-\infty,\infty]$. Since $F_\Gamma$ is convex, and in particular, $s \mapsto s\log s$ is strictly convex on $(0,\infty)$, the minimizer of $F_\Gamma$ is unique; cf. [37].

Finally, we obtain (2.29) from (1.3) (with $\psi_\Gamma$ and $c_\Gamma$ replacing $\psi$ and $c$, respectively), (1.2), and (2.28). □

# 3 Dielectric Boundary Force

## 3.1 Electrostatic Free Energy of a Dielectric Boundary

Let $\Gamma$ be a given dielectric boundary as described in the assumption A1 in Subsection 2.1. We denote by

$$E[\Gamma] = \min_{c\in\mathcal{X}_+} F_\Gamma[c], \tag{3.1}$$

the minimum electrostatic free energy given in Theorem 2.2 (cf. (2.29)). Let $\psi_\Gamma \in \hat{\phi}_{\rm C} + H^1(\Omega)$ be the corresponding solution to the BVP of PB equation (1.7). Recall that all the functions $\hat{\phi}_{\rm C}$, $\hat{\phi}_0$, $\hat{\phi}_\infty$, $\phi_{\Gamma,\infty}$, and $\hat{\phi}_{\Gamma,\infty}$ are defined in Subsection 2.1, with a hat corresponding to $\sum_{i=1}^{N} Q_i\delta_{x_i}$, $\Gamma$ to $-\nabla\cdot\varepsilon_\Gamma\nabla$ and otherwise to $-\varepsilon_-\Delta$, and $\infty$ or 0 to the boundary value $\phi_\infty$ or 0, respectively.

**Lemma 3.1.** *We have*

$$E[\Gamma] = -\int_\Omega \frac{\varepsilon_\Gamma}{2}|\nabla(\psi_\Gamma - \hat\phi_{\Gamma,\infty})|^2 dx - \int_{\Omega_+} B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) dx$$
$$+ \frac{\varepsilon_- - \varepsilon_+}{2}\int_{\Omega_+} \nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\hat\phi_0\, dx + \frac{1}{2}\sum_{i=1}^N Q_i(\hat\phi_\infty - \hat\phi_{\rm C})(x_i). \tag{3.2}$$

*Proof.* We first prove an elementary identity. Let $u \in C^2(\Omega_-)\cap C^1(\overline{\Omega_-})$ be such that $\Delta u = 0$ in $\Omega_-$. Let $v \in \hat\phi_{\rm C} + H^1(\Omega_-)\cap C(\overline{\Omega_-})$, in particular, $v = \hat\phi_{\rm C}$, $\hat\phi_0$, $\hat\phi_\infty$, or $\hat\phi_{\Gamma,\infty}$ (restricted onto $\Omega_-$). Denote $B_\alpha = \cup_{i=1}^N B(x_i,\alpha)$ for $0<\alpha\ll 1$ and $\nu$ the unit normal at $\partial B(\alpha) = \cup_{i=1}^N \partial B(x_i,\alpha)$, pointing toward $x_i$ $(i=1,\dots,N)$. Since the unit normal $n$ at $\Gamma$ points from $\Omega_-$ to $\Omega_+$, and since $v = \hat\phi_{\rm C} + \hat v$ for some $\hat v \in H^1(\Omega_-)\cap C(\overline{\Omega_-})$ and $\hat\phi_{\rm C}$ is given in (1.6), we have

$$\begin{aligned}\int_{\Omega_-}\nabla u\cdot\nabla v\, dx &= \lim_{\alpha\to 0^+}\int_{\Omega_-\setminus B_\alpha}\nabla u\cdot\nabla v\, dx\\ &= \int_\Gamma \partial_n u\, v\, dS + \lim_{\alpha\to 0^+}\sum_{i=1}^N\int_{\partial B(x_i,\alpha)}\partial_\nu u\, v\, dS\\ &= \int_\Gamma \partial_n u\, v\, dS.\end{aligned} \tag{3.3}$$

Denoting now $W = (1/2)\sum_{i=1}^N Q_i(\hat\phi_\infty - \hat\phi_{\rm C})(x_i)$, we have by (3.1) and (2.29) that

$$E[\Gamma] = \frac{1}{2}\sum_{i=1}^N Q_i(\hat\phi_{\Gamma,\infty} - \hat\phi_\infty)(x_i) + \frac{1}{2}\sum_{i=1}^N Q_i(\psi_\Gamma - \hat\phi_{\Gamma,\infty})(x_i)$$
$$+ \int_{\Omega_+}\left[\frac{1}{2}(\psi_\Gamma - \phi_{\Gamma,\infty})B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) - B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dx + W. \tag{3.4}$$

We first consider the first term in (3.4). Note that the unit vector $n$ normal to $\Gamma$ points from $\Omega_-$ to $\Omega_+$. Denoting $u^s = u|_{\Omega_s}$ $(s=+,-)$, we have by Green's formula that

$$\begin{aligned}&\frac{1}{2}\sum_{i=1}^N Q_i(\hat\phi_{\Gamma,\infty} - \hat\phi_\infty)(x_i)\\ &= \int_\Omega \frac{\varepsilon_-}{2}\nabla\hat\phi_0\cdot\nabla(\hat\phi_{\Gamma,\infty} - \hat\phi_\infty)\, dx \qquad [\text{by (2.12) with } \hat\phi = \hat\phi_0 \text{ and } \eta = \hat\phi_{\Gamma,\infty} - \hat\phi_\infty]\\ &= \int_{\Omega_-} \frac{\varepsilon_-}{2}\nabla\hat\phi_0\cdot\nabla(\hat\phi_{\Gamma,\infty} - \hat\phi_\infty)\, dx + \int_{\Omega_+} \frac{\varepsilon_-}{2}\nabla\hat\phi_0\cdot\nabla(\hat\phi_{\Gamma,\infty} - \hat\phi_\infty)\, dx\\ &= \int_\Gamma \frac{\varepsilon_-}{2}\hat\phi_0\partial_n(\hat\phi^-_{\Gamma,\infty} - \hat\phi_\infty)\, dS + \int_{\Omega_+} \frac{\varepsilon_-}{2}\nabla\hat\phi_0\cdot\nabla(\hat\phi_{\Gamma,\infty} - \hat\phi_\infty)\, dx \qquad [\text{by (3.3)}]\\ &= \int_\Gamma \frac{\varepsilon_+}{2}\hat\phi_0\partial_n\hat\phi^+_{\Gamma,\infty}\, dS - \int_\Gamma \frac{\varepsilon_-}{2}\hat\phi_0\partial_n\hat\phi_\infty\, dS \qquad [\text{by (2.16)}]\end{aligned}$$

$$
\begin{aligned}
&\quad + \int_{\Omega_+} \frac{\varepsilon_-}{2} \nabla\hat{\phi}_0 \cdot \nabla(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_\infty)\, dx \\
&= -\int_{\partial\Omega_+} \frac{\varepsilon_+}{2} \hat{\phi}_0 \partial_n \hat{\phi}_{\Gamma,\infty}\, dS + \int_{\partial\Omega_+} \frac{\varepsilon_-}{2} \hat{\phi}_0 \partial_n \hat{\phi}_\infty\, dS \qquad [\text{since } \hat{\phi}_0 = 0 \text{ on } \partial\Omega] \\
&\quad + \int_{\Omega_+} \frac{\varepsilon_-}{2} \nabla\hat{\phi}_0 \cdot \nabla(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_\infty)\, dx \\
&= -\int_{\Omega_+} \frac{\varepsilon_+}{2} \nabla\hat{\phi}_0 \cdot \nabla\hat{\phi}_{\Gamma,\infty}\, dx + \int_{\Omega_+} \frac{\varepsilon_-}{2} \nabla\hat{\phi}_0 \cdot \nabla\hat{\phi}_\infty\, dx \\
&\quad + \int_{\Omega_+} \frac{\varepsilon_-}{2} \nabla\hat{\phi}_0 \cdot \nabla(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_\infty)\, dx \\
&= \frac{\varepsilon_- - \varepsilon_+}{2} \int_{\Omega_+} \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla\hat{\phi}_0\, dx.
\end{aligned}
$$

Considering now the second and third terms in (3.4), we have

$$
\begin{aligned}
&\frac{1}{2}\sum_{i=1}^{N} Q_i(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})(x_i) \\
&\quad + \int_{\Omega_+} \left[\frac{1}{2}\left(\psi_\Gamma - \phi_{\Gamma,\infty}\right) B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) - B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dx \\
&= \sum_{i=1}^{N} Q_i(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})(x_i) - \frac{1}{2}\sum_{i=1}^{N} Q_i(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})(x_i) \\
&\quad + \int_{\Omega_+} \left[\frac{1}{2}(\psi_\Gamma - \phi_{\Gamma,\infty}) B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) - B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dx \\
&= \int_\Omega \varepsilon_\Gamma \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})\, dx \quad [\text{by (2.13)}] \\
&\quad - \frac{1}{2}\int_\Omega \left[\varepsilon_\Gamma \nabla\psi_\Gamma \cdot \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty}) + \chi_+ B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})\right] \quad [\text{by (2.22)}] \\
&\quad + \int_{\Omega_+} \left[\frac{1}{2}\left(\psi_\Gamma - \phi_{\Gamma,\infty}\right) B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) - B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dx \\
&= -\int_\Omega \frac{\varepsilon_\Gamma}{2} |\nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})|^2 dx - \int_{\Omega_+} B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) dx \\
&\quad + \int_\Omega \frac{\varepsilon_\Gamma}{2} \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty}) \cdot \nabla\hat{\phi}_{\Gamma,\infty}\, dx + \int_{\Omega_+} \frac{1}{2}(\hat{\phi}_{\Gamma,\infty} - \phi_{\Gamma,\infty}) B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) dx \\
&= -\int_\Omega \frac{\varepsilon_\Gamma}{2} |\nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})|^2 dx - \int_{\Omega_+} B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right) dx \\
&\quad + \int_\Omega \frac{\varepsilon_\Gamma}{2} \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty}) \cdot \nabla\hat{\phi}_{\Gamma,\infty}\, dx
\end{aligned}
$$

$$
\begin{aligned}
&+\int_{\Omega_+}\frac{\varepsilon_+}{2}(\hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty})\Delta(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})\,dx \quad \text{[by (2.24) and (2.15) ]}\\
=&-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}|\nabla(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})|^2dx-\int_{\Omega_+}B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dx\\
&+\int_{\Omega}\frac{\varepsilon_\Gamma}{2}\nabla(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})\cdot\nabla(\hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty})\,dx \quad \text{[by (2.3)]}\\
&-\int_{\Omega_+}\frac{\varepsilon_+}{2}\nabla(\hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty})\cdot\nabla(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})\,dx\\
&-\int_{\Gamma}\frac{\varepsilon_+}{2}\partial_n(\psi_\Gamma^+-\hat{\phi}_{\Gamma,\infty}^+)(\hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty})\,dS \quad \text{[since } \hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty}=0 \text{ on } \partial\Omega\text{]}\\
=&-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}|\nabla(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})|^2dx-\int_{\Omega_+}B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dx\\
&+\int_{\Omega_-}\frac{\varepsilon_-}{2}\nabla(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})\cdot\nabla(\hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty})\,dx\\
&-\int_{\Gamma}\frac{\varepsilon_-}{2}\partial_n(\psi_\Gamma^--\hat{\phi}_{\Gamma,\infty}^-)(\hat{\phi}_{\Gamma,\infty}-\phi_{\Gamma,\infty})\,dS \quad \text{[by (2.24) with } \psi=\psi_\Gamma \text{ and (2.16) ]}\\
=&-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}|\nabla(\psi_\Gamma-\hat{\phi}_{\Gamma,\infty})|^2dx-\int_{\Omega_+}B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dx. \quad \text{[by (3.3)]}
\end{aligned}
$$

Now (3.2) follows directly from (3.4) and the above two expressions. □

We define $G_\Gamma : \hat{\phi}_{\Gamma,\infty}+H_0^1(\Omega)\to\mathbb{R}\cup\{-\infty\}$ by

$$
G_\Gamma[\psi]=-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}|\nabla(\psi-\hat{\phi}_{\Gamma,\infty})|^2dx-\int_{\Omega_+}B\left(\psi-\frac{\phi_{\Gamma,\infty}}{2}\right)dx+g_{\Gamma,\infty},
$$

where

$$
g_{\Gamma,\infty}=\frac{\varepsilon_--\varepsilon_+}{2}\int_{\Omega_+}\nabla\hat{\phi}_{\Gamma,\infty}\cdot\nabla\hat{\phi}_0\,dx+\frac{1}{2}\sum_{i=1}^{N}Q_i(\hat{\phi}_\infty-\hat{\phi}_{\mathrm{C}})(x_i).
$$

We shall call $G_\Gamma$ the PB energy functional. Note that by Lemma 3.1, $E[\Gamma]=G_\Gamma[\psi_\Gamma]$. In fact, we have the following variational principle for the PB energy functional.

**Theorem 3.1.** *The Euler–Lagrange equation of the PB energy functional $G_\Gamma : \hat{\phi}_{\Gamma,\infty}+H_0^1(\Omega)\to\mathbb{R}\cup\{-\infty\}$ is exactly the dielectric-boundary PB equation. Moreover, the functional $G_\Gamma[\cdot]$ is uniquely maximized over $\hat{\phi}_{\Gamma,\infty}+H_0^1(\Omega)$ by the solution $\psi_\Gamma$ to the BVP of the PB equation* (1.7)*, and the maximum value is exactly $E[\Gamma]$.*

*Proof.* Direct calculations verify that the Euler–Lagrange equation for the PB energy functional $G_\Gamma[\cdot]$ is indeed the dielectric-boundary PB equation; cf. Definition 2.1. Hence, $\psi_\Gamma$ is a solution to the Euler–Lagrange equation, and is further the unique maximizer of the strictly concave functional $G_\Gamma$. These, together with Lemma 3.1, imply that the maximum value of the free energy is $G_\Gamma[\psi_\Gamma]=E[\Gamma]$. □

We remark that the PB functional $G_\Gamma[\cdot]$ is maximized, not minimized, among all the admissible electrostatic potentials; see [9] for related discussions.

## 3.2 Definition and Formula of the Dielectric Boundary Force

Let $\Gamma$ be a dielectric boundary as given in the assumption A1 in Subsection 2.1. Let $\phi : \mathbb{R}^3 \to \mathbb{R}$ be the signed distance function to $\Gamma$, negative in $\Omega_-$ (inside $\Gamma$) and positive in $\mathbb{R}^3 \setminus \overline{\Omega_-}$ (outside $\Gamma$). Then, $n = \nabla\phi$ is exactly the unit normal along $\Gamma$, pointing from $\Omega_-$ to $\Omega_+$. Since $\Gamma$ is assumed to be of the class $C^3$, there exists $d_0 > 0$ with

$$d_0 < \frac{1}{2}\min\left(\operatorname{dist}(\Gamma, \partial\Omega), \min_{1\le i\le N} \operatorname{dist}(x_i, \Gamma)\right)$$

such that the signed distance function $\phi$ is a $C^3$-function and $\nabla\phi \neq 0$ in the neighborhood

$$\mathcal{N}_0(\Gamma) = \{x \in \Omega : \operatorname{dist}(x, \Gamma) < d_0\} \tag{3.5}$$

in $\Omega$ of $\Gamma$; cf. [27] (Section 14.6) and [34]. Define

$$\mathcal{V} = \{V \in C^2_c(\mathbb{R}^3, \mathbb{R}^3) : \operatorname{supp}(V) \subset \mathcal{N}_0(\Gamma)\}. \tag{3.6}$$

Let $V \in \mathcal{V}$. For any $X \in \mathbb{R}^3$, let $x = x(t, X)$ be the unique solution to the initial-value problem

$$\dot{x} = V(x) \quad (t \in \mathbb{R}) \qquad \text{and} \qquad x(0, X) = X, \tag{3.7}$$

where a dot denotes the derivative with respect to $t$. Define $T_t(X) = x(t, X)$ for any $X \in \mathbb{R}^3$ and any $t \in \mathbb{R}$. Then, $\{T_t\}_{t\in\mathbb{R}}$ is a family of diffeomorphisms and $C^2$-maps from $\mathbb{R}^3$ to $\mathbb{R}^3$ with $T_0 = I$ the identity map and $T_{-t} = T_t^{-1}$ for any $t \in \mathbb{R}$.

Let $t \in \mathbb{R}$. Since $\operatorname{supp}(V) \subset \mathcal{N}_0(\Gamma) \subset \Omega$, we have $T_t(\Omega) = \Omega$ and $T_t(\partial\Omega) = \partial\Omega$. Clearly, $T_t(\Omega_-) \subset \Omega$ and $T_t(\Omega_+) = \Omega \setminus \overline{T_t(\Omega_-)}$. Moreover, $\Gamma_t := T_t(\Gamma) = \partial T_t(\Omega_-) = \overline{T_t(\Omega_-)} \cap \overline{T_t(\Omega_+)}$ is of class $C^2$. Note that $x_i \in T_t(\Omega_-)$ and $T_t(x_i) = x_i$ for all $i = 1, \ldots, N$. Analogous to (1.5), $\varepsilon_{\Gamma_t}$ is defined correspondingly with respect to $T_t(\Omega_-)$ and $T_t(\Omega_+)$. We shall denote $\Gamma_t = \Gamma_t(V)$ to indicate the dependence of $\Gamma_t$ on $V \in \mathcal{V}$. For each $t \in \mathbb{R}$, the electrostatic free energy $E[\Gamma_t(V)]$ is defined in (3.1) (cf. also (3.2)) with $\Gamma_t = \Gamma_t(V)$ replacing $\Gamma$.

**Definition 3.1.** *The first variation of $E[\Gamma]$ with respect to $V \in \mathcal{V}$ is*

$$\delta_{\Gamma,V} E[\Gamma] = \frac{d}{dt} E[\Gamma_t(V)]\bigg|_{t=0} = \lim_{t\to 0^+} \frac{E[\Gamma_t(V)] - E[\Gamma]}{t},$$

*if the limit exists.*

We recall that the tangential gradient along a dielectric boundary $\Gamma$ is given by $\nabla_\Gamma = (I - n \otimes n)\nabla$, where $I$ is the identity matrix. The following theorem provides an explicit formula of the first variation $\delta_{\Gamma,V} E[\Gamma]$, and its proof is given in Section 5:

**Theorem 3.2.** *Let $\psi_\Gamma \in \hat{\phi}_C + H_0^1(\Omega)$ be the unique weak solution to the BVP of the dielectric-boundary PB equation* (1.7)*. Then, for any $V \in \mathcal{V}$, the first variation $\delta_{\Gamma,V} E[\Gamma]$ exists, and is given by*

$$\delta_{\Gamma,V} E[\Gamma] = \int_\Gamma q_\Gamma (V \cdot n)\, dS,$$

*where*

$$\begin{aligned} q_\Gamma = &-\frac{1}{2}\left(\frac{1}{\varepsilon_+} - \frac{1}{\varepsilon_-}\right)\left(|\varepsilon_\Gamma \partial_n \psi_\Gamma|^2 - \varepsilon_\Gamma \partial_n \psi_\Gamma \varepsilon_\Gamma \partial_n \phi_{\Gamma,\infty}\right) \\ &+ \frac{\varepsilon_+ - \varepsilon_-}{2}\left(|\nabla_\Gamma \psi_\Gamma|^2 - \nabla_\Gamma \psi_\Gamma \cdot \nabla_\Gamma \phi_{\Gamma,\infty}\right) + B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right). \end{aligned} \tag{3.8}$$

We identify $q_\Gamma$ in (3.8) as the first variation of $E[\Gamma]$ and denote it as $q_\Gamma = \delta_\Gamma E[\Gamma]$. We call $-\delta_\Gamma E[\Gamma]$ the (normal component of the) dielectric boundary force.

# 4 Some Lemmas: The Calculus of Boundary Variations

## 4.1 Properties of the Transformation $T_t$

We first recall some properties of the family of transformations $T_t : \mathbb{R}^3 \to \mathbb{R}^3$ ($t \in \mathbb{R}$) defined by (3.7) in Subsection 3.2 above via a vector field $V \in C_c^2(\mathbb{R}^3, \mathbb{R}^3)$. These properties hold true if we change $\mathbb{R}^3$ to $\mathbb{R}^d$ with a general dimension $d \geq 2$. They can be proved by direct calculations; cf. [20] (Section 4 of Chapter 9).

(1) Let $X \in \mathbb{R}^3$ and $t \in \mathbb{R}$. Let $\nabla T_t(X)$ be the gradient matrix of $T_t$ at $X$ with its entries $(\nabla T_t(X))_{ij} = \partial_j T_t^i(X)$ $(i, j = 1, 2, 3)$, where $T_t^i$ is the $i$th component of $T_t$. Let

$$J_t(X) = \det \nabla T_t(X). \tag{4.1}$$

Then for each $X \in \mathbb{R}^3$ the function $t \mapsto J_t(X)$ is a $C^2$-function and

$$\frac{dJ_t}{dt} = ((\nabla \cdot V) \circ T_t) J_t,$$

where $\circ$ denotes the composition of functions or maps. Clearly, $\nabla T_0 = I$, the identity matrix, and $J_0 = 1$. Moreover,

$$J_t(X) = 1 + t(\nabla \cdot V)(X) + H(t, X)t^2 \qquad \forall t \in \mathbb{R} \quad \forall X \in \mathbb{R}^3, \tag{4.2}$$

where $H(t, X)$ satisfies

$$\sup\{|H(t, X)| : t \in \mathbb{R}, X \in \mathbb{R}^3\} < \infty, \tag{4.3}$$

since $V$ is compactly supported.

(2) For each $t \in \mathbb{R}$, we define $A_V(t) : \mathbb{R}^3 \to \mathbb{R}^{3\times 3}$ by

$$A_V(t)(X) = J_t(X)\left(\nabla T_t(X)\right)^{-1}\left(\nabla T_t(X)\right)^{-T}, \tag{4.4}$$

where a superscript $T$ denotes the matrix transpose. The mapping $A_V(t)$ collects some terms together when the change of variable $x = T_t(X)$ is made to an integral over $\Omega$ of a function of the type $a\nabla u \cdot \nabla v$ (for some functions $a$, $u$, and $v$); cf. e.g., (4.21) and the equation above that. Clearly, $A_V(t) \in C^1(\mathbb{R}^3, \mathbb{R}^{3\times 3})$, and the $t$-derivative of $A_V(t)$ at each point in $\mathbb{R}^3$ is

$$\begin{aligned} A_V'(t) = \big[&((\nabla \cdot V) \circ T_t) - (\nabla T_t)^{-1}((\nabla V) \circ T_t)\nabla T_t \\ &-(\nabla T_t)^{-1}((\nabla V) \circ T_t)^T(\nabla T_t)\big]\, A_V(t). \end{aligned} \tag{4.5}$$

In particular

$$A_V'(0) = (\nabla \cdot V)I - \nabla V - (\nabla V)^T. \tag{4.6}$$

Moreover,

$$A_V(t)(X) = I + tA_V'(0)(X) + K(t,X)t^2 \qquad \forall t \in \mathbb{R} \quad \forall X \in \mathbb{R}^3, \tag{4.7}$$

where $K(t,X)$ satisfies

$$\sup\{|K(t,X)| : t \in \mathbb{R}, X \in \mathbb{R}^3\} < \infty. \tag{4.8}$$

(3) For any $u \in L^2(\Omega)$ and $t \in \mathbb{R}$, $u \circ T_t \in L^2(\Omega)$ and $u \circ T_t^{-1} \in L^2(\Omega)$. Moreover,

$$\lim_{t\to 0} u \circ T_t = u \qquad \text{and} \qquad \lim_{t\to 0} u \circ T_t^{-1} = u \qquad \text{in } L^2(\Omega). \tag{4.9}$$

For any $u \in H^1(\Omega)$ and $t \in \mathbb{R}$, $u \circ T_t \in H^1(\Omega)$ and $u \circ T_t^{-1} \in H^1(\Omega)$. Moreover,

$$\nabla(u \circ T_t^{-1}) = (\nabla T_t^{-1})^T\left(\nabla u \circ T_t^{-1}\right) \quad \text{and} \quad \nabla(u \circ T_t) = (\nabla T_t)^T\left(\nabla u \circ T_t\right), \tag{4.10}$$

$$\lim_{t\to 0} u \circ T_t = u \qquad \text{and} \qquad \lim_{t\to 0} u \circ T_t^{-1} = u \qquad \text{in } H^1(\Omega). \tag{4.11}$$

If $u \in H^2(\Omega)$, then

$$\lim_{t\to 0} \left\| \frac{u \circ T_t - u}{t} - \nabla u \cdot V \right\|_{H^1(\Omega)} = 0. \tag{4.12}$$

## 4.2 Continuity and Differentiability

Let $\Gamma$ be a dielectric boundary satisfying the assumptions in A1 of Subsection 2.1 and $V \in \mathcal{V}$ (cf. (3.6)). Let $\{T_t\}_{t\in\mathbb{R}}$ be the corresponding family of diffeomorphisms defined by (3.7). Let $\hat{\phi} \in W^{1,1}(\Omega)$ satisfy (2.12). We consider the approximations $\hat{\phi} \circ T_t$. Note that $\hat{\phi} \circ T_t - \hat{\phi}$ and $\nabla\hat{\phi} \cdot V$ vanish in any small neighborhood of $\cup_{i=1}^N x_i$, as $V(X) = 0$ and $T_t(X) = X$ for any $X$ in such a neighborhood and any $t \in \mathbb{R}$.

**Lemma 4.1.** *Let $\hat{\phi} \in \hat{\phi}_{\mathrm{C}} + H^1(\Omega)$ satisfy (2.12). We have $\hat{\phi} \circ T_t - \hat{\phi} \in H^1(\Omega)$ for all $t \geq 0$ and*

$$\lim_{t \to 0} \|\hat{\phi} \circ T_t - \hat{\phi}\|_{H^1(\Omega)} = 0. \tag{4.13}$$

*Moreover, $\nabla \hat{\phi} \cdot V \in H^1(\Omega)$ and*

$$\lim_{t \to 0} \left\| \frac{\hat{\phi} \circ T_t - \hat{\phi}}{t} - \nabla \hat{\phi} \cdot V \right\|_{H^1(\Omega)} = 0. \tag{4.14}$$

*Proof.* We note that both $\hat{\phi}$ and $\hat{\phi} \circ T_t$ are not in $H^1(\Omega)$ due to the singularities at $x_i$ $(i = 1, \ldots, N)$. Let $\sigma > 0$ be such that $B_\sigma := \cup_{i=1}^N B(x_i, \sigma) \subset \Omega$ and $V = 0$ on $B_\sigma$. Then, there exists $\tilde{\phi} \in C^\infty(\Omega) \cap H^2(\Omega)$ such that $\tilde{\phi} = 0$ in $B_{\sigma/2}$ and $\tilde{\phi} = \hat{\phi}$ a.e. in $\Omega \setminus B_\sigma$. These imply that $\tilde{\phi} \circ T_t - \tilde{\phi} = \hat{\phi} \circ T_t - \hat{\phi}$, and $\nabla \tilde{\phi} \cdot V = \nabla \hat{\phi} \cdot V$ a.e. in $\Omega$ for all $t$. This implies that $\nabla \hat{\phi} \cdot V \in H^1(\Omega)$. Moreover, it follows from (4.11) that

$$\lim_{t \to 0} \|\hat{\phi} \circ T_t - \hat{\phi}\|_{H^1(\Omega)} = \lim_{t \to 0} \|\tilde{\phi} \circ T_t - \tilde{\phi}\|_{H^1(\Omega)} = 0,$$

implying (4.13), and from (4.12) that

$$\lim_{t \to 0} \left\| \frac{\hat{\phi} \circ T_t - \hat{\phi}}{t} - \nabla \hat{\phi} \cdot V \right\|_{H^1(\Omega)} = \lim_{t \to 0} \left\| \frac{\tilde{\phi} \circ T_t - \tilde{\phi}}{t} - \nabla \tilde{\phi} \cdot V \right\|_{H^1(\Omega)} = 0,$$

implying (4.14). □

We recall that $\phi_{\Gamma,\infty} \in H^1(\Omega) \cap C(\overline{\Omega})$ is the unique weak solution to the BVP (1.8), defined in (2.3). Similarly, $\phi_{\Gamma_t,\infty} \in H^1(\Omega) \cap C(\overline{\Omega})$ for each $t \in \mathbb{R}$ is the unique weak solution to the same BVP with $\Gamma_t = T_t(\Gamma)$ replacing $\Gamma$. We note that the support of $V$ and hence that of $A'_V(0)$ (cf. (4.6)) do not contain any of the singularities $x_i$ $(1 \leq i \leq N)$.

**Lemma 4.2.** (1) *There exists a unique $\zeta_{\Gamma,V} \in H^1_0(\Omega)$ such that*

$$\int_\Omega \varepsilon_\Gamma \nabla \zeta_{\Gamma,V} \cdot \nabla \eta \, dx = -\int_\Omega \varepsilon_\Gamma A'_V(0) \nabla \phi_{\Gamma,\infty} \cdot \nabla \eta \, dx \qquad \forall \eta \in H^1_0(\Omega), \tag{4.15}$$

*where $A'_V(0)$ is defined in (4.6). Moreover, the mapping $V \mapsto \zeta_{\Gamma,V}$ is linear in $V$, i.e.,*

$$\zeta_{\Gamma,c_1V_1+c_2V_2} = c_1\zeta_{\Gamma,V_1} + c_2\zeta_{\Gamma,V_2} \qquad \textit{for all } V_1, V_2 \in \mathcal{V} \textit{ and } c_1, c_2 \in \mathbb{R}.$$

(2) *We have $\zeta_{\Gamma,V}|_{\Omega_s} \in H^2(\Omega_s) \cap C^1(\Omega_s)$ for $s = -$ or $+$. Moreover,*

$$\Delta \zeta_{\Gamma,V} = -\nabla \cdot [A'_V(0) \nabla \phi_{\Gamma,\infty}] = \Delta(\nabla \phi_{\Gamma,\infty} \cdot V) \qquad \textit{in } \Omega_- \cup \Omega_+, \tag{4.16}$$

$$[\![\varepsilon_\Gamma \partial_n \zeta_{\Gamma,V}]\!]_\Gamma = -[\![\varepsilon_\Gamma A'_V(0) \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma \qquad \textit{on } \Gamma. \tag{4.17}$$

(3) *We have*

$$\lim_{t\to 0} \|\phi_{\Gamma_t,\infty}\circ T_t - \phi_{\Gamma,\infty}\|_{H^1(\Omega)} = 0, \tag{4.18}$$

$$\lim_{t\to 0} \left\| \frac{\phi_{\Gamma_t,\infty}\circ T_t - \phi_{\Gamma,\infty}}{t} - \zeta_{\Gamma,V} \right\|_{H^1(\Omega)} = 0. \tag{4.19}$$

(4) *If* $V\cdot n = 0$ *on* $\Gamma$*, then* $\zeta_{\Gamma,V} = \nabla\phi_{\Gamma,\infty}\cdot V$ *in* $\Omega$.

*Proof.* (1) The existence and uniqueness of $\zeta_{\Gamma,V}\in H^1_0(\Omega)$ that satisfies (4.15) follow from the Lax–Milgram Lemma [24, 27]. By (4.6), $A'_V(0)$ is linear in $V$. Therefore, by the definition (4.15) of $\zeta_{\Gamma,V}\in H^1_0(\Omega)$, $\zeta_{\Gamma,V}$ is linear in $V$.

(2) Let s denote $-$ or $+$. Note by (2.4), (3.6), and (4.6) that $A'_V(0)\nabla\phi_{\Gamma,\infty}\in C^1(\Omega_s)\cap H^1(\Omega_s)$. For any $\eta\in C^1_c(\Omega)$ with $\mathrm{supp}\,(\eta)\subset\Omega_s$, we have by (4.15) and the Divergence Theorem that

$$\int_{\Omega_s}\varepsilon_s\nabla\zeta_{\Gamma,V}\cdot\nabla\eta\,dx = \int_{\Omega_s}\varepsilon_s\nabla\cdot[A'_V(0)\nabla\phi_{\Gamma,\infty}]\,\eta\,dx.$$

Hence, $-\Delta\zeta_{\Gamma,V} = \nabla\cdot[A'_V(0)\nabla\phi_{\Gamma,\infty}]$ in $\Omega_s$. Since the right-hand side is in $L^2(\Omega_s)\cap C(\Omega_s)$, it follows from the elliptic regularity theory [27, 35] that $\zeta_{\Gamma,V}|_{\Omega_s}\in H^2(\Omega_s)\cap C^1(\Omega_s)$, after a possible modification of the value of $\zeta_{\Gamma,V}$ on a set of zero Lebesgue measure. Moreover, the first equality in (4.16) follows.

Let us denote by $V^i$ $(i=1,2,3)$ the components of $V$. With the conventional summation notation (i.e., repeated indices are summed), we have by (4.6), (2.4), and (2.6) that

$$\begin{aligned}
&-\nabla\cdot(A'_V(0)\nabla\phi_{\Gamma,\infty})\\
&\quad=\nabla\cdot\left[\nabla V+(\nabla V)^T-(\nabla\cdot V)I\right]\nabla\phi_{\Gamma,\infty}\\
&\quad=\partial_i\left(\partial_j V^i\partial_j\phi_{\Gamma,\infty}+\partial_i V^j\partial_j\phi_{\Gamma,\infty}-\partial_k V^k\partial_i\phi_{\Gamma,\infty}\right)\\
&\quad=2\partial_{ij}\phi_{\Gamma,\infty}\partial_i V^j+\partial_{ii}V^j\partial_j\phi_{\Gamma,\infty}\quad[\text{since }\partial_{ii}\phi_{\Gamma,\infty}=0]\\
&\quad=\partial_{ii}\partial_j\phi_{\Gamma,\infty}V^j+2\partial_{ij}\phi_{\Gamma,\infty}\partial_i V^j+\partial_{ii}V^j\partial_j\phi_{\Gamma,\infty}\quad[\text{since }\partial_{ii}\phi_{\Gamma,\infty}=0]\\
&\quad=\partial_{ii}\left(\partial_j\phi_{\Gamma,\infty}V^j\right)\\
&\quad=\Delta(\nabla\phi_{\Gamma,\infty}\cdot V)\qquad\text{in }\Omega_-\cup\Omega_+,
\end{aligned}\tag{4.20}$$

implying the second equation in (4.16).

Since $\zeta_{\Gamma,V}\in H^1_0(\Omega)$ and $\Delta\zeta_{\Gamma,V}\in L^2(\Omega_s)$ for s being $-$ or $+$, and since the unit normal $n$ at the $\Gamma$ points from $\Omega_-$ to $\Omega_+$, we have by the Divergence Theorem that both sides of the equation in (4.15) are

$$\begin{aligned}
\int_\Omega\varepsilon_\Gamma\nabla\zeta_{\Gamma,V}\cdot\nabla\eta\,dx &= \int_{\Omega_-}\varepsilon_-\nabla\zeta_{\Gamma,V}\cdot\nabla\eta\,dx+\int_{\Omega_+}\varepsilon_+\nabla\zeta_{\Gamma,V}\cdot\nabla\eta\,dx\\
&=-\int_{\Omega_-}\varepsilon_-\Delta\zeta_{\Gamma,V}\eta\,dx-\int_{\Omega_+}\varepsilon_+\Delta\zeta_{\Gamma,V}\eta\,dx-\int_\Gamma[\![\varepsilon_\Gamma\partial_n\zeta_{\Gamma,V}]\!]_\Gamma\eta\,dS,
\end{aligned}$$

and

$$
\begin{aligned}
&-\int_\Omega \varepsilon_\Gamma A_V'(0)\nabla\phi_{\Gamma,\infty}\cdot\nabla\eta\,dx\\
&\quad=-\int_{\Omega_-}\varepsilon_- A_V'(0)\nabla\phi_{\Gamma,\infty}\cdot\nabla\eta\,dx-\int_{\Omega_+}\varepsilon_+ A_V'(0)\nabla\phi_{\Gamma,\infty}\cdot\nabla\eta\,dx\\
&\quad=\int_{\Omega_-}\varepsilon_-\nabla\cdot(A_V'(0)\nabla\phi_{\Gamma,\infty})\eta\,dx+\int_{\Omega_+}\varepsilon_+\nabla\cdot(A_V'(0)\nabla\phi_{\Gamma,\infty})\eta\,dx\\
&\qquad+\int_\Gamma[\![\varepsilon_\Gamma A_V'(0)\nabla\phi_{\Gamma,\infty}\cdot n]\!]_\Gamma\eta\,dS,
\end{aligned}
$$

respectively. These, together with (4.16), imply (4.17).

(3) Replacing $\Gamma$, $\phi_{\Gamma,\infty}$, and $\eta$ by $\Gamma_t$, $\phi_{\Gamma_t,\infty}$, and $\eta\circ T_t^{-1}$ for $t\in\mathbb{R}$, respectively, in the weak formulation (2.3), we get by the change of variable $x=T_t(X)$ that

$$
\int_\Omega \varepsilon_\Gamma A_V(t)\nabla(\phi_{\Gamma_t,\infty}\circ T_t)\cdot\nabla\eta\,dX=0\qquad\forall\eta\in H_0^1(\Omega).
$$

This and (2.3) imply for any $\eta\in H_0^1(\Omega)$ that

$$
\int_\Omega \varepsilon_\Gamma\nabla(\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty})\cdot\nabla\eta\,dX=\int_\Omega\varepsilon_\Gamma[I-A_V(t)]\nabla(\phi_{\Gamma_t,\infty}\circ T_t)\cdot\nabla\eta\,dX. \tag{4.21}
$$

It follows from a change of variable, (4.2), (4.3), (4.7), and (4.8) that $\|\nabla(\phi_{\Gamma_t,\infty}\circ T_t)\|_{L^2(\Omega)}$ is bounded uniformly in $t$. Setting $\eta=\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}\in H_0^1(\Omega)$ in (4.21), we then obtain (4.18) by (4.7), (4.8), and the Cauchy–Schwarz and Poincaré inequalities.

Dividing both sides of (4.21) by $t\neq0$ and setting now $\eta=(\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty})/t-\zeta_{\Gamma,V}$ in the resulting equation and also in (4.15), we have by the Cauchy–Schwarz inequality that

$$
\begin{aligned}
&\int_\Omega\varepsilon_\Gamma\left|\nabla\left(\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right)\right|^2dX\\
&\quad=\int_\Omega\varepsilon_\Gamma\left[\frac{I-A_V(t)}{t}+A_V'(0)\right]\nabla(\phi_{\Gamma_t,\infty}\circ T_t)\cdot\nabla\left(\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right)dX\\
&\qquad+\int_\Omega\varepsilon_\Gamma A_V'(0)\nabla\left[\phi_{\Gamma,\infty}-\phi_{\Gamma_t,\infty}\circ T_t\right]\cdot\nabla\left(\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right)dX\\
&\quad\le C\left\|\frac{A_V(t)-I-tA_V'(0)}{t}\right\|_{L^\infty(\Omega)}\|\phi_{\Gamma_t,\infty}\circ T_t\|_{H^1(\Omega)}\left\|\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right\|_{H^1(\Omega)}\\
&\qquad+C\|\phi_{\Gamma,\infty}-\phi_{\Gamma_t,\infty}\circ T_t\|_{H^1(\Omega)}\left\|\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right\|_{H^1(\Omega)}.
\end{aligned}
$$

This, together with Poincaré's inequality, (4.7), (4.8), and (4.18), leads to (4.19).

(4) Assume now $V\cdot n=0$ on $\Gamma$. This means that $V$ is tangent to $\Gamma$ at every point in $\Gamma$. Since $\Gamma$ is a compact manifold, by a classical result on the initial-value problem of differential

equations on a differentiable manifold, each trajectory $T_t(X)$ ($t \in \mathbb{R}$) defined by the vector-field $V$ that starts from $T_0(X) = X \in \Gamma$ will stay in $\Gamma$ for all $t \in \mathbb{R}$ [4, 36]. Therefore, $\Gamma_t = \Gamma$ for all $t \in \mathbb{R}$. Let $\eta \in L^2(\Omega)$ and $t \neq 0$. We have by the properties of the transformations $T_t$ ($t \in \mathbb{R}$) (4.9), (4.12), and (4.1)–(4.3) that

$$\begin{aligned}
\int_\Omega \frac{\phi_{\Gamma_t,\infty} \circ T_t - \phi_{\Gamma,\infty}}{t} \eta \, dX &= \int_\Omega \frac{\phi_{\Gamma,\infty} \circ T_t}{t} \eta \, dX - \int_\Omega \frac{\phi_{\Gamma,\infty}\eta}{t} \, dX \\
&= \int_\Omega \frac{\phi_{\Gamma,\infty}}{t} \left(\eta \circ T_t^{-1}\right) \det \nabla T_t^{-1} dx - \int_\Omega \frac{\phi_{\Gamma,\infty}\eta}{t} \, dx \\
&= \int_\Omega \phi_{\Gamma,\infty} \left( \frac{\eta \circ T_t^{-1} - \eta}{t} \det \nabla T_t^{-1} + \eta \frac{\det \nabla T_t^{-1} - 1}{t} \right) dx \\
&\to - \int_\Omega \phi_{\Gamma,\infty} \nabla \eta \cdot V \, dx - \int_\Omega \phi_{\Gamma,\infty} \eta (\nabla \cdot V) \, dx \\
&= \int_\Omega (\nabla \phi_{\Gamma,\infty} \cdot V) \eta \, dx \qquad \text{as } t \to 0.
\end{aligned}$$

Since $\eta \in L^2(\Omega)$ is arbitrary, this and (4.19) imply that $\zeta_{\Gamma,V} = \nabla \phi_{\Gamma,\infty} \cdot V$ in $\Omega$. □

We recall that $\hat{\phi}_{\Gamma,\infty}$ is determined by (2.13) and the boundary condition $\hat{\phi}_{\Gamma,\infty} = \phi_\infty$ on $\partial\Omega$. For each $t \in \mathbb{R}$, we denote by $\hat{\phi}_{\Gamma_t,\infty}$ the unique function that is defined by (2.13) with $\Gamma_t$ replacing $\Gamma$ and the same boundary condition $\hat{\phi}_{\Gamma_t,\infty} = \phi_\infty$ on $\partial\Omega$. Note again that the support of $V$ or $A'_V(0)$ contains no singularities $x_i$ ($i = 1, \dots, N$).

**Lemma 4.3.** (1) *There exists a unique $\xi_{\Gamma,V} \in H_0^1(\Omega)$ such that*

$$\int_\Omega \varepsilon_\Gamma \nabla \xi_{\Gamma,V} \cdot \nabla \eta \, dx = - \int_\Omega \varepsilon_\Gamma A'_V(0) \nabla \hat{\phi}_{\Gamma,\infty} \cdot \nabla \eta \, dx \qquad \forall \eta \in H_0^1(\Omega). \tag{4.22}$$

(2) *We have $\xi_{\Gamma,V}|_{\Omega_s} \in H^2(\Omega_s) \cap C^1(\Omega_s)$ for $s = -$ or $+$. Moreover,*

$$\begin{aligned}
&\Delta \xi_{\Gamma,V} = -\nabla \cdot A'_V(0) \nabla \hat{\phi}_{\Gamma,\infty} = \Delta(\nabla \hat{\phi}_{\Gamma,\infty} \cdot V) \qquad \text{in } \Omega_- \cup \Omega_+, \\
&[\![ \varepsilon_\Gamma \partial_n \xi_{\Gamma,V} ]\!]_\Gamma = -[\![ \varepsilon_\Gamma A'_V(0) \partial_n \hat{\phi}_{\Gamma,\infty} ]\!]_\Gamma \qquad \text{on } \Gamma.
\end{aligned} \tag{4.23}$$

(3) *We have*

$$\begin{aligned}
&\lim_{t \to 0} \| \hat{\phi}_{\Gamma_t,\infty} \circ T_t - \hat{\phi}_{\Gamma,\infty} \|_{H^1(\Omega)} = 0, \\
&\lim_{t \to 0} \left\| \frac{\hat{\phi}_{\Gamma_t,\infty} \circ T_t - \hat{\phi}_{\Gamma,\infty}}{t} - \xi_{\Gamma,V} \right\|_{H^1(\Omega)} = 0.
\end{aligned}$$

*Proof.* The proof is the same as and simpler than that of the next lemma, Lemma 4.4, as there is an extra term $B$ there, which can be set to 0 here. The only exception is the second equality in (4.23) which can be obtained by the same calculations as in (4.20). □

We recall that $\psi_\Gamma \in \hat{\phi}_{\rm C} + H^1(\Omega) \cap C(\overline{\Omega}) = \hat{\phi}_{\Gamma,\infty} + H^1(\Omega) \cap C(\overline{\Omega})$ is the unique weak solution to the BVP of the dielectric-boundary PB equation (1.7); cf. Definition 2.1. For each $t \in \mathbb{R}$, we denote by $\psi_{\Gamma_t} \in \hat{\phi}_{\rm C} + H^1(\Omega) \cap C(\overline{\Omega})$ the unique solution to the same BVP with $\Gamma_t$ replacing $\Gamma$.

**Lemma 4.4.** (1) *There exists a unique $\omega_{\Gamma,V} \in H^1_0(\Omega)$ such that*

$$\begin{aligned}
&\int_\Omega \left[ \varepsilon_\Gamma \nabla \omega_{\Gamma,V} \cdot \nabla \eta + \chi_+ B'' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) \omega_{\Gamma,V} \eta \right] dx \\
&\quad = - \int_\Omega \varepsilon_\Gamma A_V'(0) \nabla \psi_\Gamma \cdot \nabla \eta \, dx \\
&\qquad - \int_{\Omega_+} \left[ (\nabla \cdot V) B' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) - \frac{\zeta_{\Gamma,V}}{2} B'' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) \right] \eta \, dx \qquad \forall \eta \in H^1_0(\Omega).
\end{aligned} \tag{4.24}$$

(2) *We have $\omega_{\Gamma,V}|_{\Omega_s} \in H^2(\Omega_s) \cap C^1(\Omega_s)$ for $s = -$ or $+$. Moreover,*

$$\Delta \omega_{\Gamma,V} = -\nabla \cdot A_V'(0) \nabla \psi_\Gamma = \Delta(\nabla \psi_\Gamma \cdot V) \qquad \text{in } \Omega_-, \tag{4.25}$$

$$\begin{aligned}
&\varepsilon_+ \Delta \omega_{\Gamma,V} - B'' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) \omega_{\Gamma,V} = -\varepsilon_+ \nabla \cdot A_V'(0) \nabla \psi_\Gamma \\
&\quad + (\nabla \cdot V) B' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) - \frac{\zeta_{\Gamma,V}}{2} B'' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) \qquad \text{in } \Omega_+,
\end{aligned} \tag{4.26}$$

$$[\![ \varepsilon_\Gamma \partial_n \omega_{\Gamma,V} ]\!]_\Gamma = -[\![ \varepsilon_\Gamma A_V'(0) \partial_n \psi_\Gamma ]\!]_\Gamma \qquad \text{on } \Gamma. \tag{4.27}$$

(3) *We have*

$$\lim_{t \to 0} \| \psi_{\Gamma_t} \circ T_t - \psi_\Gamma \|_{H^1(\Omega)} = 0, \tag{4.28}$$

$$\lim_{t \to 0} \left\| \frac{\psi_{\Gamma_t} \circ T_t - \psi_\Gamma}{t} - \omega_{\Gamma,V} \right\|_{H^1(\Omega)} = 0. \tag{4.29}$$

*Proof.* (1) Since $B'' > 0$, the support of $V$ does not contain any of the singularities $x_i$ ($i = 1, \ldots, N$), and $\psi_\Gamma$ and $\phi_{\Gamma,\infty}$ are uniformly bounded on the union of the support of $V$ and $\Omega_+$ (cf. (2.18) and (2.23)), the existence and uniqueness of $\omega_{\Gamma,V} \in H^1_0(\Omega)$ that satisfies (4.24) follows from the Lax–Milgram Lemma [24, 27].

(2) Choosing $\eta \in C^1_{\rm c}(\Omega)$ in (4.24) with $\mathrm{supp}\,(\eta) \subset \Omega_-$ and applying the Divergence Theorem, we obtain the first equation of (4.25) a.e. in $\Omega_-$. Since the right-hand side of this first equation is in $L^2(\Omega_-) \cap C(\Omega_-)$, it follows from the regularity theory [27, 35] that, with a possible modification of the value of $\omega_{\Gamma,V}$ on a set of zero Lebesgue measure, $\omega_{\Gamma,V}|_{\Omega_-} \in H^2(\Omega_-) \cap C^1(\Omega_-)$. Now, the first equation in (4.25) holds for each point in $\Omega_-$. The second equation is similar to that in (4.16) (cf. (4.20)). By similar arguments, we obtain that $\omega_{\Gamma,V}|_{\Omega_+} \in H^2(\Omega_+) \cap C^1(\Omega_+)$ and (4.26). By splitting each of those two integrals in (4.24) that has the term $\nabla \eta$ into integrals over $\Omega_-$ and $\Omega_+$, respectively, using the Divergence Theorem, and using (4.25) and (4.26), we obtain (4.27).

(3) Let $\hat{\phi}_{\rm C}$ be given as in (1.6) and $t \in \mathbb{R}$. Denote $\psi_{\rm r} = \psi_\Gamma - \hat{\phi}_{\rm C}$ and $\psi_{{\rm r},t} = \psi_{\Gamma_t} - \hat{\phi}_{\rm C}$. We first prove (4.28). By (4.13) (with $\hat{\phi} = \hat{\phi}_{\rm C}$) in Lemma 4.1, it suffices to prove that

$$\lim_{t\to 0} \|\psi_{{\rm r},t} \circ T_t - \psi_{\rm r}\|_{H^1(\Omega)} = 0. \tag{4.30}$$

By Definition 2.1 and (2.12) (cf. also (2.14)), we have

$$\begin{aligned} &\int_\Omega \left[\varepsilon_\Gamma \nabla\psi_r \cdot \nabla\eta + \chi_+ B'\left(\psi_r + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma,\infty}}{2}\right)\eta\right] dx \\ &\quad = -(\varepsilon_+ - \varepsilon_-)\int_{\Omega_+} \nabla\hat{\phi}_{\rm C} \cdot \nabla\eta \, dx \qquad \forall \eta \in H^1_0(\Omega). \end{aligned} \tag{4.31}$$

Replacing $\Gamma$, $\Omega_+$, $\psi$, and $\eta$ in (4.31) by $\Gamma_t = T_t(\Gamma)$, $T_t(\Omega_+)$, $\psi_{\Gamma_t}$, and $\eta = \eta \circ T_t^{-1}$, respectively, we obtain by the change of variable $x = T_t(X)$ and (4.4) that

$$\begin{aligned} &\int_\Omega \left[\varepsilon_\Gamma A_V(t)\nabla(\psi_{r,t} \circ T_t) \cdot \nabla\eta + \chi_+ B'\left(\left(\psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2}\right) \circ T_t\right)\eta \, J_t\right] dX \\ &\quad = -(\varepsilon_+ - \varepsilon_-)\int_{\Omega_+} A_V(t)\nabla(\hat{\phi}_{\rm C} \circ T_t) \cdot \nabla\eta \, dX \qquad \forall \eta \in H^1_0(\Omega). \end{aligned} \tag{4.32}$$

Subtracting (4.31) from (4.32) and rearranging terms, we get

$$\begin{aligned} &\int_\Omega \varepsilon_\Gamma \left[\nabla(\psi_{{\rm r},t} \circ T_t) - \nabla\psi_{\rm r}\right] \cdot \nabla\eta \, dX \\ &\quad = -\int_\Omega \varepsilon_\Gamma [A_V(t) - I]\nabla(\psi_{{\rm r},t} \circ T_t) \cdot \nabla\eta \, dX \\ &\qquad - \int_{\Omega_+} B'\left(\left(\psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2}\right) \circ T_t\right)(J_t - 1)\eta \, dX \\ &\qquad - \int_{\Omega_+} \left[B'\left(\left(\psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2}\right) \circ T_t\right) - B'\left(\psi_{\rm r} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma,\infty}}{2}\right)\right]\eta \, dX \\ &\qquad - (\varepsilon_+ - \varepsilon_-)\int_{\Omega_+} [\nabla(\hat{\phi}_{\rm C} \circ T_t) - \nabla\hat{\phi}_{\rm C}] \cdot \nabla\eta \, dX \\ &\qquad - (\varepsilon_+ - \varepsilon_-)\int_{\Omega_+} [A_V(t) - I]\nabla(\hat{\phi}_{\rm C} \circ T_t) \cdot \nabla\eta \, dX \qquad \forall \eta \in H^1_0(\Omega). \end{aligned} \tag{4.33}$$

Setting $\eta = \psi_{{\rm r},t} \circ T_t - \psi_{\rm r}$, we have by the uniform bound of all $\psi_{{\rm r},t}$ and $\phi_{\Gamma_t,\infty}$ (cf. (2.23) and (2.5)), the Mean-Value Theorem, and the convexity of $B$ that

$$\begin{aligned} &-\left[B'\left(\left(\psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2}\right) \circ T_t\right) - B'\left(\psi_{\rm r} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma,\infty}}{2}\right)\right]\eta \\ &\quad = -B''(\lambda_t)\left(\psi_{{\rm r},t} \circ T_t - \psi_{\rm r} + \hat{\phi}_{\rm C} \circ T_t - \hat{\phi}_{\rm C} + \frac{1}{2}\phi_{\Gamma_t,\infty} \circ T_t - \frac{1}{2}\phi_{\Gamma,\infty}\right)(\psi_{{\rm r},t} \circ T_t - \psi_{\rm r}) \end{aligned}$$

$$
\begin{aligned}
&= -B''(\lambda_t)(\psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}})^2 \\
&\quad - B''(\lambda_t)\left(\hat\phi_{\mathrm{C}}\circ T_t - \hat\phi_{\mathrm{C}} + \frac{1}{2}\phi_{\Gamma_t,\infty}\circ T_t - \frac{1}{2}\phi_{\Gamma,\infty}\right)(\psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}}) \\
&\le C|(\hat\phi_{\mathrm{C}}\circ T_t - \hat\phi_{\mathrm{C}})(\psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}})| + C\,|(\phi_{\Gamma_t,\infty}\circ T_t - \phi_{\Gamma,\infty})(\psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}})|\,, \qquad (4.34)
\end{aligned}
$$

where $\lambda_t$ is in between $(\psi_{\mathrm{r},t} + \hat\phi_{\mathrm{C}} - \phi_{\Gamma_t,\infty}/2)\circ T_t$ and $\psi_{\mathrm{r}} + \hat\phi_{\mathrm{C}} - \phi_{\Gamma,\infty}/2$ at each point in $\Omega_+$, and the constant $C > 0$ is independent of $t$ and $\Gamma$. Now, the combination of (4.33) with $\eta = \psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}}$ and (4.34), together with the uniform bounds for $\psi_{\mathrm{r},t}$ and $\phi_{\Gamma_t,\infty}$, and the Cauchy–Schwarz and Poincaré inequalities, leads to

$$
\begin{aligned}
\|\psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}}\|_{H^1(\Omega)} \le{}& C\|A_V(t) - I\|_{L^\infty(\Omega)}\|\psi_{\mathrm{r},t}\circ T_t\|_{H^1(\Omega)} + C\|J_t - 1\|_{L^\infty(\Omega)} \\
&+ C\|\hat\phi_{\mathrm{C}}\circ T_t - \hat\phi_{\mathrm{C}}\|_{H^1(\Omega_+)} + C\,\|\phi_{\Gamma_t,\infty}\circ T_t - \phi_{\Gamma,\infty}\|_{H^1(\Omega)} \\
&+ C\,\|A_V(t) - I\|_{L^\infty(\Omega_+)}\,\|\hat\phi_{\mathrm{C}}\circ T_t\|_{H^1(\Omega_+)}.
\end{aligned}
$$

Now the convergence (4.30) follows from (4.2), (4.3), (4.7), (4.8), the uniform bound of $\psi_{\mathrm{r},t}$, Lemma 4.1 (with $\hat\phi = \hat\phi_{\mathrm{C}}$), and Lemma 4.2.

We now prove (4.29). Let us denote $\hat\omega_{\Gamma,V} = \omega_{\Gamma,V} - \nabla\hat\phi_{\mathrm{C}}\cdot V$. By Lemma 4.1 (cf. (4.14) with $\hat\phi = \hat\phi_{\mathrm{C}}$), we need only to prove that

$$
\lim_{t\to 0}\left\|\frac{\psi_{\mathrm{r},t}\circ T_t - \psi_{\mathrm{r}}}{t} - \hat\omega_{\Gamma,V}\right\|_{H^1(\Omega)} = 0. \qquad (4.35)
$$

Since the support of $V$ and $A_V'(0)$ does not contain any of the singularities $x_i$ ($i = 1,\dots,N$), the Divergence Theorem and the calculations in (4.20) imply that

$$
\begin{aligned}
&\int_\Omega [A_V'(0)\nabla\hat\phi_{\mathrm{C}} + \nabla(\nabla\hat\phi_{\mathrm{C}}\cdot V)]\cdot\nabla\eta\,dx \\
&\quad = -\int_\Omega [\nabla\cdot(A_V'(0)\nabla\hat\phi_{\mathrm{C}}) + \Delta(\nabla\hat\phi_{\mathrm{C}}\cdot V)]\,\eta\,dx \\
&\quad = 0 \qquad \forall\eta\in H_0^1(\Omega).
\end{aligned}
$$

This allows us to rewrite (4.24) into the following equation for $\hat\omega_{\Gamma,V}$:

$$
\begin{aligned}
&\int_\Omega\left[\varepsilon_\Gamma\nabla\hat\omega_{\Gamma,V}\cdot\nabla\eta + \chi_+ B''\left(\psi_{\mathrm{r}} + \hat\phi_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2}\right)\hat\omega_{\Gamma,V}\eta\right]dX \\
&\quad = -\int_\Omega \varepsilon_\Gamma A_V'(0)\nabla(\psi_\Gamma - \hat\phi_{\mathrm{C}})\cdot\nabla\eta\,dX \\
&\qquad -\int_{\Omega_+}\left[B'\left(\psi_{\mathrm{r}} + \hat\phi_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2}\right)(\nabla\cdot V)\right. \\
&\qquad\quad \left. + B''\left(\psi_{\mathrm{r}} + \hat\phi_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2}\right)\left(\nabla\hat\phi_{\mathrm{C}}\cdot V - \frac{\zeta_{\Gamma,V}}{2}\right)\right]\eta\,dX
\end{aligned}
$$

$$- (\varepsilon_+ - \varepsilon_-) \int_{\Omega_+} [A_V'(0)\nabla\hat{\phi}_{\mathrm{C}} + \nabla(\nabla\hat{\phi}_{\mathrm{C}} \cdot V)] \cdot \nabla\eta \, dX \qquad \forall \eta \in H_0^1(\Omega). \tag{4.36}$$

Multiplying both sides of (4.33) by $1/t$ and combining the resulting equation with (4.36), we obtain by rearranging terms that

$$\begin{aligned}
&\int_\Omega \varepsilon_\Gamma \nabla \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} - \hat{\omega}_{\Gamma,V} \right) \cdot \nabla\eta \, dX \\
&\quad = -\int_\Omega \varepsilon_\Gamma \left[ \left( \frac{A_V(t) - I}{t} \right) \nabla(\psi_{\mathrm{r},t} \circ T_t) - A_V'(0)\nabla\psi_{\mathrm{r}} \right] \cdot \nabla\eta \, dX \\
&\qquad - \int_{\Omega_+} \left[ B' \left( \left( \psi_{\mathrm{r},t} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma_t,\infty}}{2} \right) \circ T_t \right) \left( \frac{J_t - 1}{t} \right) \right. \\
&\qquad\quad \left. -B' \left( \psi_{\mathrm{r}} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2} \right) (\nabla \cdot V) \right] \eta \, dX \\
&\qquad - \int_{\Omega_+} \left\{ \frac{1}{t} \left[ B' \left( \left( \psi_{\mathrm{r},t} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma_t,\infty}}{2} \right) \circ T_t \right) - B' \left( \psi_{\mathrm{r}} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2} \right) \right] \right. \\
&\qquad\quad \left. -B'' \left( \psi_{\mathrm{r}} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2} \right) \left( \hat{\omega}_{\Gamma,V} + \nabla\hat{\phi}_{\mathrm{C}} \cdot V - \frac{\zeta_{\Gamma,V}}{2} \right) \right\} \eta \, dX \\
&\qquad - (\varepsilon_+ - \varepsilon_-) \int_{\Omega_+} \left[ \left( \frac{A_V(t) - I}{t} \right) \nabla(\hat{\phi}_{\mathrm{C}} \circ T_t) - A_V'(0)\nabla\hat{\phi}_{\mathrm{C}} \right] \cdot \nabla\eta \, dX \\
&\qquad - (\varepsilon_+ - \varepsilon_-) \int_{\Omega_+} \nabla \left( \frac{\hat{\phi}_{\mathrm{C}} \circ T_t - \hat{\phi}_{\mathrm{C}}}{t} - \nabla\hat{\phi}_{\mathrm{C}} \cdot V \right) \cdot \nabla\eta \, dX \qquad \forall \eta \in H_0^1(\Omega).
\end{aligned} \tag{4.37}$$

Specifying $\eta = (\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}})/t - \hat{\omega}_{\Gamma,V} \in H_0^1(\Omega)$, we have by the fact that $B'' > 0$, the Mean-Value Theorem, the uniform bound in $\Omega_+$ for all the functions $\hat{\phi}_C$ (cf. (1.6)) $\psi_{\mathrm{r},t}$ (cf. (2.5), (2.18), (2.23)), $\zeta_{\Gamma,V}$ (cf. part (2) of Lemma 4.2), and $\omega_{\Gamma,V}$ (cf. part (2) of Lemma 4.4) that in $\Omega_+$

$$\begin{aligned}
&- \left\{ \frac{1}{t} \left[ B' \left( \left( \psi_{\mathrm{r},t} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma_t,\infty}}{2} \right) \circ T_t \right) - B' \left( \psi_{\mathrm{r}} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2} \right) \right] \right. \\
&\quad \left. -B'' \left( \psi_{\mathrm{r}} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2} \right) \left( \hat{\omega}_{\Gamma,V} + \nabla\hat{\phi}_{\mathrm{C}} \cdot V - \frac{\zeta_{\Gamma,V}}{2} \right) \right\} \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} - \hat{\omega}_{\Gamma,V} \right) \\
&\quad = -B'' (\xi_t) \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} + \frac{\hat{\phi}_{\mathrm{C}} \circ T_t - \hat{\phi}_{\mathrm{C}}}{t} - \frac{\phi_{\Gamma_t,\infty} \circ T_t - \phi_{\Gamma,\infty}}{2t} \right) \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} - \hat{\omega}_{\Gamma,V} \right) \\
&\qquad + B'' \left( \psi_{\mathrm{r}} + \hat{\phi}_{\mathrm{C}} - \frac{\phi_{\Gamma,\infty}}{2} \right) \left( \hat{\omega}_{\Gamma,V} + \nabla\hat{\phi}_{\mathrm{C}} \cdot V - \frac{\zeta_{\Gamma,V}}{2} \right) \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} - \hat{\omega}_{\Gamma,V} \right) \\
&\quad = -B'' (\xi_t) \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} - \hat{\omega}_{\Gamma,V} \right)^2 \\
&\qquad - B'' (\xi_t) \left( \hat{\omega}_{\Gamma,V} + \frac{\hat{\phi}_{\mathrm{C}} \circ T_t - \hat{\phi}_{\mathrm{C}}}{t} - \frac{\phi_{\Gamma_t,\infty} \circ T_t - \phi_{\Gamma,\infty}}{2t} \right) \left( \frac{\psi_{\mathrm{r},t} \circ T_t - \psi_{\mathrm{r}}}{t} - \hat{\omega}_{\Gamma,V} \right)
\end{aligned}$$

$$
\begin{aligned}
&+\left[B''\left(\psi_{\mathrm{r}}+\hat{\phi}_{\mathrm{C}}-\frac{\phi_{\Gamma,\infty}}{2}\right)-B''(\xi_t)\right]\left(\hat{\omega}_{\Gamma,V}+\nabla\hat{\phi}_{\mathrm{C}}\cdot V-\frac{\zeta_{\Gamma,V}}{2}\right)\left(\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right)\\
&+B''(\xi_t)\left(\hat{\omega}_{\Gamma,V}+\nabla\hat{\phi}_{\mathrm{C}}\cdot V-\frac{\zeta_{\Gamma,V}}{2}\right)\left(\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right)\\
\leq\,&-B''\left(\xi_t\right)\left(\frac{\hat{\phi}_{\mathrm{C}}\circ T_t-\hat{\phi}_{\mathrm{C}}}{t}-\nabla\hat{\phi}_{\mathrm{C}}\cdot V-\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{2t}+\frac{\zeta_{\Gamma,V}}{2}\right)\\
&\cdot\left(\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right)\\
&+B'''(\sigma_t)\left(\psi_{\mathrm{r}}+\hat{\phi}_{\mathrm{C}}-\frac{\phi_{\Gamma,\infty}}{2}-\xi_t\right)\left(\hat{\omega}_{\Gamma,V}+\nabla\hat{\phi}_{\mathrm{C}}\cdot V-\frac{\zeta_{\Gamma,V}}{2}\right)\left(\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right)\\
\leq\,&C\left(\left|\frac{\hat{\phi}_{\mathrm{C}}\circ T_t-\hat{\phi}_{\mathrm{C}}}{t}-\nabla\hat{\phi}_{\mathrm{C}}\cdot V\right|+\left|\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right|\right)\left|\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right|\\
&+C\left(\left|\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}\right|+\left|\hat{\phi}_{\mathrm{C}}\circ T_t-\hat{\phi}_{\mathrm{C}}\right|+\left|\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}\right|\right)\left|\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right|,
\end{aligned}
$$

where $\xi_t$ and $\sigma_t$ are in between $(\psi_{\mathrm{r},t}+\hat{\phi}_{\mathrm{C}}-\phi_{\Gamma_t,\infty}/2)\circ T_t$ and $\psi_{\mathrm{r}}+\hat{\phi}_{\mathrm{C}}-\phi_{\Gamma,\infty}/2$ at each point in $\Omega_+$. Now, combining this inequality and the identity (4.37) with $\eta=(\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}})/t-\hat{\omega}_{\Gamma,V}\in H_0^1(\Omega)$, we obtain by the Poincaré and Cauchy–Schwarz inequalities and rearranging terms that

$$
\begin{aligned}
&\left\|\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}-\hat{\omega}_{\Gamma,V}\right\|_{H^1(\Omega)}^2\\
&\quad\leq C\int_\Omega\left|\left(\frac{A_V(t)-I}{t}\right)\nabla(\psi_{\mathrm{r},t}\circ T_t)-A_V'(0)\nabla\psi_{\mathrm{r}}\right|^2dX\\
&\qquad+C\int_{\Omega_+}\left|B'\left(\left(\psi_{\mathrm{r},t}+\hat{\phi}_{\mathrm{C}}-\frac{\phi_{\Gamma_t,\infty}}{2}\right)\circ T_t\right)\left(\frac{J_t-1}{t}\right)\right.\\
&\qquad\quad\left.-B'\left(\psi_{\mathrm{r}}+\hat{\phi}_{\mathrm{C}}-\frac{\phi_{\Gamma,\infty}}{2}\right)(\nabla\cdot V)\right|^2dX\\
&\qquad+C\left(\left\|\frac{\hat{\phi}_{\mathrm{C}}\circ T_t-\hat{\phi}_{\mathrm{C}}}{t}-\nabla\hat{\phi}_{\mathrm{C}}\cdot V\right\|_{H^1(\Omega_+)}^2+\left\|\frac{\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}}{t}-\zeta_{\Gamma,V}\right\|_{L^2(\Omega_+)}^2\right.\\
&\qquad\left.+\|\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}\|_{L^2(\Omega_+)}^2+\|\hat{\phi}_{\mathrm{C}}\circ T_t-\hat{\phi}_{\mathrm{C}}\|_{L^2(\Omega_+)}^2+\|\phi_{\Gamma_t,\infty}\circ T_t-\phi_{\Gamma,\infty}\|_{L^2(\Omega_+)}^2\right)\\
&\qquad+C\int_{\Omega_+}\left|\left(\frac{A_V(t)-I}{t}\right)\nabla(\hat{\phi}_{\mathrm{C}}\circ T_t)-A_V'(0)\nabla\hat{\phi}_{\mathrm{C}}\right|^2dX\\
&\quad=C\left[S_1(t)+S_2(t)+S_3(t)+S_4(t)\right].
\end{aligned}
\tag{4.38}
$$

It follows from (4.6)–(4.8), Lemma 4.1 (with $\hat{\phi} = \hat{\phi}_{\rm C}$), and (4.28) that

$$\begin{aligned} S_1(t) &= \int_\Omega \left| \left[ \frac{A_V(t) - I}{t} \right] \nabla(\psi_{{\rm r},t} \circ T_t) - A_V'(0) \nabla \psi_{\rm r} \right|^2 dX \\ &\le 2 \int_\Omega \left| \left[ \frac{A_V(t) - I}{t} - A_V'(0) \right] \nabla(\psi_{{\rm r},t} \circ T_t) \right|^2 dX + 2 \int_\Omega |A_V'(0) \nabla(\psi_{{\rm r},t} \circ T_t - \psi_{\rm r})|^2 \, dX \\ &\to 0 \qquad \text{as } t \to 0. \end{aligned} \tag{4.39}$$

By the uniform boundedness of $\psi_{{\rm r},t}$ and $\phi_{\Gamma_t,\infty}$ (cf. (2.5), (2.18), (2.23)) the Mean-Value Theorem, (4.2) and (4.3), Lemmas 4.1 and 4.2, and (4.28), we have

$$\begin{aligned} S_2(t) &= \int_{\Omega_+} \left| B' \left( \left( \psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2} \right) \circ T_t \right) \left( \frac{J_t - 1}{t} \right) - B' \left( \psi_{\rm r} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma,\infty}}{2} \right) (\nabla \cdot V) \right|^2 dX \\ &\le 2 \int_{\Omega_+} \left| B' \left( \left( \psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2} \right) \circ T_t \right) \left( \frac{J_t - 1}{t} - \nabla \cdot V \right) \right|^2 dX \\ &\quad + 2 \int_{\Omega_+} \left| B' \left( \left( \psi_{{\rm r},t} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma_t,\infty}}{2} \right) \circ T_t \right) - B' \left( \psi_{\rm r} + \hat{\phi}_{\rm C} - \frac{\phi_{\Gamma,\infty}}{2} \right) \right|^2 |\nabla \cdot V|^2 dX \\ &\le C \int_{\Omega_+} \left| \frac{J_t - 1}{t} - \nabla \cdot V \right|^2 dX \\ &\quad + C \int_{\Omega_+} (|\psi_{{\rm r},t} \circ T_t - \psi_{\rm r}|^2 + |\hat{\phi}_{\rm C} \circ T_t - \hat{\phi}_{\rm C}|^2 + |\phi_{\Gamma_t,\infty} \circ T_t - \phi_{\Gamma,\infty}|^2) \, dX \\ &\to 0 \qquad \text{as } t \to 0. \end{aligned} \tag{4.40}$$

By Lemma 4.1 (with $\hat{\phi} = \hat{\phi}_{\rm C}$), Lemma 4.2, and (4.28), we have

$$\begin{aligned} S_3(t) &= \left\| \frac{\hat{\phi}_{\rm C} \circ T_t - \hat{\phi}_{\rm C}}{t} - \nabla \hat{\phi}_{\rm C} \cdot V \right\|_{H^1(\Omega_+)}^2 + \left\| \frac{\phi_{\Gamma_t,\infty} \circ T_t - \phi_{\Gamma,\infty}}{t} - \zeta_{\Gamma,V} \right\|_{L^2(\Omega_+)} \\ &\quad + \|\psi_{{\rm r},t} \circ T_t - \psi_{\rm r}\|_{L^2(\Omega_+)} + \|\hat{\phi}_{\rm C} \circ T_t - \hat{\phi}_{\rm C}\|_{L^2(\Omega_+)}^2 + \|\phi_{\Gamma_t,\infty} \circ T_t - \phi_{\Gamma,\infty}\|_{L^2(\Omega_+)}^2 \\ &\quad \to 0 \qquad \text{as } t \to 0. \end{aligned} \tag{4.41}$$

It follows from (4.6)–(4.8) and Lemma 4.1 (with $\hat{\phi} = \hat{\phi}_{\rm C}$) that

$$\begin{aligned} S_4(t) &= \int_{\Omega_+} \left| \left[ \frac{A_V(t) - I}{t} \right] \nabla(\hat{\phi}_{\rm C} \circ T_t) - A_V'(0) \nabla \hat{\phi}_{\rm C} \right|^2 dX \\ &\le C \int_{\Omega_+} \left| \left[ \frac{A_V(t) - I}{t} - A_V'(0) \right] \nabla(\hat{\phi}_{\rm C} \circ T_t) \right|^2 + C \int_{\Omega_+} |\nabla(\hat{\phi}_{\rm C} \circ T_t - \hat{\phi}_{\rm C})|^2 dX \\ &\to 0 \qquad \text{as } t \to 0. \end{aligned} \tag{4.42}$$

Now the desired convergence (4.35) follows from (4.38)–(4.42). □

# 5 Proof of Theorem 3.2

*Proof of Theorem 3.2.* Fix $V \in \mathcal{V}$ (cf. (3.6)). Let $\{T_t\}_{t\in\mathbb{R}}$ be the family of diffeomorphisms from $\mathbb{R}^3$ to $\mathbb{R}^3$ defined by $T_t(X) = x(t, X)$ as the solution to the initial-value problem (3.7). We proceed in five steps. In Step 1, we calculate the limit as $t \to 0$ that defines the variation $\delta_{\Gamma,V}E[\Gamma]$; cf. Definition 3.1. In Step 2, we simplify the expression of $\delta_{\Gamma,V}E[\Gamma]$. In Step 3, we convert all the volume integrals in $\delta_{\Gamma,V}E[\Gamma]$ into surface integrals on the boundary $\Gamma$, except one volume integral that involves the $B'$ term. In Step 4, we rewrite the surface integrals to have the desired form (i.e., with a factor $V \cdot n$ in the integrand). Finally, in Step 5, we treat the only volume integral term that involves $B'$ to get the desired formula.

*Step 1.* Let $t \in \mathbb{R}$. We recall that $\phi_{\Gamma_t,\infty}$, $\hat{\phi}_{\Gamma_t,\infty}$, and $\psi_{\Gamma_t}$ are the solutions to (2.3), (2.13), and (2.22) with $\Gamma_t = T_t(\Gamma)$ replacing $\Gamma$, respectively, and that all these functions have the boundary value $\phi_\infty$ on $\partial\Omega$. Recall that $\hat{\phi}_0$ and $\hat{\phi}_\infty$ are defined by (2.9) and (2.10). We denote in this proof

$$\psi_{\mathrm{r}} = \psi_\Gamma - \hat{\phi}_{\Gamma,\infty} \quad \text{and} \quad \psi_{\mathrm{r},t} = \psi_{\Gamma_t} - \hat{\phi}_{\Gamma_t,\infty}. \tag{5.1}$$

By (3.2) with $\Gamma_t$ replacing $\Gamma$, the definition of $A_V(t)$ (4.4) and $J_t$ (4.1), and the change of variable $x = T_t(X)$, we have

$$\begin{aligned}
E[\Gamma_t] &= -\int_\Omega \frac{\varepsilon_{\Gamma_t}}{2}|\nabla\psi_{\mathrm{r},t}|^2 dx - \int_{T_t(\Omega_+)} B\left(\psi_{\Gamma_t} - \frac{\phi_{\Gamma_t,\infty}}{2}\right) dx \\
&\quad + \frac{\varepsilon_- - \varepsilon_+}{2}\int_{T_t(\Omega_+)} \nabla\hat{\phi}_{\Gamma_t,\infty}\cdot\nabla\hat{\phi}_0\, dx + W \\
&= -\int_\Omega \frac{\varepsilon_\Gamma}{2}\left[A_V(t)\nabla(\psi_{r,t}\circ T_t)\cdot\nabla(\psi_{r,t}\circ T_t)\right] dX \\
&\quad - \int_{\Omega_+} B\left(\left(\psi_{\Gamma_t} - \frac{\phi_{\Gamma_t,\infty}}{2}\right)\circ T_t\right) J_t\, dX \\
&\quad + \frac{\varepsilon_- - \varepsilon_+}{2}\int_{\Omega_+} A_V(t)\nabla(\hat{\phi}_{\Gamma_t,\infty}\circ T_t)\cdot\nabla(\hat{\phi}_0\circ T_t)\, dX + W,
\end{aligned}$$

where $W = (1/2)\sum_{i=1}^N Q_i(\hat{\phi}_\infty - \hat{\phi}_{\mathrm{C}})(x_i)$ is independent of $\Gamma$.

By the definition of $\delta_{\Gamma,V}E[\Gamma]$ (cf. Definition 3.1), we need to calculate $(d/dt)|_{t=0}E[\Gamma_t]$. This amounts to justifying the interchange of the differentiation against $t$ and the integration over $\Omega$, and then applying the product and chain rules of differentiation. Since the differentiation against $t$ of functions $\phi_{\Gamma_t,\infty}$, $\hat{\phi}_{\Gamma_t,\infty}$, and $\psi_{\Gamma_t}$ are defined using the $H^1(\Omega)$-norm applied to the corresponding quotients (cf. Lemmas 4.2–4.4), we proceed with the limit $t \to 0$ of $(E[\Gamma_t] - E[\Gamma])/t$ and apply the results from those lemmas.

It follows from (3.2) and the above expression of $E[\Gamma_t]$ that

$$\begin{aligned}
\frac{E[\Gamma_t] - E[\Gamma]}{t} &= -\int_\Omega \frac{\varepsilon_\Gamma}{2t}\left[A_V(t)\nabla(\psi_{\mathrm{r},t}\circ T_t)\cdot\nabla(\psi_{\mathrm{r},t}\circ T_t) - \nabla\psi_{\mathrm{r}}\cdot\nabla\psi_{\mathrm{r}}\right] dX \\
&\quad - \int_{\Omega_+}\frac{1}{t}\left[B\left(\left(\psi_{\Gamma_t} - \frac{\phi_{\Gamma_t,\infty}}{2}\right)\circ T_t\right) J_t - B\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\right] dX
\end{aligned}$$

$$+\frac{\varepsilon_- - \varepsilon_+}{2}\int_{\Omega_+}\frac{1}{t}\left[A_V(t)\nabla(\hat{\phi}_{\Gamma_t,\infty}\circ T_t)\cdot\nabla(\hat{\phi}_0\circ T_t)-\nabla\hat{\phi}_{\Gamma,\infty}\cdot\nabla\hat{\phi}_0\right]dX$$
$$=-\delta_1(t)-\delta_2(t)+\frac{\varepsilon_- - \varepsilon_+}{2}\delta_3(t). \tag{5.2}$$

By rearranging the terms, we obtain that

$$\begin{aligned}\delta_1(t)=&\int_\Omega\frac{\varepsilon_\Gamma}{2}\left[\frac{A_V(t)-I-tA_V'(0)}{t}\right]\nabla(\psi_{\mathrm{r},t}\circ T_t)\cdot\nabla(\psi_{\mathrm{r},t}\circ T_t)\,dX\\&+\int_\Omega\frac{\varepsilon_\Gamma}{2}A_V'(0)\nabla(\psi_{\mathrm{r},t}\circ T_t)\cdot\nabla(\psi_{\mathrm{r},t}\circ T_t)\,dX\\&+\int_\Omega\frac{\varepsilon_\Gamma}{2}\left[\nabla(\psi_{\mathrm{r},t}\circ T_t)+\nabla\psi_{\mathrm{r}}\right]\cdot\nabla\left(\frac{\psi_{\mathrm{r},t}\circ T_t-\psi_{\mathrm{r}}}{t}\right)dX.\end{aligned}$$

It thus follows from (4.7), (4.8), Lemma 4.3, and Lemma 4.4 that

$$\lim_{t\to0}\delta_1(t)=\int_\Omega\varepsilon_\Gamma\left[\frac{1}{2}A_V'(0)\nabla\psi_{\mathrm{r}}\cdot\nabla\psi_{\mathrm{r}}+\nabla\psi_{\mathrm{r}}\cdot\nabla(\omega_{\Gamma,V}-\xi_{\Gamma,V})\right]dX, \tag{5.3}$$

where $\xi_{\Gamma,V}$ and $\omega_{\Gamma,V}$ are defined in (4.22) in Lemma 4.3 and (4.24) in Lemma 4.4, respectively.

Denote $q=\psi_\Gamma-\phi_{\Gamma,\infty}/2$ and $q_t=(\psi_{\Gamma_t}-\phi_{\Gamma_t,\infty}/2)\circ T_t$. The second term $\delta_2(t)$ in (5.2) can be written as

$$\delta_2(t)=\int_{\Omega_+}\frac{J_t-1}{t}B(q_t)\,dX+\int_{\Omega_+}\frac{B(q_t)-B(q)}{t}\,dX. \tag{5.4}$$

It follows from Lemma 4.2 and Lemma 4.4 that $q_t\to q$ in $L^2(\Omega)$. Moreover, by (2.5), (2.18), and (2.23), the $L^\infty(\Omega)$-norm of $q_t$ is bounded uniformly in $t\in\mathbb{R}$. Hence, $B(q_t)\to B(q)$ in $L^2(\Omega_+)$ as $t\to0$. This, together with (4.2) and (4.3), implies that

$$\lim_{t\to0}\int_{\Omega_+}\frac{J_t-1}{t}B(q_t)\,dX=\int_{\Omega_+}(\nabla\cdot V)B(q)\,dX. \tag{5.5}$$

Now Taylor's expansion implies that

$$\begin{aligned}&\frac{B(q_t(X))-B(q(X))}{t}\\&\quad=B'(q(X))\frac{q_t(X)-q(X)}{t}+\frac{1}{2}B''(\eta_t(X))[q_t(X)-q(X)]\frac{q_t(X)-q(X)}{t},\quad\text{a.e. } X\in\Omega_+,\end{aligned}$$

where $\eta_t(X)$ is in between $q(X)$ and $q_t(X)$, and its $L^\infty(\Omega)$-norm is bounded uniformly in $t$. It then follows from Lemma 4.2 and Lemma 4.4 that

$$\left|\int_{\Omega_+}B''(\eta_t)(q_t-q)\frac{q_t-q}{t}\,dX\right|\le C\|q_t-q\|_{L^2(\Omega_+)}\left\|\frac{q_t-q}{t}\right\|_{L^2(\Omega_+)}\to0\qquad\text{as } t\to0,$$

where $C$ is a constant independent of $t$. Consequently, Lemma 4.2 and Lemma 4.4 imply that

$$\lim_{t\to 0}\int_{\Omega_+}\frac{B(q_t)-B(q)}{t}\,dX=\lim_{t\to 0}\int_{\Omega_+}B'(q)\frac{q_t-q}{t}\,dX=\int_{\Omega_+}B'(q)\left(\omega_{\Gamma,V}-\frac{\zeta_{\Gamma,V}}{2}\right)dX,$$

where $\omega_{\Gamma,V}$ and $\zeta_{\Gamma,V}$ are given in (4.24) and (4.15), respectively. This, together with (5.4) and (5.5), and our definition of $q$ and $q_t$, implies that

$$\lim_{t\to 0}\delta_2(t)=\int_{\Omega_+}\left[(\nabla\cdot V)B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)+B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)\left(\omega_{\Gamma,V}-\frac{\zeta_{\Gamma,V}}{2}\right)\right]dX. \tag{5.6}$$

Rearranging the terms, we have

$$\begin{aligned}\delta_3(t)=&\int_{\Omega_+}\frac{A_V(t)-I-tA_V'(0)}{t}\nabla(\hat\phi_{\Gamma_t,\infty}\circ T_t)\cdot\nabla(\hat\phi_0\circ T_t)\,dX\\&+\int_{\Omega_+}\frac{\nabla(\hat\phi_{\Gamma_t,\infty}\circ T_t)-\nabla\hat\phi_{\Gamma,\infty}}{t}\cdot\nabla(\hat\phi_0\circ T_t)\,dx\\&+\int_{\Omega_+}\nabla\hat\phi_{\Gamma,\infty}\cdot\frac{\nabla(\hat\phi_0\circ T_t)-\nabla\hat\phi_0}{t}\,dX\\&+\int_{\Omega_+}A_V'(0)\nabla(\hat\phi_{\Gamma_t,\infty}\circ T_t)\cdot\nabla(\hat\phi_0\circ T_t)\,dX.\end{aligned}$$

Therefore, we have by (4.7), (4.8), Lemma 4.1 (with $\hat\phi=\hat\phi_0$), and Lemma 4.3 that

$$\lim_{t\to 0}\delta_3(t)=\int_{\Omega_+}\left[\nabla\xi_{\Gamma,V}\cdot\nabla\hat\phi_0+\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\hat\phi_0\right]dX. \tag{5.7}$$

It now follows from Definition 3.1, (5.2), (5.3), (5.6), and (5.7) that the first variation $\delta_\Gamma E[\Gamma]$ exists and is given by

$$\begin{aligned}\delta_{\Gamma,V}E[\Gamma]=&\underbrace{-\int_\Omega\frac{\varepsilon_\Gamma}{2}A_V'(0)\nabla\psi_{\rm r}\cdot\nabla\psi_{\rm r}\,dX}_{M_1}+\underbrace{\int_\Omega\varepsilon_\Gamma\nabla\psi_{\rm r}\cdot\nabla\xi_{\Gamma,V}\,dX}_{M_2}\underbrace{-\int_\Omega\varepsilon_\Gamma\nabla\psi_{\rm r}\cdot\nabla\omega_{\Gamma,V}\,dX}_{M_3}\\&\underbrace{-\int_{\Omega_+}\left[(\nabla\cdot V)B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)+B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)\left(\omega_{\Gamma,V}-\frac{\zeta_{\Gamma,V}}{2}\right)\right]dX}_{M_4}\\&\underbrace{+\frac{\varepsilon_--\varepsilon_+}{2}\int_{\Omega_+}\nabla\xi_{\Gamma,V}\cdot\nabla\hat\phi_0\,dX}_{M_5}\\&\underbrace{+\frac{\varepsilon_--\varepsilon_+}{2}\int_{\Omega_+}\left[\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\hat\phi_0\right]dX}_{M_6}\end{aligned}$$

$$= M_1 + M_2 + M_3 + M_4 + M_5 + M_6. \tag{5.8}$$

*Step 2.* We now simplify this expression. By Lemma 4.3, our notation $\psi_{\rm r} = \psi_\Gamma - \hat{\phi}_{\Gamma,\infty}$, and the fact that the support of $V$ or $A_V'(0)$ contains no singularities $x_i$ $(1 \le i \le N)$, we can express the sum of the first two integrals above as

$$\begin{aligned} M_1 + M_2 &= -\int_\Omega \frac{\varepsilon_\Gamma}{2} A_V'(0) \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty}) \cdot \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})\, dX \\ &\quad - \int_\Omega \varepsilon_\Gamma A_V'(0) \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})\, dX \\ &= -\int_\Omega \frac{\varepsilon_\Gamma}{2} A_V'(0) \nabla\psi_\Gamma \cdot \nabla\psi_\Gamma\, dX + \int_\Omega \frac{\varepsilon_\Gamma}{2} A_V'(0) \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla\hat{\phi}_{\Gamma,\infty}\, dX. \end{aligned} \tag{5.9}$$

Note that the last two integrals exist as the singularities $x_i$ $(1 \le i \le N)$ of $\psi_\Gamma$ and $\hat{\phi}_{\Gamma,\infty}$ are outside the support of $V$ and $A_V'(0)$ is given in (4.6). By (2.22) and (2.13), we have

$$\int_\Omega \left[ \varepsilon_\Gamma \nabla\psi_r \cdot \nabla\eta + \chi_+ B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\eta \right] dX = 0$$

for all $\eta \in C_{\rm c}^1(\Omega)$ and hence all $\eta \in H_0^1(\Omega)$. Setting $\eta = \omega_{\Gamma,V}$, we get the two-$\omega_{\Gamma,V}$ terms in (5.8) (one is $M_3$ and the other is part of $M_4$) cancelled:

$$\int_\Omega \left[ \varepsilon_\Gamma \nabla\psi_{\rm r} \cdot \nabla\omega_{\Gamma,V} + \chi_+ B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)\omega_{\Gamma,V} \right] dX = 0. \tag{5.10}$$

To simplify $M_5$, we note that we can replace $\eta$ in (2.12) (with $\hat{\phi} = \hat{\phi}_0$) and (2.13) by $\xi_{\Gamma,V} \in H_0^1(\Omega)$, as $\xi_{\Gamma,V}|_{\Omega_-} \in C^2(\Omega_-)$; cf. the remark six lines below (2.12) and that below (2.14). It then follows that

$$\begin{aligned} M_5 &= \frac{\varepsilon_-}{2} \int_{\Omega_+} \nabla\xi_{\Gamma,V} \cdot \nabla\hat{\phi}_0\, dX - \frac{\varepsilon_+}{2} \int_{\Omega_+} \nabla\xi_{\Gamma,V} \cdot \nabla\hat{\phi}_0\, dX \\ &= -\int_\Omega \frac{\varepsilon_\Gamma}{2} \nabla\xi_{\Gamma,V} \cdot \nabla\hat{\phi}_0\, dX + \frac{1}{2}\sum_{i=1}^N Q_i \xi_{\Gamma,V}(x_i) \quad \text{[by (2.12) with } \hat{\phi} = \hat{\phi}_0\text{]} \\ &= -\int_\Omega \frac{\varepsilon_\Gamma}{2} \nabla\xi_{\Gamma,V} \cdot \nabla\hat{\phi}_0\, dX + \int_\Omega \frac{\varepsilon_\Gamma}{2} \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla\xi_{\Gamma,V}\, dX \quad \text{[by (2.13)]} \\ &= \int_\Omega \frac{\varepsilon_\Gamma}{2} \nabla\xi_{\Gamma,V} \cdot \nabla(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_0 - \phi_{\Gamma,\infty})\, dX \quad \text{[by (2.3)]} \\ &= -\int_\Omega \frac{\varepsilon_\Gamma}{2} A_V'(0) \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla(\hat{\phi}_{\Gamma,\infty} - \hat{\phi}_0 - \phi_{\Gamma,\infty})\, dX. \quad \text{[by Lemma 4.3]} \end{aligned} \tag{5.11}$$

Since $\hat{\phi}_0$ is harmonic in the support of $V$ that excludes all $x_i$ $(i = 1, \dots, N)$, we have by the same calculations as in (4.20) that $\nabla \cdot \left[\nabla(\nabla\hat{\phi}_0 \cdot V) + A_V'(0)\nabla\hat{\phi}_0\right] = 0$ in $\Omega$. Thus, since

the normal $n$ along $\Gamma$ points from $\Omega_-$ to $\Omega_+$, we have by the Divergence Theorem that

$$\begin{aligned}
&\frac{\varepsilon_-}{2}\int_{\Omega_+}\nabla\hat\phi_{\Gamma,\infty}\cdot[\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_0]\,dX\\
&\quad=-\frac{\varepsilon_-}{2}\int_{\Gamma}\hat\phi_{\Gamma,\infty}[\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_0]\cdot n\,dS\\
&\quad=-\frac{\varepsilon_-}{2}\int_{\Omega_-}\nabla\hat\phi_{\Gamma,\infty}\cdot[\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_0]\,dX.
\end{aligned}$$

Therefore, since $A_V'(0)$ (cf. (4.6)) is symmetric,

$$\begin{aligned}
M_6&=\frac{\varepsilon_--\varepsilon_+}{2}\int_{\Omega_+}\nabla\hat\phi_{\Gamma,\infty}\cdot[\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_0]\,dX\\
&=-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}[\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla(\nabla\hat\phi_0\cdot V)+A_V'(0)\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\hat\phi_0]\,dX\\
&=-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}A_V'(0)\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\hat\phi_0\,dX.\quad[\text{by (2.13)}]
\end{aligned}\tag{5.12}$$

It now follows from (5.8)–(5.12) that

$$\begin{aligned}
\delta_{\Gamma,V}E[\Gamma]&=\underbrace{-\int_{\Omega}\frac{\varepsilon_\Gamma}{2}A_V'(0)\nabla\psi_\Gamma\cdot\nabla\psi_\Gamma\,dX}_{P_1}+\underbrace{\int_{\Omega}\frac{\varepsilon_\Gamma}{2}A_V'(0)\nabla\hat\phi_{\Gamma,\infty}\cdot\nabla\phi_{\Gamma,\infty}\,dX}_{P_2}\\
&\quad+\underbrace{\int_{\Omega_+}\left[\frac{\zeta_{\Gamma,V}}{2}B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)-(\nabla\cdot V)B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)\right]dX}_{P_3}\\
&=P_1+P_2+P_3.
\end{aligned}\tag{5.13}$$

*Step 3.* We convert most of these volume integrals into surface integrals on $\Gamma$. We shall use the following identities that can be verified by using the Divergence Theorem and approximations by smooth functions:

$$\int_D(\nabla\cdot U)\nabla a\cdot\nabla b\,dx=-\int_D U\cdot(\nabla^2a\nabla b+\nabla^2b\nabla a)\,dx+\int_{\partial D}(\nabla a\cdot\nabla b)(U\cdot\nu)\,dx,\tag{5.14}$$

$$\int_D(\nabla U)\nabla a\cdot\nabla b\,dx=-\int_D U\cdot(\Delta a\nabla b+\nabla^2b\nabla a)\,dx+\int_{\partial D}(\nabla a\cdot\nu)(\nabla b\cdot U)\,dx.\tag{5.15}$$

Here, $D\subset\mathbb{R}^3$ is a bounded open set with a $C^1$ boundary $\partial D$, $U\in H^1(D,\mathbb{R}^3)$, $a,b\in H^2(D)$, $\nabla^2a$ is the Hessian matrix of $a$, and $\nu$ is the unit exterior normal at the boundary $\partial D$. If in addition $\Delta a=\Delta b=0$ in $D$, then we have by (5.14) and (5.15) that

$$\int_D(\nabla U+(\nabla U)^T-(\nabla\cdot U)I)\nabla a\cdot\nabla b\,dx$$

$$= \int_{\partial D} [(\nabla a \cdot U) \cdot (\nabla b \cdot \nu) + (\nabla b \cdot U) \cdot (\nabla a \cdot \nu) - (\nabla a \cdot \nabla b) \cdot (U \cdot \nu)] \, dS. \tag{5.16}$$

Note that $V = 0$ in a neighborhood of all $x_i$ $(1 \le i \le N)$ and $V = 0$ on $\partial\Omega$ and that the unit normal vector $n$ on $\Gamma$ points from $\Omega_-$ to $\Omega_+$. By Theorem 2.1, $\Delta\psi_\Gamma = 0$ on $\Omega_- \cap \text{supp}\,(V)$ and $\varepsilon_+ \Delta\psi_\Gamma = B'(\psi_\Gamma - \phi_{\Gamma,\infty}/2)$ on $\Omega_+$. Therefore, writing $u^s = u|_{\Omega_s}$ $(s = +, -)$, we have by (4.6), (5.14), and (5.15) that

$$\begin{aligned}
P_1 &= \int_\Omega \frac{\varepsilon_\Gamma}{2} [\nabla V + (\nabla V)^T - (\nabla \cdot V) I] \nabla\psi_\Gamma \cdot \nabla\psi_\Gamma \, dX \\
&= \int_{\Omega_-} \varepsilon_- (\nabla V) \nabla\psi_\Gamma \cdot \nabla\psi_\Gamma \, dX + \int_{\Omega_+} \varepsilon_+ (\nabla V) \nabla\psi_\Gamma \cdot \nabla\psi_\Gamma \, dX \\
&\quad - \int_{\Omega_-} \frac{\varepsilon_-}{2} (\nabla \cdot V) \nabla\psi_\Gamma \cdot \nabla\psi_\Gamma \, dX - \int_{\Omega_+} \frac{\varepsilon_+}{2} (\nabla \cdot V) \nabla\psi_\Gamma \cdot \nabla\psi_\Gamma \, dX \\
&= - \int_{\Omega_-} \varepsilon_- V \cdot (\Delta\psi_\Gamma \nabla\psi_\Gamma + \nabla^2\psi_\Gamma \nabla\psi_\Gamma) \, dX + \int_\Gamma \varepsilon_- (\nabla\psi_\Gamma^- \cdot V)(\nabla\psi_\Gamma^- \cdot n) \, dS \\
&\quad - \int_{\Omega_+} \varepsilon_+ V \cdot (\Delta\psi_\Gamma \nabla\psi_\Gamma + \nabla^2\psi_\Gamma \nabla\psi_\Gamma) \, dX - \int_\Gamma \varepsilon_+ (\nabla\psi_\Gamma^+ \cdot V)(\psi_\Gamma^+ \cdot n) \, dS \\
&\quad + \int_{\Omega_-} \varepsilon_- V \cdot \nabla^2\psi_\Gamma \nabla\psi_\Gamma \, dX - \int_\Gamma \frac{\varepsilon_-}{2} |\nabla\psi_\Gamma^-|^2 (V \cdot n) \, dS \\
&\quad + \int_{\Omega_+} \varepsilon_+ V \cdot \nabla^2\psi_\Gamma \nabla\psi_\Gamma \, dX + \int_\Gamma \frac{\varepsilon_+}{2} |\nabla\psi_\Gamma^+|^2 (V \cdot n) \, dS \\
&= - \int_{\Omega_-} \varepsilon_- \Delta\psi_\Gamma (\nabla\psi_\Gamma \cdot V) \, dX + \int_\Gamma \varepsilon_- (\nabla\psi_\Gamma^- \cdot V)(\nabla\psi_\Gamma^- \cdot n) \, dS - \int_\Gamma \frac{\varepsilon_-}{2} |\nabla\psi_\Gamma^-|^2 (V \cdot n) \, dS \\
&\quad - \int_{\Omega_+} \varepsilon_+ \Delta\psi_\Gamma (\nabla\psi_\Gamma \cdot V) \, dX - \int_\Gamma \varepsilon_+ (\nabla\psi_\Gamma^+ \cdot V)(\nabla\psi_\Gamma^+ \cdot n) \, dS + \int_\Gamma \frac{\varepsilon_+}{2} |\nabla\psi_\Gamma^+|^2 (V \cdot n) \, dS \\
&= \int_\Gamma \varepsilon_- (\nabla\psi_\Gamma^- \cdot V)(\nabla\psi_\Gamma^- \cdot n) \, dS - \int_\Gamma \varepsilon_+ (\nabla\psi_\Gamma^+ \cdot V)(\nabla\psi_\Gamma^+ \cdot n) \, dS \\
&\quad - \int_\Gamma \frac{\varepsilon_-}{2} |\nabla\psi_\Gamma^-|^2 (V \cdot n) \, dS + \int_\Gamma \frac{\varepsilon_+}{2} |\nabla\psi_\Gamma^+|^2 (V \cdot n) \, dS \\
&\quad - \int_{\Omega_+} B' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) (\nabla\psi_\Gamma \cdot V) \, dX,
\end{aligned} \tag{5.17}$$

where a superscript $-$ or $+$ denotes the restriction to $\Omega_-$ or $\Omega_+$, respectively.

Since $\hat{\phi}_{\Gamma,\infty}$ and $\phi_{\Gamma,\infty}$ are harmonic in $\Omega_- \cap \text{supp}\,(V)$ and $\Omega_+$, and since the normal $n$ points from $\Omega_-$ to $\Omega_+$, we have by (4.6), (5.16), and the notation of jumps (2.8) that

$$\begin{aligned}
P_2 = &\int_{\Omega_-} \frac{\varepsilon_-}{2} [(\nabla \cdot V) I - \nabla V - (\nabla V)^T] \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla\phi_{\Gamma,\infty} \, dX \\
&+ \int_{\Omega_+} \frac{\varepsilon_+}{2} [(\nabla \cdot V) I - \nabla V - (\nabla V)^T] \nabla\hat{\phi}_{\Gamma,\infty} \cdot \nabla\phi_{\Gamma,\infty} \, dX
\end{aligned}$$

$$= \frac{1}{2}\int_\Gamma [\![\varepsilon_\Gamma(\nabla\hat{\phi}_{\Gamma,\infty}\cdot V)(\nabla\phi_{\Gamma,\infty}\cdot n)+\varepsilon_\Gamma(\nabla\hat{\phi}_{\Gamma,\infty}\cdot n)(\nabla\phi_{\Gamma,\infty}\cdot V)$$
$$-\varepsilon_\Gamma(\nabla\hat{\phi}_{\Gamma,\infty}\cdot\nabla\phi_{\Gamma,\infty})(V\cdot n)]\!]_\Gamma\, dS. \tag{5.18}$$

Using the Divergence Theorem and noting again that the normal $n$ at $\Gamma$ points from $\Omega_-$ to $\Omega_+$, we obtain

$$\begin{aligned}
P_3 &= \int_{\Omega_+}\frac{\zeta_{\Gamma,V}}{2}B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dX+\int_{\Omega_+}V\cdot B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)\left(\nabla\psi_\Gamma-\frac{\nabla\phi_{\Gamma,\infty}}{2}\right)dX\\
&\quad+\int_\Gamma B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)(V\cdot n)\,dS\\
&= \int_{\Omega_+}\left[\frac{1}{2}\left(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V\right)+\nabla\psi_\Gamma\cdot V\right]B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dX\\
&\quad+\int_\Gamma B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)(V\cdot n)\,dS.
\end{aligned}\tag{5.19}$$

It now follows from (5.13) and (5.17)–(5.19) that

$$\begin{aligned}
\delta_{\Gamma,V}E[\Gamma] &= \int_\Gamma\varepsilon_-(\nabla\psi_\Gamma^-\cdot V)(\nabla\psi_\Gamma^-\cdot n)\,dS-\int_\Gamma\varepsilon_+(\nabla\psi_\Gamma^+\cdot V)(\nabla\psi_\Gamma^+\cdot n)\,dS\\
&\quad-\int_\Gamma\frac{\varepsilon_-}{2}|\nabla\psi_\Gamma^-|^2(V\cdot n)\,dS+\int_\Gamma\frac{\varepsilon_+}{2}|\nabla\psi_\Gamma^+|^2(V\cdot n)\,dS\\
&\quad+\frac{1}{2}\int_\Gamma[\![\varepsilon_\Gamma(\nabla\hat{\phi}_{\Gamma,\infty}\cdot V)(\nabla\phi_{\Gamma,\infty}\cdot n)]\!]_\Gamma\,dS\\
&\quad+\frac{1}{2}\int_\Gamma[\![\varepsilon_\Gamma(\nabla\hat{\phi}_{\Gamma,\infty}\cdot n)(\nabla\phi_{\Gamma,\infty}\cdot V)]\!]_\Gamma\,dS\\
&\quad-\frac{1}{2}\int_\Gamma[\![\varepsilon_\Gamma(\nabla\hat{\phi}_{\Gamma,\infty}\cdot\nabla\phi_{\Gamma,\infty})(V\cdot n)]\!]_\Gamma\,dS\\
&\quad+\int_{\Omega_+}\frac{1}{2}\left(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V\right)B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dX\\
&\quad+\int_\Gamma B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)(V\cdot n)\,dS.
\end{aligned}\tag{5.20}$$

*Step 4.* We express the surface integrals into those with the factor $V\cdot n$ in the integrand. Note that on each side of $\Gamma$, we can write

$$\nabla\psi_\Gamma=(\nabla\psi_\Gamma\cdot n)n+\nabla_\Gamma\psi_\Gamma=\partial_n\psi_\Gamma n+\nabla_\Gamma\psi_\Gamma\quad\text{on }\Gamma,$$

where $\nabla_\Gamma\psi_\Gamma=(I-n\otimes n)\nabla\psi_\Gamma$ is the tangential derivative. Clearly $n\cdot\nabla_\Gamma\psi_\Gamma=0$. Moreover, $\nabla_\Gamma\psi_\Gamma^+=\nabla_\Gamma\psi_\Gamma^-$ on $\Gamma$. Thus, $\nabla\left(\psi_\Gamma^+-\psi_\Gamma^-\right)=(\partial_n\psi_\Gamma^+-\partial_n\psi_\Gamma^-)n$ on $\Gamma$. By Theorem 2.1, we have also $\varepsilon_+\nabla\psi_\Gamma^+\cdot n=\varepsilon_-\nabla\psi_\Gamma^-\cdot n=\varepsilon_\Gamma\nabla\psi_\Gamma\cdot n$ on $\Gamma$. Therefore, the first four terms in (5.20) are

$$\int_\Gamma\varepsilon_-(\nabla\psi_\Gamma^-\cdot V)(\nabla\psi_\Gamma^-\cdot n)\,dS-\int_\Gamma\varepsilon_+(\nabla\psi_\Gamma^+\cdot V)(\nabla\psi_\Gamma^+\cdot n)\,dS$$

$$
\begin{aligned}
&- \int_\Gamma \frac{\varepsilon_-}{2} |\nabla \psi_\Gamma^-|^2 (V \cdot n)\, dS + \int_\Gamma \frac{\varepsilon_+}{2} |\nabla \psi_\Gamma^+|^2 (V \cdot n)\, dS \\
&= - \int_\Gamma \varepsilon_\Gamma \partial_n \psi_\Gamma (\partial_n \psi_\Gamma^+ - \partial_n \psi_\Gamma^-)(V \cdot n)\, dS \\
&\quad + \int_\Gamma \frac{\varepsilon_+}{2} |\nabla \psi_\Gamma^+|^2 (V \cdot n)\, dS - \int_\Gamma \frac{\varepsilon_-}{2} |\nabla \psi_\Gamma^-|^2 (V \cdot n)\, dS \\
&= - \int_\Gamma \varepsilon_+ |\partial_n \psi_\Gamma^+|^2 (V \cdot n)\, dS + \int_\Gamma \varepsilon_- |\partial_n \psi_\Gamma^-|^2 (V \cdot n)\, dS \\
&\quad + \int_\Gamma \frac{\varepsilon_+}{2} |\partial_n \psi_\Gamma^+|^2 (V \cdot n)\, dS + \int_\Gamma \frac{\varepsilon_+}{2} |\nabla_\Gamma \psi_\Gamma|^2 (V \cdot n)\, dS \\
&\quad - \int_\Gamma \frac{\varepsilon_-}{2} |\partial_n \psi_\Gamma^-|^2 (V \cdot n)\, dS - \int_\Gamma \frac{\varepsilon_-}{2} |\nabla_\Gamma \psi_\Gamma|^2 (V \cdot n)\, dS \\
&= -\frac{1}{2} \left( \frac{1}{\varepsilon_+} - \frac{1}{\varepsilon_-} \right) \int_\Gamma |\varepsilon_\Gamma \partial_n \psi_\Gamma|^2 (V \cdot n)\, dS + \frac{\varepsilon_+ - \varepsilon_-}{2} \int_\Gamma |\nabla_\Gamma \psi_\Gamma|^2 (V \cdot n)\, dS. \qquad (5.21)
\end{aligned}
$$

Similarly, on each side of $\Gamma$, we have with $u_\Gamma = \phi_{\Gamma,\infty}$ or $\hat{\phi}_{\Gamma,\infty}$ that

$$
\begin{aligned}
\nabla u_\Gamma \cdot V &= (\partial_n u_\Gamma n + \nabla_\Gamma u_\Gamma) \cdot ((V \cdot n) n + (I - n \otimes n) V) \\
&= \partial_n u_\Gamma (V \cdot n) + \nabla_\Gamma u_\Gamma (I - n \otimes n) V.
\end{aligned}
$$

Moreover, $\varepsilon_+ \partial_n u_\Gamma^+ = \varepsilon_- \partial_n u_\Gamma^-$ and $\partial_\Gamma u_\Gamma^+ = \partial_\Gamma u_\Gamma^-$ on $\Gamma$. Therefore, the next three terms in (5.20) become

$$
\begin{aligned}
&\frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma (\nabla \hat{\phi}_{\Gamma,\infty} \cdot V)(\nabla \phi_{\Gamma,\infty} \cdot n) ]\!]_\Gamma \, dS + \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma (\nabla \hat{\phi}_{\Gamma,\infty} \cdot n)(\nabla \phi_{\Gamma,\infty} \cdot V) ]\!]_\Gamma \, dS \\
&\quad - \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma (\nabla \hat{\phi}_{\Gamma,\infty} \cdot \nabla \phi_{\Gamma,\infty})(V \cdot n) ]\!]_\Gamma \, dS \\
&= \int_\Gamma [\![ \varepsilon_\Gamma \partial_n \hat{\phi}_{\Gamma,\infty} \partial_n \phi_{\Gamma,\infty} ]\!]_\Gamma (V \cdot n)\, dS \\
&\quad - \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma (\partial_n \hat{\phi}_{\Gamma,\infty} \partial_n \phi_{\Gamma,\infty} + \nabla_\Gamma \hat{\phi}_{\Gamma,\infty} \cdot \nabla_\Gamma \phi_{\Gamma,\infty}) ]\!]_\Gamma (V \cdot n)\, dS \\
&= \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma \partial_n \hat{\phi}_{\Gamma,\infty} \partial_n \phi_{\Gamma,\infty} ]\!]_\Gamma (V \cdot n)\, dS - \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma \nabla_\Gamma \hat{\phi}_{\Gamma,\infty} \cdot \nabla_\Gamma \phi_{\Gamma,\infty} ]\!]_\Gamma (V \cdot n)\, dS. \qquad (5.22)
\end{aligned}
$$

It now follows from (5.20)–(5.22) that

$$
\begin{aligned}
\delta_{\Gamma,V} E[\Gamma] = &-\frac{1}{2} \left( \frac{1}{\varepsilon_+} - \frac{1}{\varepsilon_-} \right) \int_\Gamma |\varepsilon_\Gamma \partial_n \psi_\Gamma|^2 (V \cdot n)\, dS + \frac{\varepsilon_+ - \varepsilon_-}{2} \int_\Gamma |\nabla_\Gamma \psi_\Gamma|^2 (V \cdot n)\, dS \\
&+ \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma \partial_n \hat{\phi}_{\Gamma,\infty} \partial_n \phi_{\Gamma,\infty} ]\!]_\Gamma (V \cdot n)\, dS - \frac{1}{2} \int_\Gamma [\![ \varepsilon_\Gamma \nabla_\Gamma \hat{\phi}_{\Gamma,\infty} \cdot \nabla_\Gamma \phi_{\Gamma,\infty} ]\!]_\Gamma (V \cdot n)\, dS \\
&+ \int_{\Omega_+} \frac{1}{2} \left( \zeta_{\Gamma,V} - \nabla \phi_{\Gamma,\infty} \cdot V \right) B' \left( \psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2} \right) dX
\end{aligned}
$$

$$+\int_\Gamma B\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)(V\cdot n)\,dS. \tag{5.23}$$

*Step 5.* We finally rewrite the volume integral above into a surface integral on the boundary $\Gamma$. Recall from the beginning of Subsection 3.2 that the signed distance function $\phi:\mathbb{R}^3\to\mathbb{R}$ with respect to $\Gamma$ is a $C^3$-function and $\nabla\phi\neq 0$ in the neighborhood $\mathcal{N}_0(\Gamma)$ of $\Gamma$. We extend $n=\nabla\phi$ on $\Gamma$ to $\mathcal{N}_0(\Gamma)$, i.e., we define $n=\nabla\phi$ at every point in $\mathcal{N}_0(\Gamma)$. Note that $n\in C^2(\mathcal{N}_0(\Gamma))$. Since $V\in\mathcal{V}$ vanishes outside $\mathcal{N}_0(\Gamma)$, both the normal component $(V\cdot n)n$ and the tangential component $V-(V\cdot n)n=(I-n\otimes n)V$ of $V$ are in the class of vector fields $\mathcal{V}$; cf. (3.6). Since $V=(V\cdot n)n+(I-n\otimes n)V$ and $(I-n\otimes n)V\cdot n=0$, we have by Lemma 4.2 (part (1) and part (4)) that

$$\begin{aligned}\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V&=\zeta_{\Gamma,(V\cdot n)n+(I-n\otimes n)V}-\nabla\phi_{\Gamma,\infty}\cdot[(V\cdot n)n+(I-n\otimes n)V]\\&=\zeta_{\Gamma,(V\cdot n)n}-\nabla\phi_{\Gamma,\infty}\cdot(V\cdot n)n+\zeta_{\Gamma,(I-n\otimes n)V}-\nabla\phi_{\Gamma,\infty}\cdot(I-n\otimes n)V\\&=\zeta_{\Gamma,(V\cdot n)n}-\nabla\phi_{\Gamma,\infty}\cdot(V\cdot n)n\qquad\text{in }\Omega.\end{aligned}$$

Therefore, we may assume that

$$V=(V\cdot n)n\qquad\text{in }\mathcal{N}_0(\Gamma). \tag{5.24}$$

By Lemma 4.2, $\zeta_{\Gamma,V}|_{\Omega_s}\in H^2(\Omega_s)$ for $s=-$ or $+$. Thus, by (4.16), $\Delta(\nabla\phi_{\Gamma,\infty}\cdot V)\in L^2(\Omega_s)$ for $s=-$ or $+$. Therefore,

$$\nabla\phi_{\Gamma,\infty}\cdot V\in H^2(\Omega_s)\qquad\text{for s }=-\text{ or }+. \tag{5.25}$$

Recall from (5.1) that $\psi_{\rm r}=\psi_\Gamma-\hat\phi_{\Gamma,\infty}\in H^1_0(\Omega)$. Note by Theorem 2.1 that $\Delta\psi_{\rm r}=0$ in $\Omega_-$ and $\varepsilon_+\Delta\psi_{\rm r}=B'(\psi_\Gamma-\phi_{\Gamma,\infty}/2)$ in $\Omega_+$. Note also by (4.16) in Lemma 4.2 that $\Delta(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V)=0$ in $\Omega_-\cup\Omega_+$. We then obtain by Green's second identity with our convention that the normal $n$ at $\Gamma$ pointing from $\Omega_-$ to $\Omega_+$ and the fact that $[\![\varepsilon_\Gamma\zeta_{\Gamma,V}\partial_n\psi_{\rm r}]\!]_\Gamma=0$ which follows from the third equation in (2.24) that twice of the volume term in (5.23) is

$$\begin{aligned}Q&:=\int_{\Omega_+}(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V)\,B'\left(\psi_\Gamma-\frac{\phi_{\Gamma,\infty}}{2}\right)dX\\&=\int_{\Omega_+}\varepsilon_+\left[(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V)\,\Delta\psi_{\rm r}-\psi_{\rm r}\Delta\left(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V\right)\right]dX\\&\quad+\int_{\Omega_-}\varepsilon_-\left[(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V)\,\Delta\psi_{\rm r}-\psi_{\rm r}\Delta\left(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V\right)\right]dX\\&=-\int_\Gamma[\![\varepsilon_\Gamma\left[(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V)\,\partial_n\psi_{\rm r}-\psi_{\rm r}\partial_n\left(\zeta_{\Gamma,V}-\nabla\phi_{\Gamma,\infty}\cdot V\right)\right]]\!]_\Gamma\,dS\\&=\int_\Gamma[\![\varepsilon_\Gamma(\nabla\phi_{\Gamma,\infty}\cdot V)\partial_n\psi_{\rm r}]\!]_\Gamma\,dS+\int_\Gamma[\![\varepsilon_\Gamma\psi_{\rm r}\partial_n\zeta_{\Gamma,V}]\!]_\Gamma\,dS-\int_\Gamma[\![\varepsilon_\Gamma\psi_{\rm r}\partial_n(\nabla\phi_{\Gamma,\infty}\cdot V)]\!]_\Gamma\,dS\\&=Q_1+Q_2-Q_3.\end{aligned} \tag{5.26}$$

It follows from (5.24) that

$$Q_1 = \int_\Gamma [\![\varepsilon_\Gamma (\nabla \phi_{\Gamma,\infty} \cdot V) \partial_n \psi_{\rm r}]\!]_\Gamma \, dS = \int_\Gamma [\![\varepsilon_\Gamma \partial_n \phi_{\Gamma,\infty} \partial_n \psi_{\rm r}]\!]_\Gamma (V \cdot n) dS. \tag{5.27}$$

Since $[\![\psi_{\rm r}]\!]_\Gamma = 0$ and $[\![\varepsilon_\Gamma \partial_n \phi_{\Gamma,\infty}]\!]_\Gamma = 0$, we have by Lemma 4.2 (cf. (4.17)) that

$$\begin{aligned}
Q_2 &= \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \partial_n \zeta_{\Gamma,V}]\!]_\Gamma \, dS \\
&= -\int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} A_V'(0) \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma \, dS \\
&= \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \left[\nabla V + (\nabla V)^T - (\nabla \cdot V) I\right] \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma \, dS \\
&= \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \left[\nabla V + (\nabla V)^T\right] \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma \, dS \\
&= \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \nabla \phi_{\Gamma,\infty} \cdot \left[\nabla V + (\nabla V)^T\right] n]\!]_\Gamma \, dS.
\end{aligned} \tag{5.28}$$

Denoting by $n^j$ the $j$th component of $n$ and noting that $\partial_i n^j n^j = (1/2)\partial_i \|n\|^2 = 0$, we obtain on each side of $\Gamma$ (i.e., on $\mathcal{N}_0(\Gamma) \cap \Omega_-$ and $\mathcal{N}_0(\Gamma) \cap \Omega_+$) that

$$\begin{aligned}
&\nabla \phi_{\Gamma,\infty} \cdot (\nabla V + (\nabla V)^T) n \\
&\quad = \partial_i \phi_{\Gamma,\infty} \left(\partial_j V^i + \partial_i V^j\right) n^j \\
&\quad = \partial_i \phi_{\Gamma,\infty} \partial_j ((V \cdot n) n^i) n^j + \partial_i \phi_{\Gamma,\infty} \partial_i ((V \cdot n) n^j) n^j \qquad \text{[by (5.24)]} \\
&\quad = \partial_i \phi_{\Gamma,\infty} \partial_j (V \cdot n) n^i n^j + \partial_i \phi_{\Gamma,\infty} (V \cdot n) \partial_j n^i n^j \\
&\qquad + \partial_i \phi_{\Gamma,\infty} \partial_i (V \cdot n) n^j n^j + \partial_i \phi_{\Gamma,\infty} (V \cdot n) \partial_i n^j n^j \\
&\quad = (\nabla \phi_{\Gamma,\infty} \cdot n) \nabla (V \cdot n) \cdot n + \nabla \phi_{\Gamma,\infty} \cdot ((\nabla n) n)(V \cdot n) + \nabla \phi_{\Gamma,\infty} \cdot \nabla (V \cdot n).
\end{aligned}$$

This and (5.28), together with the fact that $[\![\varepsilon_\Gamma \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma = 0$ on $\Gamma$, lead to

$$\begin{aligned}
Q_2 &= \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \nabla \phi_{\Gamma,\infty} \cdot (\nabla n) n]\!]_\Gamma (V \cdot n) \, dS + \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \nabla \phi_{\Gamma,\infty} \cdot \nabla (V \cdot n)]\!]_\Gamma \, dS \\
&= Q_{2,1} + Q_{2,2}.
\end{aligned} \tag{5.29}$$

To further simplify these terms, let us recall the surface divergence $\nabla_\Gamma v$ along $\Gamma$ and its integral on $\Gamma$ for a vector field $v$ that belongs to $H^1$ of a neighborhood of $\Gamma$

$$\nabla_\Gamma \cdot v = \nabla \cdot v - (\nabla v) n \cdot n, \tag{5.30}$$

$$\int_\Gamma \nabla_\Gamma \cdot v \, dS = 2 \int_\Gamma H (v \cdot n) \, dS, \tag{5.31}$$

where H is the mean curvature; cf. [20] (Section 5 of Chapter 9).

Consider the term $Q_{2,1}$ in (5.29). Since $n = \nabla\phi$ is a unit vector field, we have $n \cdot (\nabla n)n = n^i \partial_j n^i n^j = (1/2) n^j \partial_j (n^i n^i) = 0$. Hence, on each side of $\Gamma$, we have

$$\nabla\phi_{\Gamma,\infty} \cdot (\nabla n)n = \nabla_\Gamma \phi_{\Gamma,\infty} \cdot (\nabla n)n. \tag{5.32}$$

Let us denote $\alpha_\Gamma = \psi_{\rm r} \nabla_\Gamma \phi_{\Gamma,\infty}$ and note that $[\![\alpha_\Gamma]\!]_\Gamma = 0$. Hence $\alpha_\Gamma \in H^1(\mathcal{N}_0(\Gamma), \mathbb{R}^3)$. Note also that $\alpha_\Gamma \cdot n = 0$. Thus,

$$(\nabla\alpha_\Gamma)n \cdot n + \alpha_\Gamma \cdot (\nabla n)n = \nabla(\alpha_\Gamma \cdot n) \cdot n = 0 \qquad \text{in } \mathcal{N}_0(\Gamma). \tag{5.33}$$

This implies that

$$(\nabla\alpha_\Gamma n) \cdot n = -\alpha_\Gamma \cdot (\nabla n)n \in H^1(\mathcal{N}_0(\Gamma)). \tag{5.34}$$

By (5.24), we have for $s = -$ or $+$ that

$$\begin{aligned} \nabla(\nabla\phi_{\Gamma,\infty} \cdot V) \cdot n &= \nabla((\nabla\phi_{\Gamma,\infty} \cdot n)(V \cdot n)) \cdot n \\ &= (\nabla(\nabla\phi_{\Gamma,\infty} \cdot n) \cdot n)(V \cdot n) + (\nabla\phi_{\Gamma,\infty} \cdot n)\nabla(V \cdot n) \cdot n \qquad \text{in } \Omega_s \cap \mathcal{N}_0(\Gamma). \end{aligned}$$

This, together with (2.4) and (5.25), implies for $s = -$ or $+$ that

$$(\nabla(\nabla\phi_{\Gamma,\infty} \cdot n) \cdot n)(V \cdot n) \in H^1(\Omega_s \cap \mathcal{N}_0(\Gamma)). \tag{5.35}$$

Therefore, since $\nabla_\Gamma \phi_{\Gamma,\infty} = \nabla\phi_{\Gamma,\infty} - (\nabla\phi_{\Gamma,\infty} \cdot n)n$, $\Delta\phi_{\Gamma,\infty} = 0$ in $\Omega_-$ and $\Omega_+$, and $\psi_{\rm r}$ and $\phi_{\Gamma,\infty}$ are in $W^{1,\infty}$ on each side of $\Gamma$, we can verify that for $s = -$ or $+$

$$\begin{aligned} (\nabla \cdot \alpha_\Gamma)(V \cdot n) &= (\nabla\psi_{\rm r} \cdot \nabla\phi_{\Gamma,\infty})(V \cdot n) - (\nabla\psi_{\rm r} \cdot n)(\nabla\phi_{\Gamma,\infty} \cdot n)(V \cdot n) \\ &\quad - \psi_{\rm r}(\nabla(\nabla\phi_{\Gamma,\infty} \cdot n) \cdot n)(V \cdot n) - \psi_{\rm r}(\nabla\phi_{\Gamma,\infty} \cdot n)(\nabla \cdot n)(V \cdot n) \in H^1(\Omega_s \cap \mathcal{N}_0(\Gamma)). \end{aligned} \tag{5.36}$$

By (5.34), (5.36), and (5.30) (with $\alpha_\Gamma$ replacing $v$), we have for $s = -$ or $+$ that

$$(\nabla_\Gamma \cdot \alpha_\Gamma)(V \cdot n) = (\nabla \cdot \alpha_\Gamma)(V \cdot n) - (\nabla\alpha_\Gamma n \cdot n)(V \cdot n) \in H^1(\Omega_s \cap \mathcal{N}_0(\Gamma)). \tag{5.37}$$

With all the regularity results (5.34), (5.36), and (5.37), we have now by (5.32), (5.33), and (5.30) (with $\alpha_\Gamma$ replacing $v$) that

$$\begin{aligned} Q_{2,1} &= \int_\Gamma [\![\varepsilon_\Gamma \alpha_\Gamma \cdot (\nabla n)n]\!]_\Gamma (V \cdot n)\, dS \\ &= -\int_\Gamma [\![\varepsilon_\Gamma (\nabla\alpha_\Gamma)n \cdot n]\!]_\Gamma (V \cdot n)\, dS \\ &= \int_\Gamma [\![\varepsilon_\Gamma (\nabla_\Gamma \cdot \alpha_\Gamma - \nabla \cdot \alpha_\Gamma)]\!]_\Gamma (V \cdot n)\, dS. \end{aligned} \tag{5.38}$$

Consider now the term $Q_{2,2}$ in (5.29). On each side of $\Gamma$,

$$\begin{aligned} \nabla\phi_{\Gamma,\infty} \cdot \nabla(V \cdot n) &= [(\nabla\phi_{\Gamma,\infty} \cdot n)n + \nabla_\Gamma \phi_{\Gamma,\infty}] \cdot [(\nabla(V \cdot n) \cdot n)n + \nabla_\Gamma (V \cdot n)] \\ &= (\nabla\phi_{\Gamma,\infty} \cdot n)(\nabla(V \cdot n) \cdot n) + \nabla_\Gamma \phi_{\Gamma,\infty} \cdot \nabla_\Gamma (V \cdot n). \end{aligned}$$

Since $[\![\psi_{\rm r}]\!]_\Gamma = 0$ and $[\![\varepsilon_\Gamma \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma = 0$, we thus have

$$\begin{aligned}
&[\![\varepsilon_\Gamma \psi_{\rm r} \nabla \phi_{\Gamma,\infty} \cdot \nabla (V \cdot n)]\!]_\Gamma \\
&\quad = [\![\varepsilon_\Gamma \psi_{\rm r} (\nabla \phi_{\Gamma,\infty} \cdot n)(\nabla (V \cdot n) \cdot n)]\!]_\Gamma + [\![\varepsilon_\Gamma \psi_{\rm r} \nabla_\Gamma \phi_{\Gamma,\infty} \cdot \nabla_\Gamma (V \cdot n)]\!]_\Gamma \\
&\quad = [\![\varepsilon_\Gamma \alpha_\Gamma \cdot \nabla_\Gamma (V \cdot n)]\!]_\Gamma. \qquad (5.39)
\end{aligned}$$

One can verify that on both sides of $\Gamma$

$$\nabla_\Gamma \cdot ((V \cdot n)\alpha_\Gamma) = (V \cdot n) \nabla_\Gamma \cdot \alpha_\Gamma + \alpha_\Gamma \cdot \nabla_\Gamma (V \cdot n).$$

Consequently, we have by (5.29), (5.39), (5.31), and the fact that $\nabla_\Gamma \phi_{\Gamma,\infty} \cdot n = 0$ on each side of $\Gamma$ that

$$\begin{aligned}
Q_{2,2} &= \int_\Gamma [\![\varepsilon_\Gamma \alpha_\Gamma \cdot \nabla_\Gamma (V \cdot n)]\!]_\Gamma \, dS \\
&= \int_\Gamma [\![\varepsilon_\Gamma \nabla_\Gamma \cdot ((V \cdot n)\alpha_\Gamma)]\!]_\Gamma \, dS - \int_\Gamma [\![\varepsilon_\Gamma (V \cdot n) \nabla_\Gamma \cdot \alpha_\Gamma]\!]_\Gamma \, dS \\
&= \int_\Gamma [\![2\varepsilon_\Gamma H ((V \cdot n)\alpha_\Gamma \cdot n)]\!]_\Gamma \, dS - \int_\Gamma [\![\varepsilon_\Gamma \nabla_\Gamma \cdot \alpha_\Gamma]\!]_\Gamma (V \cdot n) \, dS \\
&= - \int_\Gamma [\![\varepsilon_\Gamma \nabla_\Gamma \cdot \alpha_\Gamma]\!]_\Gamma (V \cdot n) \, dS.
\end{aligned}$$

This, together with (5.29), (5.38), and the notation $\alpha_\Gamma = \psi_{\rm r} \nabla_\Gamma \phi_{\Gamma,\infty}$, implies that

$$Q_2 = - \int_\Gamma [\![\varepsilon_\Gamma \nabla \cdot \alpha_\Gamma]\!]_\Gamma (V \cdot n) \, dS = - \int_\Gamma [\![\varepsilon_\Gamma \nabla \cdot (\psi_{\rm r} \nabla_\Gamma \phi_{\Gamma,\infty})]\!]_\Gamma (V \cdot n) \, dS. \qquad (5.40)$$

Now, let us calculate the term $Q_3$ in (5.26). Since $V = (V \cdot n)n$ (cf. (5.24)), we have from both sides of $\Gamma$ that

$$\begin{aligned}
\nabla (\nabla \phi_{\Gamma,\infty} \cdot V) \cdot n &= \nabla \left( (\nabla \phi_{\Gamma,\infty} \cdot n)(V \cdot n) \right) \cdot n \\
&= \nabla (\nabla \phi_{\Gamma,\infty} \cdot n) \cdot n (V \cdot n) + (\nabla \phi_{\Gamma,\infty} \cdot n) \nabla (V \cdot n) \cdot n.
\end{aligned}$$

Since $[\![\varepsilon_\Gamma \nabla \phi_{\Gamma,\infty} \cdot n]\!]_\Gamma = 0$, we have by (5.26) and (5.35) that

$$Q_3 = \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \nabla (\nabla \phi_{\Gamma,\infty} \cdot V) \cdot n]\!]_\Gamma \, dS = \int_\Gamma [\![\varepsilon_\Gamma \psi_{\rm r} \nabla (\nabla \phi_{\Gamma,\infty} \cdot n) \cdot n]\!]_\Gamma (V \cdot n) \, dS. \qquad (5.41)$$

It now follows from (5.26), (5.27), (5.40), and (5.41) that

$$Q = \int_\Gamma [\![\varepsilon_\Gamma [\partial_n \phi_{\Gamma,\infty} \partial_n \psi_{\rm r} - \nabla \cdot (\psi_{\rm r} \nabla_\Gamma \phi_{\Gamma,\infty}) - \psi_{\rm r} \nabla (\nabla \phi_{\Gamma,\infty} \cdot n) \cdot n]]\!]_\Gamma (V \cdot n) \, dS. \qquad (5.42)$$

By the definition of the tangential gradient, the fact that $\Delta \phi_{\Gamma,\infty} = 0$ on both sides of $\Gamma$ (cf. (2.6)), and $\nabla \cdot n = 2H$ on $\Gamma$, we can simplify the terms inside the pair of brackets in (5.42). On both sides of $\Gamma$, we have

$$\partial_n \phi_{\Gamma,\infty} \partial_n \psi_{\rm r} - \nabla \cdot (\psi_{\rm r} \nabla_\Gamma \phi_{\Gamma,\infty}) - \psi_{\rm r} \nabla (\nabla \phi_{\Gamma,\infty} \cdot n) \cdot n$$

$$
\begin{aligned}
&= \partial_n\phi_{\Gamma,\infty}\partial_n\psi_{\rm r} - \nabla\cdot[\psi_{\rm r}\nabla\phi_{\Gamma,\infty} - \psi_{\rm r}(\nabla\phi_{\Gamma,\infty}\cdot n)n] - \psi_{\rm r}\nabla(\nabla\phi_{\Gamma,\infty}\cdot n)\cdot n\\
&= \partial_n\phi_{\Gamma,\infty}\partial_n\psi_{\rm r} - \nabla\psi_{\rm r}\cdot\nabla\phi_{\Gamma,\infty} - \psi_{\rm r}\Delta\phi_{\Gamma,\infty}\\
&\quad + \nabla(\psi_{\rm r}(\nabla\phi_{\Gamma,\infty}\cdot n))\cdot n + \psi_{\rm r}(\nabla\phi_{\Gamma,\infty}\cdot n)(\nabla\cdot n) - \psi_{\rm r}\nabla(\nabla\phi_{\Gamma,\infty}\cdot n)\cdot n\\
&= \partial_n\phi_{\Gamma,\infty}\partial_n\psi_{\rm r} - \nabla\psi_{\rm r}\cdot\nabla\phi_{\Gamma,\infty} + (\nabla\phi_{\Gamma,\infty}\cdot n)(\nabla\psi_{\rm r}\cdot n) + \psi_{\rm r}(\nabla\phi_{\Gamma,\infty}\cdot n)(\nabla\cdot n)\\
&= 2\partial_n\phi_{\Gamma,\infty}\partial_n\psi_{\rm r} - [(\nabla\psi_{\rm r}\cdot n)n + \nabla_\Gamma\psi_{\rm r}]\,[(\nabla\phi_{\Gamma,\infty}\cdot n)n + \nabla_\Gamma\phi_{\Gamma,\infty}] + 2H\psi_{\rm r}\partial_n\phi_{\Gamma,\infty}\\
&= \partial_n\phi_{\Gamma,\infty}\partial_n\psi_{\rm r} - \nabla_\Gamma\phi_{\Gamma,\infty}\cdot\nabla_\Gamma\psi_{\rm r} + 2H\psi_{\rm r}\partial_n\phi_{\Gamma,\infty}.
\end{aligned}
$$

Plug this into (5.42). Noting that $\psi_{\rm r} = \psi_\Gamma - \hat{\phi}_{\Gamma,\infty}$ and that all $\nabla_\Gamma\psi_{\rm r}$, $\nabla_\Gamma\phi_{\Gamma,\infty}$, $\varepsilon_\Gamma\partial_n(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})$, and $\varepsilon_\Gamma\partial_n\phi_{\Gamma,\infty}$ are continuous across the boundary $\Gamma$, we obtain that

$$
\begin{aligned}
Q &= \int_\Gamma [\![\varepsilon_\Gamma(\partial_n\psi_{\rm r}\partial_n\phi_{\Gamma,\infty} - \nabla_\Gamma\psi_{\rm r}\cdot\nabla_\Gamma\phi_{\Gamma,\infty})]\!]_\Gamma(V\cdot n)\,dS\\
&= \int_\Gamma [\![\varepsilon_\Gamma[\partial_n(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})\partial_n\phi_{\Gamma,\infty} - \nabla_\Gamma(\psi_\Gamma - \hat{\phi}_{\Gamma,\infty})\cdot\nabla_\Gamma\phi_{\Gamma,\infty}]]\!]_\Gamma(V\cdot n)\,dS. \qquad (5.43)
\end{aligned}
$$

Finally, we obtain by (5.23), (5.26), and (5.43) that some of the terms in $\delta_{\Gamma,V}E[\Gamma]$ (5.23) are simplified into

$$
\begin{aligned}
&\frac{1}{2}\int_\Gamma [\![\varepsilon_\Gamma\partial_n\hat{\phi}_{\Gamma,\infty}\partial_n\phi_{\Gamma,\infty}]\!]_\Gamma(V\cdot n)\,dS - \frac{1}{2}\int_\Gamma [\![\varepsilon_\Gamma\nabla_\Gamma\hat{\phi}_{\Gamma,\infty}\cdot\nabla_\Gamma\phi_{\Gamma,\infty}]\!]_\Gamma(V\cdot n)\,dS\\
&\quad + \int_{\Omega_+}\frac{1}{2}\left(\zeta_{\Gamma,V} - \nabla\phi_{\Gamma,\infty}\cdot V\right)B'\left(\psi_\Gamma - \frac{\phi_{\Gamma,\infty}}{2}\right)dX\\
&= \frac{1}{2}\int_\Gamma [\![\varepsilon_\Gamma\partial_n\hat{\phi}_{\Gamma,\infty}\partial_n\phi_{\Gamma,\infty}]\!]_\Gamma(V\cdot n)\,dS - \frac{1}{2}\int_\Gamma [\![\varepsilon_\Gamma\nabla_\Gamma\hat{\phi}_{\Gamma,\infty}\cdot\nabla_\Gamma\phi_{\Gamma,\infty}]\!]_\Gamma(V\cdot n)\,dS + \frac{1}{2}Q\\
&= \frac{1}{2}\int_\Gamma [\![\varepsilon_\Gamma(\partial_n\psi_\Gamma\partial_n\phi_{\Gamma,\infty} - \nabla_\Gamma\psi_\Gamma\cdot\nabla_\Gamma\phi_{\Gamma,\infty})]\!]_\Gamma(V\cdot n)\,dS\\
&= \frac{1}{2}\int_\Gamma \varepsilon_+\partial_n\psi_\Gamma^+\partial_n\phi_{\Gamma,\infty}^+(V\cdot n)dS - \frac{1}{2}\int_\Gamma \varepsilon_-\partial_n\psi_\Gamma^-\partial_n\phi_{\Gamma,\infty}^-(V\cdot n)dS\\
&\quad - \frac{\varepsilon_+}{2}\int_\Gamma \nabla_\Gamma\psi_\Gamma\cdot\nabla_\Gamma\phi_{\Gamma,\infty}(V\cdot n)\,dS + \frac{\varepsilon_-}{2}\int_\Gamma \nabla_\Gamma\psi_\Gamma\cdot\nabla_\Gamma\phi_{\Gamma,\infty}(V\cdot n)\,dS\\
&= \frac{1}{2}\left(\frac{1}{\varepsilon_+} - \frac{1}{\varepsilon_-}\right)\int_\Gamma \varepsilon_\Gamma\partial_n\psi_\Gamma\varepsilon_\Gamma\partial_n\phi_{\Gamma,\infty}(V\cdot n)dS - \frac{\varepsilon_+ - \varepsilon_-}{2}\int_\Gamma \nabla_\Gamma\psi_\Gamma\cdot\nabla_\Gamma\phi_{\Gamma,\infty}(V\cdot n)\,dS.
\end{aligned}
$$

This and (5.23) imply the desired formula (3.8). The proof is complete. □

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# References


[1] R. Adams. *Sobolev Spaces*. Academic Press, New York, 1975.

[2] D. Andelman. Electrostatic properties of membranes: The Poisson–Boltzmann theory. In R. Lipowsky and E. Sackmann, editors, *Handbook of Biological Physics*, volume 1, pages 603–642. Elsevier, 1995.

[3] N. A. Baker, D. Sept, S. Joseph, M. J. Holst, and J. A. McCammon. Electrostatics of nanosystems: application to microtubules and the ribosome. *Proc. Natl. Acad. Sci. USA*, 98:10037–10041, 2001.

[4] W. M. Boothby. *An Introduction to Differentiable Manifolds and Riemannian Geometry*, volume 120 of *Pure and Applied Mathematics*. Academic Press, 2nd edition, 2002.

[5] D. Bucur and G. Buttazzo. *Variational Methods in Shape Optimization Problems*. Progress in Nonlinear Differential Equations and Their Applications. Birkhäuser, Boston, 2005.

[6] Q. Cai, X. Ye, and R. Luo. Dielectric pressure in continuum electrostatic solvation of biomolecules. *Phys. Chem. Chem. Phys.*, 14:15917–15925, 2012.

[7] Q. Cai, X. Ye, J. Wang, and R. Luo. Dielectric boundary forces in numerical Poisson–Boltzmann methods: Theory and numerical strategies. *Chem. Phys. Lett.*, 514:368–373, 2011.

[8] D. L. Chapman. A contribution to the theory of electrocapillarity. *Phil. Mag.*, 25:475–481, 1913.

[9] J. Che, J. Dzubiella, B. Li, and J. A. McCammon. Electrostatic free energy and its variations in implicit solvent models. *J. Phys. Chem. B*, 112:3058–3069, 2008.

[10] L. Cheng, B. Li, M. White, and S. Zhou. Motion of a cylindrical dielectric boundary. *SIAM J. Applied Math.*, 73:594–616, 2013.

[11] L.-T. Cheng, J. Dzubiella, J. A. McCammon, and B. Li. Application of the level-set method to the implicit solvation of nonpolar molecules. *J. Chem. Phys.*, 127:084503, 2007.

[12] L.-T. Cheng, Z. Wang, P. Setny, J. Dzubiella, B. Li, and J. A. McCammon. Interfaces and hydrophobic interactions in receptor-ligand systems: A level-set variational implicit solvent approach. *J. Chem. Phys.*, 131:144102, 2009.

[13] L.-T. Cheng, Y. Xie, J. Dzubiella, J. A. McCammon, J. Che, and B. Li. Coupling the level-set method with molecular mechanics for variational implicit solvation of nonpolar molecules. *J. Chem. Theory Comput.*, 5:257–266, 2009.

[14] M. Chipot, D. Kinderlehrer, and G. V. Caffarelli. Smoothness of linear laminates. *Arch. Rational Mech. Anal.*, 96:81–96, 1986.

[15] B. Chu. *Molecular Forces, Based on the Lecture of Peter J. W. Debye*. Interscience, John Wiley & Sons, 1967.

[16] C. J. Cramer and D. G. Truhlar. Implicit solvation models: Equilibria, structure, spectra, and dynamics. *Chem. Rev.*, 99:2161–2200, 1999.

[17] S. Dai, B. Li, and J. Lu. Convergence of phase-field free energy and boundary force for molecular solvation. *Arch. Rational Mech. Anal.*, 227(1):105–147, 2018.

[18] M. E. Davis and J. A. McCammon. Electrostatics in biomolecular structure and dynamics. *Chem. Rev.*, 90:509–521, 1990.

[19] P. Debye and E. Hückel. Zur theorie der elektrolyte. *Physik. Zeitschr.*, 24:185–206, 1923.

[20] M. C. Delfour and J.-P. Zolésio. *Shapes and Geometries: Analysis, Differential Calculus, and Optimization*. SIAM, 1987.

[21] J. Dzubiella, J. M. J. Swanson, and J. A. McCammon. Coupling hydrophobicity, dispersion, and electrostatics in continuum solvent models. *Phys. Rev. Lett.*, 96:087802, 2006.

[22] J. Dzubiella, J. M. J. Swanson, and J. A. McCammon. Coupling nonpolar and polar solvation free energies in implicit solvent models. *J. Chem. Phys.*, 124:084905, 2006.

[23] J. Elschner, J. Rehberg, and G. Schmidt. Optimal regularity for elliptic transmission problems including $C^1$ interfaces. *Interfaces and Free Boundaries*, 9:233–252, 2007.

[24] L. C. Evans. *Partial Differential Equations*, volume 19 of *Graduate Studies in Mathematics*. Amer. Math. Soc., 2nd edition, 2010.

[25] F. Fixman. The Poisson–Boltzmann equation and its application to polyelecrolytes. *J. Chem. Phys.*, 70:4995–5005, 1979.

[26] F. Fogolari and J. M. Briggs. On the variational approach to Poisson–Boltzmann free energies. *Chem. Phys. Lett.*, 281:135–139, 1997.

[27] D. Gilbarg and N. S. Trudinger. *Elliptic Partial Differential Equations of Second Order*. Springer-Verlag, 2nd edition, 1998.

[28] M. Gouy. Sur la constitution de la charge électrique a la surface d'un électrolyte. *J. de Phys.*, 9:457–468, 1910.

[29] P. Grochowski and J. Trylska. Continuum molecular electrostatics, salt effects and counterion binding—A review of the Poisson–Boltzmann model and its modifications. *Biopolymers*, 89:93–113, 2008.

[30] J. Huang and J. Zou. Some new a priori estimates for second-order elliptic and parabolic interface problems. *J. Diff. Equations*, 184:570–586, 2002.

[31] J. Huang and J. Zou. Uniform a priori estimates for elliptic and static Maxwell interface problems. *Disc. Cont. Dyn. Syst. B*, 7(1):145–170, 2007.

[32] J. N. Israelachvili. *Intermolecular and Surface Forces*. Academic Press, 3rd edition, 2010.

[33] B. Kawohl, O. Pironneau, L. Tartar, and J.-P. Zolésio. *Optimal Shape Design*, volume 1740 of *Lecture Notes in Mathematics*. Springer, 2000.

[34] S. G. Krantz and H. R. Parks. Distance to $C^k$ hypersurfaces. *J. Diff. Equations*, 40:116–120, 1981.

[35] O. A. Ladyzhenskaya and N. N. Ural'tseva. *Linear and Quasilinear Elliptic Equations*, volume 46 of *Mathematics in Science and Engeneering*. Academic Press, 1968.

[36] J. Lee. *Introduction to Smooth Manifolds*, volume 218 of *Graduate Texts in Mathematics*. Springer, 2nd edition, 2012.

[37] B. Li. Minimization of electrostatic free energy and the Poisson–Boltzmann equation for molecular solvation with implicit solvent. *SIAM J. Math. Anal.*, 40:2536–2566, 2009. See also an erratum in *SIAM. J. Math. Anal.*, 43, 2776–2777, 2011.

[38] B. Li, X. Cheng, and Z. Zhang. Dielectric boundary force in molecular solvation with the Poisson–Boltzmann free energy: A shape derivative approach. *SIAM J. Applied Math.*, 71:2093–2111, 2011.

[39] B. Li, P. Liu, Z. Xu, and S. Zhou. Ionic size effects: Generalized Boltzmann distributions, counterion stratification, and modified Debye length. *Nonlinearity*, 26:2899–2922, 2013.

[40] B. Li and Y. Liu. Diffused solute-solvent interface with Poisson–Boltzmann electrostatics: Free-energy variation and sharp-interface limit. *SIAM J. Applied Math*, 75(5):2072–2092, 2015.

[41] B. Li., H. Sun, and S. Zhou. Stability of a cylindrical solute-solvent interface: Effect of geometry, electrostatics, and hydrodynamics. *SIAM J. Applied Math.*, 75:907–928, 2015.

[42] Y. Y. Li and M. Vogelius. Gradient estimates for solutions to divergence form elliptic equations with discontinuous coefficients. *Arch. Rational Mech. Anal.*, 153:91–151, 2000.

[43] W. Littman, G. Stampacchia, and H. F. Weinberger. Regular points for elliptic equations with discontinuous coefficients. *Ann. Scuola Norm. Sup. Pisa*, 3:43–77, 1963.

[44] X. Liu, Y. Qiao, and B. Z. Lu. Analysis of the mean field free energy functional of electrolyte solution with nonhomogenous boundary conditions and the generalized PB/PNP equations with inhomogeneous dielectric permittivity. *SIAM J. Appl. Math.*, 78(2):1131–1154, 2018.

[45] J. Lu and F. Otto. Nonexistence of a minimizer for Thomas–Fermi–Dirac–von Weizsäcker model. *Commun. Pure Appl. Math.*, 67(10):1605–1617, 2014.

[46] J. A. McCammon. Darwinian biophysics: Electrostatics and evolution in the kinetics of molecular binding. *Proc. Nat. Acad. Sci. USA.*, 106:7683–7684, 2009.

[47] M. Mikucki and Y. C. Zhou. Electrostatic forces on charged surfaces of bilayer lipid membranes. *SIAM J. Applied Math*, 74:1–21, 2014.

[48] E. S. Reiner and C. J. Radke. Variational approach to the electrostatic free energy in charged colloidal suspensions: general theory for open systems. *J. Chem. Soc. Faraday Trans.*, 86:3901–3912, 1990.

[49] K. A. Sharp and B. Honig. Electrostatic interactions in macromolecules: Theory and applications. *Annu. Rev. Biophys. Biophys. Chem.*, 19:301–332, 1990.

[50] J. Sokolowski and J.-P. Zolésio. *Introduction to Shape Optimization: Shape Sensitivity Analysis*. Springer Series in Computational Mathematics. Springer, 1992.

[51] H. Sun, J. Wen, Y. Zhao, B. Li, and J. A. McCammon. A self-consistent phase-field approach to implicit solvation of charged molecules with Poisson–Boltzmann electrostatics. *J. Chem. Phys.*, 143:243110, 2015.

[52] J. Tomasi and M. Persico. Molecular interactions in solution: An overview of methods based on continuous distributions of the solvent. *Chem. Rev.*, 94:2027–2094, 1994.

[53] Z. Wang, J. Che, L.-T. Cheng, J. Dzubiella, B. Li, and J. A. McCammon. Level-set variational implicit solvation with the Coulomb-field approximation. *J. Chem. Theory Comput.*, 8:386–397, 2012.

[54] L. Xiao, Q. Cai, X. Ye, J. Wang, and R. Luo. Electrostatic forces in the Poisson–Boltzmann systems. *J. Chem. Phys.*, 139:094106, 2013.

[55] H. Yin, G. Feng, G. M. Clore, G. Hummer, and J. C. Rasaiah. Water in the polar and nonpolar cavities of the protein interleukin-1$\beta$. *J. Phys. Chem. B*, 114:16290–16297, 2010.

[56] H. Yin, G. Hummer, and J. C. Rasaiah. Metastable water clusters in the nonpolar cavities of the thermostable protein tetrabrachion. *J. Amer. Chem. Soc.*, 129:7369–7377, 2007.

[57] S. Zhou, L.-T. Cheng, J. Dzubiella, B. Li, and J. A. McCammon. Variational implicit solvation with Poisson–Boltzmann theory. *J. Chem. Theory Comput.*, 10:1454–1467, 2014.

[58] S. Zhou, R. G. Weiß, L.-T. Cheng, J. Dzubiella, J. A. McCammon, and B. Li. Variational implicit-solvent predictions of the dry-wet transition pathways for ligand-receptor binding and unbinding kinetics. *Proc. Natl Acad. Sci. USA*, 116(30):14989–14994, 2019.