On the Thermodynamic Description of Electrohydrodynamic Flows

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Abstract— **In the present work, an alternative description for electrohydrodynamic (EHD) flows in dielectric liquids is proposed. The theoretical framework is constructed based on the Helmholtz excess free energy definition for electrically charged, physical or hypothetical, interfaces in the presence of external electric field. Both the interfacial and bulk EHD flows are shown to be generated due to the gradient of the excess surface energies. It is also demonstrated that the Maxwell stress tensor can be derived from the thermodynamic formulation considering the gradient of surface excess energies. Several examples are analyzed using the proposed approach.**

I. Introduction

In almost all cases, mechanical systems with several degrees of freedom admit to a more straightforward description with reduced numbers of parameters, when they are described by the thermodynamic view. The thermodynamic approach does not rely on the internal details of the system, which might be important in the whole analysis. Therefore, this approach is logically and conceptually independent of the mechanics of the system. The thermodynamic description gives an alternative understanding of a system based on the conservation of its total energy, which interacts with the universe through the imaginary boundaries around it. This alternative description has been helpful to understand complicated processes in diverse fields such as biochemistry of cells [1], physics of phase transition [2], astrophysics of stars and stellar systems [3], and numerous heat and mass transport problems [4].

Although the thermodynamic description has been extensively used to describe several interfacial phenomena where charged interfaces are involved, very limited works can be found to describe the EHD flows based on thermodynamic description [5,6]. This might be attributed to the fact that authors have been more interested in details of mechanisms involved in charged carrier transport and its interactions with the fluid flow [5]. Moreover, in previous works, the authors have been more focused on generating bulk EHD flows inside channels or cavities. However, the electrowetting phenomenon is an exception among the other fields of EHD, where finding the thermodynamic description was

the primary goal [7]. To the knowledge of the authors, the thermodynamics of the EHD flows have not been extensively studied. Here, we propose that a thermodynamic description might provide a better intuitive understanding of the EHD flows, especially for interfacial EHD flows.

Recently, it has been shown that the electrostatic forces can be used to induce net interfacial flows. It was found that interfacial components of electrostatic forces, both the normal and tangential ones, can be used to expand liquid films over a conductive substrate. Kim et al. [8] demonstrated a selective spreading of dielectric liquids over a substrate due to the tangential component of electric forces. Mahmoudi et al. [9] reported a radial dielectric film spreading, when it is subjected to a corona discharge. The ionic bombardment of the dielectric interface through a point corona discharge expands the droplet over the grounded substrate uniformly in all radial directions. In the present work, a thermodynamic description of EHD flows is proposed. The surface free energy is defined for both physical and hypothetical interfaces in the volume. A physical interface is defined as a boundary between two different phases, for instance, liquid and vapor. A hypothetical interface is defined as an arbitrary surface within the volume of the liquid. This latter definition was found to be useful for charged dielectric liquids with non-uniform volume charge density.

In this paper, the relationship between the excess energy of the interface and surface charge density is found. It is shown that interfacial EHD flows arise when a gradient of excess energy along the surface is present. The description can be generalized for other bulk EHD flows by integrating the gradient of the excess energy over hypothetical surfaces and deriving the Maxwell stress tensor from the thermodynamic description. The present description for interfacial flows may reveal similarity between the conventional Marangoni effect and the "electrocapillary flows". In a special case where the tangential component of electrostatic forces may induce interfacial net motion, the generated flow can be directly analogous to the Marangoni effect. Since conventional capillary effects, such as the Marangoni effect, have been known for a relatively long time, it is proposed that this analogy might be particularly beneficial for further intuitive understanding of the interfacial EHD flows. The classical approach to the problem using force balance equations with the Maxwell stress tensor is usually straightforward for calculation purposes. However, the description proposed here may provide a unified theory for analyzing various bulk or interfacial EHD flows even though it may not be convenient for quantitative calculations.

II. THERMODYNAMICS OF CHARGED INTERFACES

The Helmholtz free energy of an interface, A^R, at a given reference state, R, can be expressed as [13]

$$dA^R = \left(\frac{\partial A^R}{\partial T}\right)dA + \left(\frac{\partial A^R}{\partial V}\right)dV + \sum_i \left(\frac{\partial A^R}{\partial n_i{}^R}\right)dn_i{}^R \qquad (1)$$

where T and n_i are temperature and the amount of the net electric charges at the given interface, respectively. In this paper, all the derivations are based on the Helmholtz free

energy since the volume of the fluid in the systems was assumed to be constant during the process.

The Helmholtz free energy of the interface at a modified state, A^s, can be defined as

$$dA^s = \left(\frac{\partial A^s}{\partial T}\right)dT + \left(\frac{\partial A^s}{\partial V}\right)dV + \left(\frac{\partial A^s}{\partial \tilde{A}}\right)d\tilde{A} + \sum_i \left(\frac{\partial A^s}{\partial n_i{}^s}\right)dn_i{}^s \quad (2)$$

Where:

$$\left(\frac{\partial A^s}{\partial T}\right) = -S, \quad \left(\frac{\partial A^s}{\partial V}\right) = -P, \quad \left(\frac{\partial A^s}{\partial \tilde{A}}\right) = \gamma, \quad \left(\frac{\partial A^s}{\partial n_i{}^s}\right) = \varphi \quad (3)$$

S, V, $\tilde{A}$, γ, P and φ are entropy, volume, surface area, surface tension, thermodynamic pressure and the surface electric potential, respectively.

The excess energy of the interface, A^{ex}, can be defined as the difference between the reference and modified state. For an isothermal system the excess energy of the interface can be expressed as

$$dA^{ex} = dA^s - dA^R \quad (4)$$

$$dA^{ex} = \left(\frac{\partial A^s}{\partial \tilde{A}}\right)d\tilde{A} + \sum_i \left(\frac{\partial A^s}{\partial n_i{}^s}\right)dn_i{}^{ex} \quad (5)$$

By integrating the both sides, the excess energy of the interface due to the charges can be obtained as:

$$A^{ex} = \left(\frac{\partial A^s}{\partial \tilde{A}}\right)\tilde{A} + \sum_i \left(\frac{\partial A^s}{\partial n_i{}^s}\right)n_i{}^{ex} \quad (6)$$

or:

$$A^{ex} = \gamma\tilde{A} + \sum_i \varphi_i n_i{}^{ex} \quad (7)$$

The total differential of the Helmholtz free energy can be calculated as:

$$dA^{ex} = \gamma d\tilde{A} + \sum_i \varphi_i dn_i{}^{ex} + \tilde{A}d\gamma + \sum_i n_i{}^{ex}d\varphi_i \quad (8)$$

Comparing Eqs (1) and (2), the two last terms must be equal to zero:

$$\tilde{A}d\gamma + \sum_i n_i{}^{ex}d\varphi_i = 0 \quad (9)$$

hence,

$$\tilde{A}d\gamma = -\sum_i n_i{}^{ex}d\varphi_i = 0 \quad (10)$$

or:

$$-d\gamma = \sum_i \sigma_i{}^{ex}d\varphi_i \quad (11)$$

Therefore, the excess surface charge density, $\sigma_i{}^{ex} = n_i{}^{ex}/\tilde{A}$, surface tension, γ, and surface potential, φ_i, can be related by:

$$\frac{d\gamma}{d\varphi} = -\sigma_i{}^{ex} \quad (12)$$

The above equation is formally Lippman's equation [7]. More accurately:

$$\left(\frac{\partial \gamma}{\partial \varphi}\right)_T = -\sigma_i{}^{ex} \qquad (13)$$

The gradient of the surface tension along an arbitrary unit vector, e_χ, in the interfacial plane can be obtained by a chain differentiation at isothermal condition:

$$\frac{d\gamma}{d\chi} e_\chi = \left(\frac{d\gamma}{d\varphi}\right)\frac{d\varphi}{d\chi} \; e_\chi \qquad (14)$$

$$\frac{d\gamma}{d\chi} e_\chi = -\sigma_i{}^{ex} \cdot \frac{d\varphi}{d\chi} \; e_\chi \qquad (15)$$

From Eq. (15), the gradient of the surface energy can be obtained by adding surface charge over the interface. In order to induce surface energy gradient, the surface electric potential should also have variation along the e_χ direction. Therefore, two conditions are required to create electrostatically-induced surface energy gradients along an arbitrary direction: (i) non-zero electric surface charge (ii) non-uniform surface potential over the interface.

In the conventional cases, a gradient of the surface energy can be developed by adding some surfactant to the interface. The local increase in the surfactant concentration over the interface will decrease the surface energy. The induced surface tension gradient leads to a thermodynamically favorable interfacial flow, which minimizes the surface energy all over the interface. This surface energy gradient capillary induced flow is known as Marangoni flow. The resulting Marangoni flow occurs from a region with low surface energy to a region with higher surface energy. Since the surface tension can be varied with temperature, surface temperature gradients may also induce thermal Marangoni flows. Here, we showed that the surface energy gradients can be induced by adding electric surface charges in the presence of non-uniform interfacial electric potential gradients. The resulting interfacial EHD flows can be referred to as "electro-capillary flow" since the generated gradient is due to the excess charge involved in such flows. Here, the non-uniform excess charge over an interface may induce the gradient in the surface energy of a given interface. We shall refer to this as the "electrical Marangoni effect" in analogy with the above description. We will now explain how the EHD interfacial flows can be understood by the concept of this electro-capillary effect through several examples.

III. THERMODYNAMICS OF HYPOTHETICAL CHARGED INTERFACES (GENERALIZED DESCRIPTION)

In order to generalize the above thermodynamic description for EHD flows in dielectric liquids, it is necessary to consider the concept of a hypothetical interface. Figure 1 describes schematically the hypothetical and physical interface for a dielectric liquid film in air. The physical interfaces normally discriminate the boundary of two different phases such as liquid/vapor, whereas the hypothetical surface can be defined arbitrarily anywhere in the bulk. For each hypothetical interface, one may define a reference state and modified state and calculate the excess energy using a similar formulation as used for a physical interface. The gradient of the excess energy creates a force which may contribute to a bulk EHD flow (see Figures 2 and 3). We define the surface energy gradient induced force for a given charged hypothetical interface as:

$$F_{EM} = \left|\widetilde{A_s} \cdot n\right| \nabla\varphi \frac{d\gamma}{d\varphi} \qquad (16)$$

Physical Interface (charged)

Air

Dielectric Liquid

Hypothetical interface inside the charge layer thickness
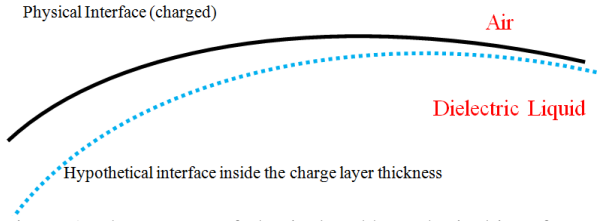
Figure 1. The concept of physical and hypothetical interfaces.

$\nabla\varphi$

Interface

F_{EM}

t

n
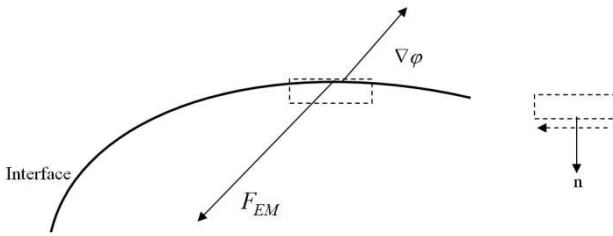
Figure 2. Interfacial electrostatic force and gradient of electric potential

U_1

$\nabla\varphi$

U_2

m"

P_1

Physical or hypothetical interface

P_2

m"

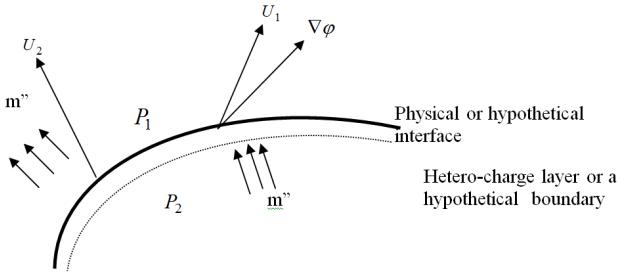Hetero-charge layer or a hypothetical boundary

Figure 3. Momentum balance at dielectric liquid interface. The interface can be hypothetical inside the bulk or physical interface

The direction of the electrical force, F_{EM}, is the same as that of the gradient in electric potential. We assumed the scalar field is the electric potential since we are working with charged interfaces. However, the scalar field can also be defined as the concentration of the surfactant or the temperature field. In the literature, authors considered the Marangoni effect as an interfacial phenomenon and surface gradient was used instead of the spatial gradient in the above equation. This might be attributed to the fact that the surface tension for physical interfaces may only vary along the surface (parallel to the interface). However, since we assumed the interface has a finite thickness, the surface energy in the volume might vary over a hypothetical interface, if the temperature, concentration or electric po-

tential vary spatially. Based on the current definition, the electrical Marangoni effect may have two components, normal and tangential. These two components appear because the gradient of the electric potential may have both normal and tangential components in the bulk. The volume force of the electro-capillary defined as:

$$F_{EMV} = \frac{\left|\tilde{A}_s \cdot n\right| \nabla \varphi \dfrac{d\gamma}{d\varphi}}{\iiint \left(\tilde{A}_s \cdot n\right) d\ell} \tag{17}$$

where ℓ is thickness of the interface.

The hypothetical interface can be defined anywhere inside the bulk. However, it might be useful if it is defined inside the hetero-charge layer where the gradient in volume charge is significant so that the difference between the surface energies of the hypothetical interface is appreciable. Hence, the volume force of the electro-capillary corresponds to

$$F_{EMV} = \frac{\left|\tilde{A}_s \cdot n\right| \nabla \varphi \dfrac{d\gamma}{d\varphi}}{\iiint \left(\tilde{A}_s \cdot n\right) d\ell} = \sigma_v E = (\nabla \cdot D)E \tag{18}$$

σ_v, E and D are volume charge density, electric field and displacement vectors.

In order to show the validity of the above definition, it would be helpful to derive the stress tensor corresponding to the force volume, F_{EMV}. Gauss's divergence theorem provides the stress tensor for the volume electro-capillary force as:

$$F_{EMV,\alpha} = \frac{\partial [F]_{\alpha\beta}}{\partial x_\beta} \tag{20}$$

$F_{EMV,\alpha}$ is the electro-capillary stress tensor. In tensor notation, the components of the force can be calculated as:

$$F_{EMV,\alpha} = (\nabla \cdot D)E = \varepsilon E_\alpha \frac{dE_\beta}{dx_\beta} = \varepsilon \left[\frac{\partial}{\partial x_\beta}(E_\alpha E_\beta) - E_\beta \frac{\partial E_\alpha}{x_\beta} \right] \tag{21}$$

Since the electric field is irrotational, $\nabla \times E = 0$, i.e. $\dfrac{\partial E_\alpha}{x_\beta} = \dfrac{\partial E_\beta}{\partial x_\alpha}$. Therefore,

$$F_{EMV,\alpha} = (\nabla \cdot D)E = \varepsilon E_\alpha \frac{dE_\beta}{dx_\beta} = \varepsilon \left[\frac{\partial}{\partial x_\beta}(E_\alpha E_\beta) - E_\beta \frac{\partial E_\beta}{x_\alpha} \right]$$

(22)

Since $E_\beta \dfrac{\partial E_\beta}{x_\alpha} = \dfrac{1}{2}\dfrac{\partial E_\beta}{\partial x_\alpha} = \dfrac{1}{2}\dfrac{\partial (E_\beta E_\beta)}{\partial x_\alpha} = \dfrac{1}{2}\dfrac{\partial (E^2)}{\partial x_\alpha} = \dfrac{1}{2}\delta_{\alpha\beta}\dfrac{\partial (E^2)}{\partial x_\beta}$, Eq. (22) can be arranged as:

$$F_{EMV,\alpha} = \varepsilon E_\alpha \frac{dE_\beta}{dx_\beta} = \varepsilon \left[\frac{\partial}{\partial x_\beta}(E_\alpha E_\beta) - \frac{1}{2}\delta_{\alpha\beta}\frac{\partial (E^2)}{\partial x_\beta} \right]$$

(23)

Eq.(23) represents the Maxwell stress tensor derived from the thermodynamic description Eq.(17).

As it was noted, the electro-capillary effect may involve both tangential and normal components of force. The tangential component, corresponding to the electrical Marangoni force, can be expressed as:

$$F_{EM} \cdot t = t.[F_{EMv}].n = A_s(t.\nabla\varphi)\frac{d\gamma}{d\varphi} = A_s(\nabla_s \cdot \varphi)\frac{d\gamma}{d\varphi}$$

(24-a)

In the current problem, the induced surface charge may cause gradients in the tangential direction, while in the conventional Marangoni effect the surfactant agents may cause similar surface energy gradients. The excess surface charge over the interface can be viewed as acting in an analogous way to surfactant agents. Therefore, the tangential flows due to the presence of excess charge have been named the "electrical Marangoni" effect. The normal component can be obtained similarly:

$$F_{EM} \cdot n = n.[F_{EMv}].n = A_s(n.\nabla\varphi)\frac{d\gamma}{d\varphi}$$

(24-b)

Conservation of the linear momentum for the interface with a finite thickness can be obtained as:

$$m''(\vec{U_1} - \vec{U_2}) + (P_1[I] - [\tau_1] - P_2[I] + [\tau_2])\vec{n} - \frac{\gamma}{R_{int}}\vec{n} + \left| A_s \cdot \vec{n} \right| \nabla\varphi \frac{d\gamma}{d\varphi} = 0$$

(25)

The normal component of the linear momentum balance equation can be expressed as:

$$m''(u_{1,n} - u_{2,n}) + (P_1 - P_2) + 2\mu_2\left(\frac{\partial u_{2,n}}{\partial n}\right) - 2\mu_1\left(\frac{\partial u_{1,n}}{\partial n}\right) - \frac{\gamma}{R_{int}} + \left| A_s \cdot \vec{n} \right|\left(\frac{d\varphi}{dn}\right)\frac{d\gamma}{d\varphi} = 0$$

(26)

The tangential component can be expressed as:

$$m''(u_{1,t} - u_{2,t}) + \mu_1\left(\frac{\partial u_{1,n}}{\partial t} + \frac{\partial u_{1,s}}{\partial n}\right) - 2\mu_2\left(\frac{\partial u_{2,n}}{\partial t} + \frac{\partial u_{2,s}}{\partial n}\right) + \left| A_s \cdot \vec{n} \right|\left(\frac{d\varphi}{dt}\right)\frac{d\gamma}{d\varphi} = 0$$

(27)

The electro-capillary effect appears in both the normal and tangential component of the momentum balance equations (see equations (26) and (27)). It can be inferred that adding surface charge in the presence of a non-uniform electric potential may contribute to both interfacial (electrical Marangoni at the physical interface) and bulk flows (volume electro-capillary force at hypothetical interfaces). Although the current approach seems to be not convenient for quantitative calculations, it gives an intuitive understanding of the analogy between the conventional interfacial flows and the electrical Marangoni effect. More importantly, it suggests a unified theory for explaining both interfacial and bulk EHD flows based on the thermodynamic description. For further understanding of the proposed description, we analyze several EHD flows based on the above description.

A. Melcher-Taylor Pumping

One of the classical experiments to demonstrate the electrocapillary phenomena at an interface was performed by Taylor and Melcher [10]. A typical Melcher-Taylor pump is depicted in Fig. 4.
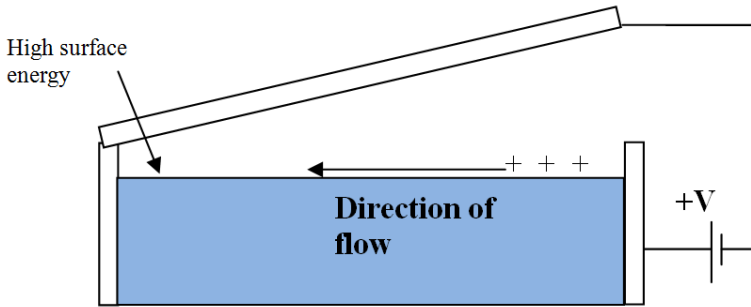

Figure 4. Thermodynamic description of Melcher-Taylor interfacial pumping effect.

Two parallel planar electrodes were fixed vertically on either side of a reservoir. An additional tilted electrode was placed on the top of the reservoir. The tilted electrode on the top and the left electrode were electrically grounded. The reservoir was partially filled with a corn oil. The right side electrode was connected to the positive polarity of the power supply. In this construction both the tangential and normal components of electric field are established over the oil interface. The normal component of the electric field induces positive surface charge at the dielectric interface. The tangential component of the electric field exerts a Coulombic force on the interface that pushes the interface from the right to the left. The resulting capillary flow can be analyzed alternatively based on the thermodynamic description. The surface energy decreases in the direction of the increasing of the surface potential. According to equation (15), the higher electric potential on the right side of the reservoir lowers the surface energy at the right of the container. The high interfacial tension at the left side of the reservoir pulls the interface towards the right side causing a counter clockwise flow. It is worthy to note that since in the absence of an electric field the excess energy of the liquid/vapor interface is zero, a large electric potential is typically necessary to establish the surface charge and a large potential difference is needed to induce such an electro-

capillary flow. This description is consistent with the balance of force description given earlier.

B. Conduction pumping

The mechanical description of the charge transport mechanism and its interaction with the fluid flow is presented in [11]. Conduction pumping can also be described by considering the thermodynamic description. Here, the hypothetical interface might be defined inside the hetero-charge layer to apply the thermodynamic description. The position of the interface can be defined anywhere inside the dielectric medium. Fig. 5 presents a figurative description of the conduction pumping based on this thermodynamic view.
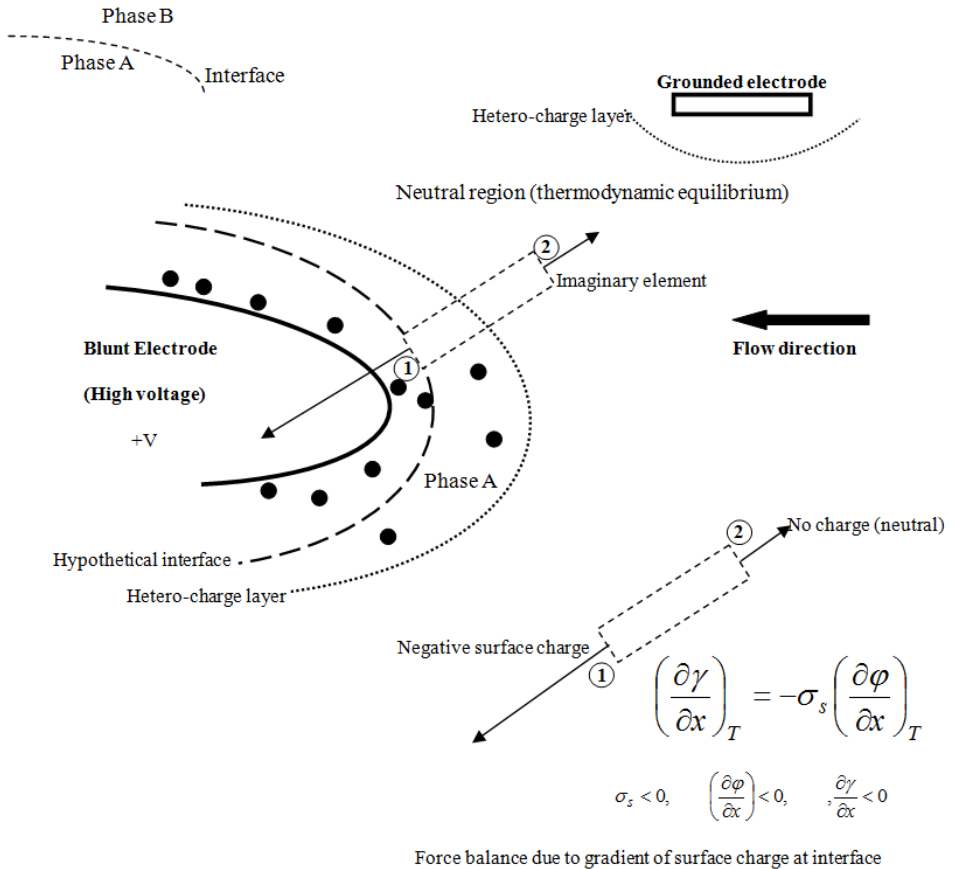

Figure 5. Electrical Marangoni effect in a conduction pump: thermodynamic description. Interface can be defined as the line between the charge layer (phase A) and the neutral region (phase B). Electrical Marangoni effect predicts the direction of pumping.

The electrode edges and the hypothetical interfaces are shown with continuous and discontinuous lines. As depicted in Fig.5, the hypothetical interface is defined inside the heterocharge layer.

Considering the imaginary element, the excess charge in the hetero-charge layer elevates the surface energy (side 1 of the element), while the surface charge is smaller far from the electrode (side 2 of the element). Therefore, a net surface energy gradient is established over the imaginary element towards the electrode. Since the electric potential at side 1 is higher than the side 2 of the element, the element is attracted to the electrode and net dielectric motion is generated towards the high potential electrode due to such surface energy gradient (see Eq. (15)). The surface charge around the high voltage electrode inside the hetero-charge layer is negative and the electric potential gradient is also negative in outwards direction. Based on Eq.(15), the surface energy gradient is also negative. The negative surface energy gradient causes an imbalance surface force and generates a surface energy gradient-driven flow towards the electrode as depicted in Fig. 5. The direction of the conduction pump based on the thermodynamic description is consistent with that of explained previously in [11,12].

C. Dielectric liquid film spreading subjected to corona discharge

It has been demonstrated that the electrostatic forces can be used for spreading of dielectric layers [8,9]. Recently, several studies were performed to show the spreading of the liquid layers using an external electric field. It was found that both interfacial components of the electrostatic forces, normal and tangential, can be used to produce liquid films over a substrate. Kim et al. [8] demonstrated a selective spreading of a dielectric liquid due to the tangential component of electric forces, which was accompanied by formation of a Taylor cone and periodic jetting occurring during the process. Mahmoudi et al. [9] reported that a dielectric droplet exposed to a perpendicular point corona discharge expands uniformly over the grounded substrate due to the normal component of the electric pressure. They also explained that the corona discharge moves the ionic cloud towards the substrate and develops a surface charge over the droplet interface. The normal component of the electric field produces a squeezing electric pressure over the dielectric interface and the droplet expands uniformly over the substrate in the radial direction.

The thermodynamic description can be applied to describe the spreading phenomenon in the presence of the electric field. Consider first the electrode configuration that was used in Kim et al. work [8]. Here a tilted high voltage electrode was positioned above a horizontal flat grounded substrate. The oil film is deposited on the grounded substrate. By applying the electric field between the electrodes, the oil film is driven from the low field to the high field region. The mechanism of spreading is similar to the Melcher-Taylor pump. Considering Eq.15, the interfacial charge over the oil interface is positive, while the gradient of the electric potential is negative. Therefore, the effective surface energy gradient is positive in the spreading direction. This means that the surface energy of the left side is lower than the surface energy of the right side. This gradient of surface energy creates a shear stress at the interface and derives the film from the left side to the right side. The spreading process is thermodynamically preferable since the spreading process drags the liquid into the lower energy region to minimize the energy of the interface as much as possible.

The concept of dielectric spreading the in the presence of corona discharge differs from the Melcher-Taylor pumping. The corona discharge-assisted spreading is mostly based on interfacial pressure and the contribution of the tangential component is not important [9]. However, the thermodynamic description gives a unified picture and describes both spreading mechanisms.

A system consisting a needle and grounded substrate is considered, where the dielectric droplet is gently deposited over the grounded substrate and positive corona voltage is applied to the needle. At the dielectric interface, the cloud of ions generated by corona discharge is deposited over the interface. In this technique, both normal and tangential components of the electric field co-exist on the droplet surface. Since the electric field is divergent, the electric potential gradient in the tangential and normal directions are both negative (see Fig. 6). Considering Eq. (15), the surface energy gradient in radial direction is positive. The charged interface attempts to minimize its energy. Since the effective surface energy at the interface just below the needle tip is small, the gradient of surface energy creates a net shear force in the spreading direction. Similarly, the gradient of surface energy in the normal direction is positive and considering the lower hypothetical interface, the resulting force pulls the physical interface towards the substrate and squeezes the dielectric film. Therefore, the net force generated by the gradients of the surface energy tends to promote the spreading process. In the previous studies of the same authors [9], it was confirmed that the tangential component of the resultant force has negligible contribution for needle-plate configuration. The squeezing force resulting from the normal component of the surface energy gradient is two orders of magnitude larger than the tangential component. Therefore, there is a substantial increase between the rate of expansion of the previous selective spreading technique [8] and the corona-discharge assisted radial spreading [9].
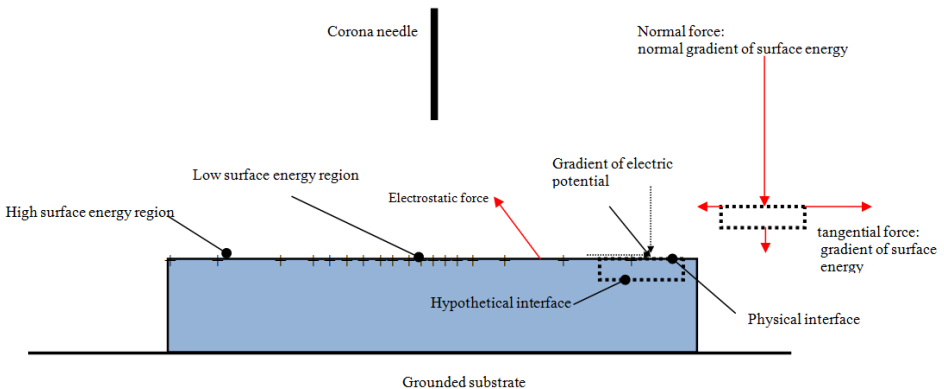

Figure 6. Thermodynamic description of dielectric film spreading when subjected to a corona discharge.

IV. CONCLUSION

In this paper, a unified thermodynamic description for both bulk and interfacial EHD flows is proposed. It was shown that the excess free charge may change the energy of a given interface and create a gradient of the energy. It was demonstrated that the bulk EHD flows can be viewed as electro-capillary flow, if the hypothetical interface is defined inside the volume, while the interfacial EHD is analogous to the Marangoni effect. Based on this thermodynamic description and considering the concept of a hypothetical interface, all the EHD flows can be considered as electro-capillary flow. Three different examples of bulk and interfacial EHD flows are analyzed thermodynamically. The proposed description is particularly important to understand the recently reported interfacial EHD flows in thin films.

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