

# Electrohydrodynamic (EHD) Instabilities

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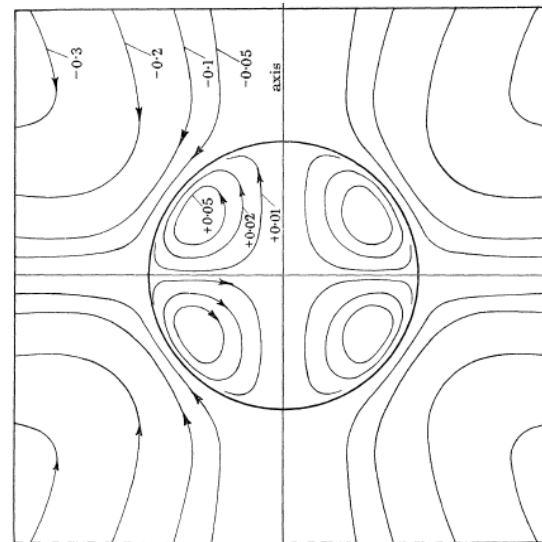
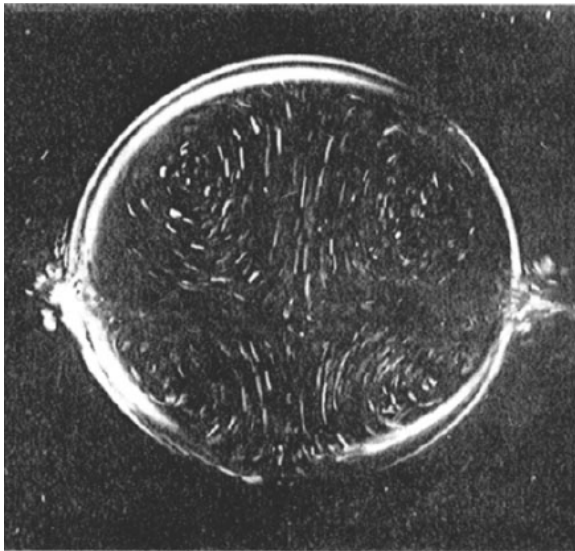
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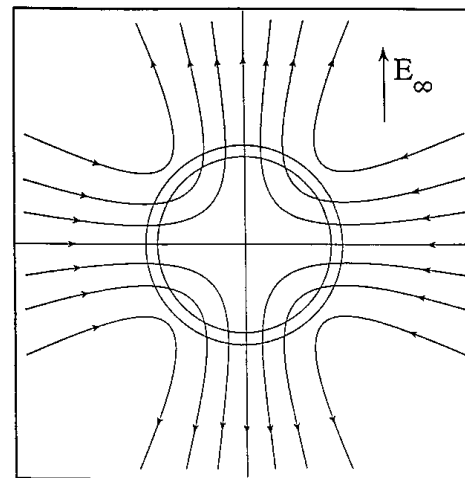
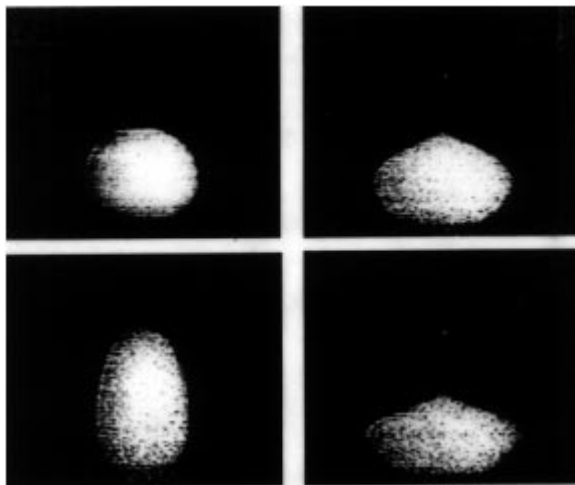
# EHD History: G. I. Taylor (1966)

- “The elongation of a drop of one dielectric fluid in another owing to the imposition of an electric field has **previously been studied assuming that the interface is uncharged and the fluids at rest.**”
- “For a steady field this is unrealistic, because **however small the conductivity of either fluid the charge associated with steady currents must accumulate at the interface** till the steady state is established.”
- “It is shown that **equilibrium can only be established in a drop when circulations are set up** both in the drop and its surroundings.”



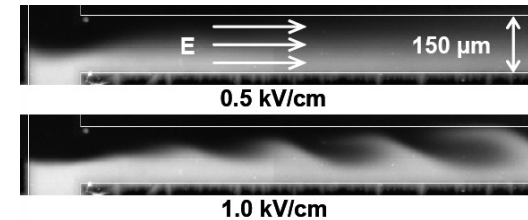
# Developments in Electrohydrodynamics

- Melcher and Taylor 1969, “**Electrohydrodynamics: a review of the role of interfacial shear stresses**,” Annu. Rev. Fluid Mech., 1, 111.
- Melcher 1981, *Continuum Electromechanics*, MIT Press.
- Saville 1997, “**Electrohydrodynamics: the Taylor-Melcher leaky dielectric model**,” Annu. Rev. Fluid Mech. 29, 27.
  - » Recent experimental studies are in better agreement with the theory.
  - » Leaky-dielectric model applies to sharp as well as diffuse interfaces.
  - » Electrohydrodynamics and electrokinetics began to merge since Taylor’s 1966 paper.



# Outline

- **Leaky-dielectric model**
  - » Ohmic model derivations
  - » Maxwell stresses
  - » Jump conditions
  - » Applications in microsystems: the high-conductivity, small-scale limit
- **Electrokinetic flow instabilities**
  - » Bulk-coupled model
  - » Temporal, convective and absolute instabilities
  - » EHD instabilities with electroosmotic convection
  - » Applications in electrokinetic assays and micromixing
- **Electrohydrodynamic cone-jets**
  - » Surface-coupled model
  - » Choking: supercritical flow and pulsating jet
  - » Varicose and whipping instabilities
  - » Applications in droplet microfluidics and electrospinning



# Primary References

- **Electrohydrodynamics**

- Leaky-Dielectric Model***

- » Melcher and Taylor 1969, Annu. Rev. Fluid Mech. 1, 111.
  - » Melcher 1974, IEEE T. Educ. E-17, 100 (and the corresponding film).
  - » Melcher 1981, Continuum Electromechanics, MIT Press.
  - » Saville 1997, Annu. Rev. Fluid. Mech, 29, 27.
  - » Castellanos 1998, Electrohydrodynamics, Springer.

- Flow Instabilities***

- » Saad 1993, Compressible Fluid Flow, 2<sup>nd</sup> Ed. Prentice Hall.
  - » Huerre and Rossi 1998, Ch.2 in Hydrodynamics and Nonlinear Instabilities (ed. Goreche and Manneville), Cambridge
  - » Eggers and Villermaux 2008, Rep. Prog. Phys. 71, 036601.

- **Electrokinetic Flow Instabilities**

- » Melcher and Schwarz 1968, Phys. Fluids, 11, 2604.
  - » Hoburg and Melcher 1976, J. Fluid Mech. 73, 333.
  - » Baygents and Baldessari 1998, Phys. Fluids. 10, 301.
  - » Lin, Storey, Oddy, Chen and Santiago 2004, Phys. Fluids, 16, 1922.
  - » Chen, Lin, Lele and Santiago 2005, J. Fluid Mech. 524, 263.
  - » Posner and Santiago 2006, J. Fluid Mech. 555, 1.

- **Electrohydrodynamic Cone-Jets**

- » Melcher and Warren 1971, J. Fluid Mech. 47, 127.
  - » Cloupeau and Prunet-Foch 1994, J. Aerosol Sci. 25, 1021.
  - » Fernandez de la Mora 1996, J. Colloid Interf. Sci. 178, 209.
  - » Ganan-Calvo 1997, J. Fluid Mech. 97, 165.
  - » Hohman, Shin, Rutledge and Brenner 2001, Phys. Fluids. 13, 2201; 2221.
  - » Chen, Saville and Aksay 2006, Appl. Phys. Lett. 89, 124103.



# Electrohydrodynamic (EHD) Instabilities

## Lectures 1-2: Leaky Dielectric Model

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# Lectures 1-2: Leaky Dielectric Model

- Ohmic model derivation
  - » Electro-diffusion of individual species
  - » Conservation laws of bulk properties
- Maxwell stresses
  - » Kelvin and Helmholtz force densities
  - » Maxwell stress for incompressible flow
- Jump conditions
  - » Surface-coupled model
  - » Bulk-coupled model
- Ohmic model for microsystems
  - » Instantaneous charge relaxation (high conductivity limit)
  - » Diffusive processes (small length scales)



# Electro-Diffusion of Ions\*

Electro-diffusion equations assuming

- Binary, monovalent electrolyte
- Dilute solution
- No reactions (see Saville 1997 for derivation involving reactions)

$$\frac{\partial c_+}{\partial t} + \mathbf{v} \cdot \nabla c_+ = D_+ \nabla^2 c_+ - m_+ F \nabla \cdot (c_+ \mathbf{E}) \quad (1.1)$$

$$\frac{\partial c_-}{\partial t} + \mathbf{v} \cdot \nabla c_- = D_- \nabla^2 c_- + m_- F \nabla \cdot (c_- \mathbf{E}) \quad (1.2)$$

**Advection**

**Diffusion**

**Electro-migration**

\*Levich VG, 1962, *Physicochemical Hydrodynamics*, Prentice-Hall.






# Bulk Properties

**Charge density:**  $\rho_f = F(c_+ - c_-)$  (1.3)

**Conductivity:**  $\sigma = F^2(c_+m_+ + c_-m_-)$  (1.4)

**Einstein's Relation:**  $D_{\pm} = RTm_{\pm}$  (1.5)

**Electro-neutrality if**  $\Theta = Fm_+ \frac{\rho_f}{\sigma} = \frac{c_+ - c_-}{c_+ + \frac{m_-}{m_+} c_-} \ll 1$  (1.6)

$$\boxed{c_+ \approx c_- = c}$$


**Not exactly equal, so there will be a charge density!**

# Algebraic Manipulation

**Eq. (1.1) – Eq. (1.2); applying Einstein's Relation**

$$(D_+ + D_-)F\nabla \cdot (c\mathbf{E}) = RT(D_+ - D_-)\nabla^2 c \quad (1.7)$$

**Substituting (1.7) into (1-1):**

$$\frac{\partial c}{\partial t} + (\mathbf{v} \cdot \nabla)c = D_{eff} \nabla^2 c \quad (1.8)$$

**Effective diffusivity**

$$D_{eff} = \frac{2D_+D_-}{D_+ + D_-} \quad (1.9)$$

**For binary, asymmetric electrolyte  
assuming electro-neutrality**

$$c = \frac{c_+}{z_-} = \frac{c_-}{z_+} \quad (1.10)$$



# “Conservation” of Conductivity

**With electro-neutrality:**

$$\sigma = F^2 (m_+ + m_-) c \quad (1.11)$$

**Eq. (1.8) becomes**

$$\boxed{\frac{\partial \sigma}{\partial t} + (\mathbf{v} \cdot \nabla) \sigma = D_{eff} \nabla^2 \sigma} \quad (1.12)$$

**Interpretation of Eq. (1.12): under the electro-neutrality assumption,**

- Each cation is almost always paired with an anion;
- Conductivity (an weighted sum of cationic and anionic concentrations) can be treated as a material property that is “conserved” in the same manner as uncharged species.



# Conservation of Charge

Eq. (1.1) – Eq. (1.2); **In an exact manner**

On RHS,  $c_+ \approx c_- = c$  applied for simplicity; See Saville 97 for exact derivation.

$$\frac{\partial \rho_f}{\partial t} + \mathbf{v} \cdot \nabla \rho_f = (D_+ - D_-) F \nabla^2 c - (m_+ + m_-) F^2 \nabla \cdot (c \mathbf{E}) \quad (1.13)$$

Rearranging (1.13); Applying incompressibility condition

$$\frac{\partial \rho_f}{\partial t} + \nabla \cdot \left\{ \rho_f \mathbf{v} - \underbrace{(D_+ - D_-) F \nabla c}_{\text{Diffusive Current}} + \sigma \mathbf{E} \right\} = 0 \quad (1.14)$$

**Charge  
Storage**

**Charge  
Convection**

**Diffusive  
Current**

**Ohmic  
Current**



# Charge Conservation in the Ohmic Limit

**If diffusive current is negligible compared to conduction current**

$$\frac{\mathbf{i}_D}{\mathbf{i}_O} \sim O\left(\frac{D_+ - D_-}{D_+ + D_-} \frac{RT / F}{E_a \delta} \frac{\Delta c}{\bar{c}}\right) = \frac{E_d}{E_a} \ll 1 \quad (1.15)$$

$\delta$ : characteristic interfacial (diffusion) length;  $E_a$ : applied E-field

**Eq. (1.14) reduces to**

$$\boxed{\frac{D\rho_f}{Dt} + \nabla \cdot (\sigma \mathbf{E}) = 0} \quad (1.16)$$

**Interpretation of Eq. (1.16):**

- Charge storage in a volume of fixed identity is brought by Ohmic current.
- Negligible diffusive current is a valid assumption when the thermal voltage is small compared to a characteristic external voltage.



# Electro-Diffusion Eqns Recasted as the Ohmic Model

$$\frac{\partial c_+}{\partial t} + \mathbf{v} \cdot \nabla c_+ = D_+ \nabla^2 c_+ - m_+ F \nabla \cdot (c_+ \mathbf{E}) \quad (1.1)$$

$$\frac{\partial c_-}{\partial t} + \mathbf{v} \cdot \nabla c_- = D_- \nabla^2 c_- + m_- F \nabla \cdot (c_- \mathbf{E}) \quad (1.2)$$

**If electro-neutral, i.e.**  $\Theta = Fm_+ \frac{\rho_f}{\sigma} \ll 1$  **(only needed for bulk-coupled model)**

$$\frac{\partial \sigma}{\partial t} + (\mathbf{v} \cdot \nabla) \sigma = D_{eff} \nabla^2 \sigma \quad (1.12)$$

**If negligible diffusive current, i.e.**  $\frac{i_D}{i_o} \sim \frac{E_d}{E_a} \ll 1$  **(for surface- & bulk- coupled models)**

$$\frac{D\rho_f}{Dt} + \nabla \cdot (\sigma \mathbf{E}) = 0 \quad (1.16)$$



# Alternative Derivation of the Ohmic Model

**Conduction current in the laboratory frame,**

$$\mathbf{i} = \mathbf{i}^*(\rho_f, \mathbf{E}) + \rho_f \mathbf{v} = \sigma \mathbf{E} + \rho_f \mathbf{v}$$

**where  $\mathbf{i}^* = \sigma \mathbf{E}$  is postulated as the constitutive law for current**

“It is evident from recently reported research that this simplest of all conduction laws can be used to understand a surprisingly wide range of electrohydrodynamic phenomena.” (Melcher & Taylor 1969)

**Conservation of charge:**

$$\frac{\partial \rho_f}{\partial t} + \nabla \cdot \mathbf{i} = 0 \quad \Longrightarrow \quad \frac{D\rho_f}{Dt} + \nabla \cdot (\sigma \mathbf{E}) = 0$$



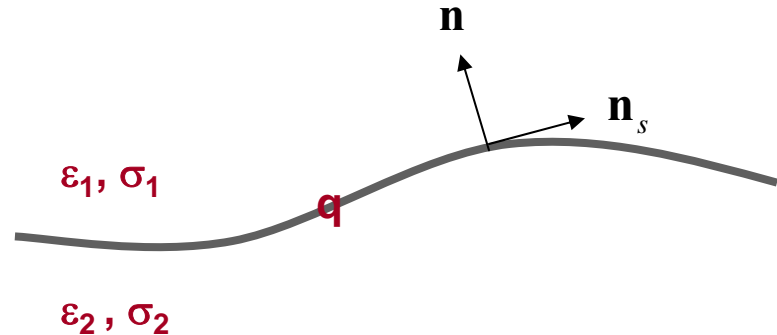
# Electric Jump Conditions

## Differential Laws:

$$\nabla \times \mathbf{E} = 0$$

$$\nabla \cdot (\varepsilon \mathbf{E}) = \rho_f$$

$$\frac{D\rho_f}{Dt} + \nabla \cdot (\sigma \mathbf{E}) = 0$$



## Jump Conditions:

$$\mathbf{n} \times [\![\mathbf{E}]\!] = 0$$

$$\mathbf{n} \cdot [\![\varepsilon \mathbf{E}]\!] = q$$

$$\frac{\partial q}{\partial t} + (\mathbf{n} \cdot \mathbf{v}) \nabla \cdot (q \mathbf{n}) + \nabla_s \cdot (\mathbf{K}_s) + \mathbf{n} \cdot [\![\sigma \mathbf{E}]\!] = 0$$

- Surface charge density:  $q$
- Jump of  $A$  across the interface:  $[\![A]\!] = A_1 - A_2$
- Surface gradient:  $\nabla_s = \nabla - \mathbf{n} \mathbf{n} \cdot \nabla$



# Charge Conservation on the Interface

$$\underbrace{\frac{\partial q}{\partial t}}_{\text{Storage of Surface Charge}} + \underbrace{(\mathbf{n} \cdot \mathbf{v}) \nabla \cdot (q\mathbf{n})}_{\text{Deformation of Interface}} + \underbrace{\nabla_s \cdot (\mathbf{K}_s)}_{\text{Surface Current}} + \underbrace{\mathbf{n} \cdot [\![\sigma \mathbf{E}]\!] }_{\text{Bulk Conduction}} = 0$$

- Interfacial deformation and its consequence on surface charge conservation is discussed in details by Castellanos 1998; Castellanos & Gonzalez 1998.

$$(\mathbf{n} \cdot \mathbf{v}) \nabla \cdot (q\mathbf{n}) = (\mathbf{n} \cdot \mathbf{v})(\mathbf{n} \cdot \nabla q) + q(\mathbf{n} \cdot \mathbf{v})(\nabla \cdot \mathbf{n})$$

- The simplest form of surface current is pure charge advection:  $\mathbf{K}_s = q\mathbf{v}_s$ . Other effects such as surface conduction are typically neglected.
- Bulk conduction is the free current density in the frame of reference moving with the boundary. **Bulk charge advection never reaches the boundary** (Melcher 1981).

# Consequence of the Ohmic Law

$$\frac{D\rho_f}{Dt} = \frac{\rho_f}{\tau_e} - \sigma \mathbf{E} \cdot \left( \frac{\nabla \sigma}{\sigma} - \frac{\nabla \varepsilon}{\varepsilon} \right)$$

- Charge relaxation time  $\tau_e = \varepsilon/\sigma$
- In regions of uniform conductivity and permittivity, for an observer following a particle of fixed identity, the net free charge decays with the relaxation time:

$$\rho_f = \rho_{f,0} \exp(-t / \tau_e)$$

- “Unless an element of material having uniform properties can be traced along a particle line to a source of net charge, it supports no net charge.”



# Charge Density Scale from Ohmic Law

$$\boxed{\frac{D\rho_f}{Dt} = \frac{\rho_f}{\tau_e} - \sigma \mathbf{E} \cdot \left( \frac{\nabla \sigma}{\sigma} - \frac{\nabla \varepsilon}{\varepsilon} \right)}$$

- If  $\rho_f$  results from conductivity gradient

$$\rho_f \sim \varepsilon \mathbf{E} \cdot \frac{\nabla \sigma}{\sigma} \left( \sim \frac{\varepsilon E_a}{\delta} \right)$$

- If  $\rho_f$  results from permittivity gradient

$$\rho_f \sim \mathbf{E} \cdot \nabla \varepsilon$$



# Maxwell Stress in Vacuum

Electric force density (due to free charge) in terms of Maxwell stress:

$$\begin{aligned}\mathbf{f} &= \rho_f \mathbf{E} = \nabla \cdot (\epsilon_0 \mathbf{E}) \mathbf{E} \\ &= \nabla \cdot (\epsilon_0 \mathbf{E} \mathbf{E}) - \epsilon_0 \mathbf{E} \cdot \nabla \mathbf{E} \quad (\nabla \times \mathbf{E} = 0) = \nabla \cdot (\epsilon_0 \mathbf{E} \mathbf{E}) - \frac{1}{2} \nabla \epsilon_0 \mathbf{E} \cdot \mathbf{E} \\ &= \nabla \cdot \left( \epsilon_0 \mathbf{E} \mathbf{E} - \frac{1}{2} \epsilon_0 E^2 \boldsymbol{\delta} \right) = \nabla \cdot \mathbf{T}^e\end{aligned}$$

$$\mathbf{T}^e = \epsilon_0 \begin{bmatrix} \frac{1}{2}(E_x^2 - E_y^2 - E_z^2) & E_x E_y & E_x E_z \\ E_x E_y & \frac{1}{2}(E_y^2 - E_z^2 - E_x^2) & E_y E_z \\ E_x E_z & E_y E_z & \frac{1}{2}(E_z^2 - E_x^2 - E_y^2) \end{bmatrix}$$



# Maxwell Stress in Dielectrics

- **Kelvin force density (assuming the force on each dipole is transmitted to macroscopic medium)**

$$\begin{aligned}\mathbf{f}_K &= \rho_f \mathbf{E} + \mathbf{P} \cdot \nabla \mathbf{E} = \nabla \cdot (\varepsilon \mathbf{E}) \mathbf{E} + (\varepsilon - \varepsilon_0) \mathbf{E} \cdot \nabla \mathbf{E} \\ &= \nabla \cdot \left( \varepsilon \mathbf{E} \mathbf{E} - \frac{1}{2} \varepsilon_0 E^2 \boldsymbol{\delta} \right)\end{aligned}$$

- **Helmholtz force density (accounting for dipole interactions)**

$$\begin{aligned}\mathbf{f}_H &= \mathbf{f}_K - \nabla \left( \frac{1}{2} (\varepsilon - \varepsilon_0) E^2 - \frac{1}{2} \rho \left( \frac{\partial \varepsilon}{\partial \rho} \right)_T E^2 \right) \\ &= \nabla \cdot \left( \varepsilon \mathbf{E} \mathbf{E} - \frac{1}{2} \varepsilon E^2 \boldsymbol{\delta} + \underbrace{\frac{1}{2} \rho \left( \frac{\partial \varepsilon}{\partial \rho} \right)_T E^2 \boldsymbol{\delta}} \right)\end{aligned}$$

**Electrostriction**  
**(typically absorbed in pressure)**



# Equivalence of Kelvin and Helmholtz Concepts

- In incompressible flow, any two force densities differing by the gradient of a scalar pressure will give rise to the same incompressible deformation.
  - » In incompressible flow, pressure becomes a “left-over” variable. It is whatever it must be to satisfy the incompressibility condition:  $\nabla \cdot \mathbf{v} = 0$
- Both force densities, if used consistently, will yield the same answer as far as incompressible mechanical deformation is concerned;
  - » Note the electric force density distributions are very different. See a classic example in Melcher 1981, Section 8.3.
  - » Kelvin force density is useful for appreciating the underlying microscopic electromechanics;
  - » Helmholtz force density is useful for predicting the consequences of electromechanical coupling.



# Maxwell Stress in Incompressible Dielectrics

**Helmholtz force density:**

$$\mathbf{f} = \rho_f \mathbf{E} - \frac{1}{2} E^2 \nabla \varepsilon = \nabla \cdot \left( \varepsilon \mathbf{E} \mathbf{E} - \frac{1}{2} \varepsilon E^2 \boldsymbol{\delta} \right) = \nabla \cdot \mathbf{T}^e$$

$$\mathbf{T}^e = \varepsilon \begin{bmatrix} \frac{1}{2}(E_x^2 - E_y^2 - E_z^2) & E_x E_y & E_x E_z \\ E_x E_y & \frac{1}{2}(E_y^2 - E_z^2 - E_x^2) & E_y E_z \\ E_x E_z & E_y E_z & \frac{1}{2}(E_z^2 - E_x^2 - E_y^2) \end{bmatrix}$$

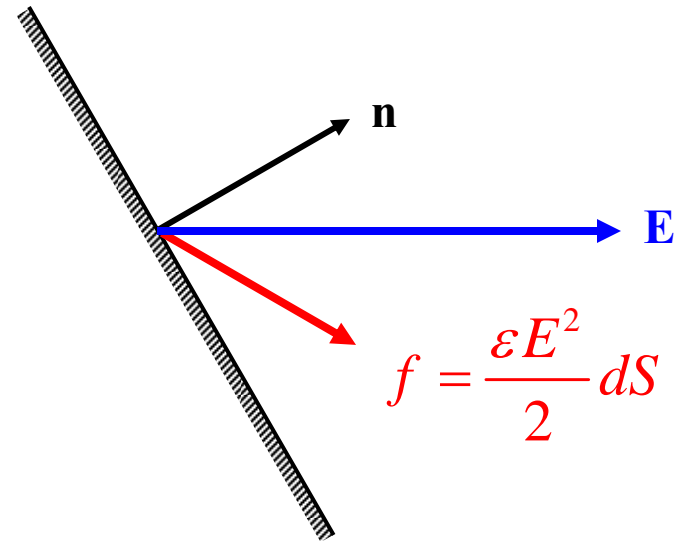
The force density reduces to that in vacuum if  $\varepsilon = \varepsilon_0$ .



# Direction of Maxwell Stress

Choose a coordinate system in which the x-axis is parallel to the direction of the field ( $E_y=E_z=0$ ).

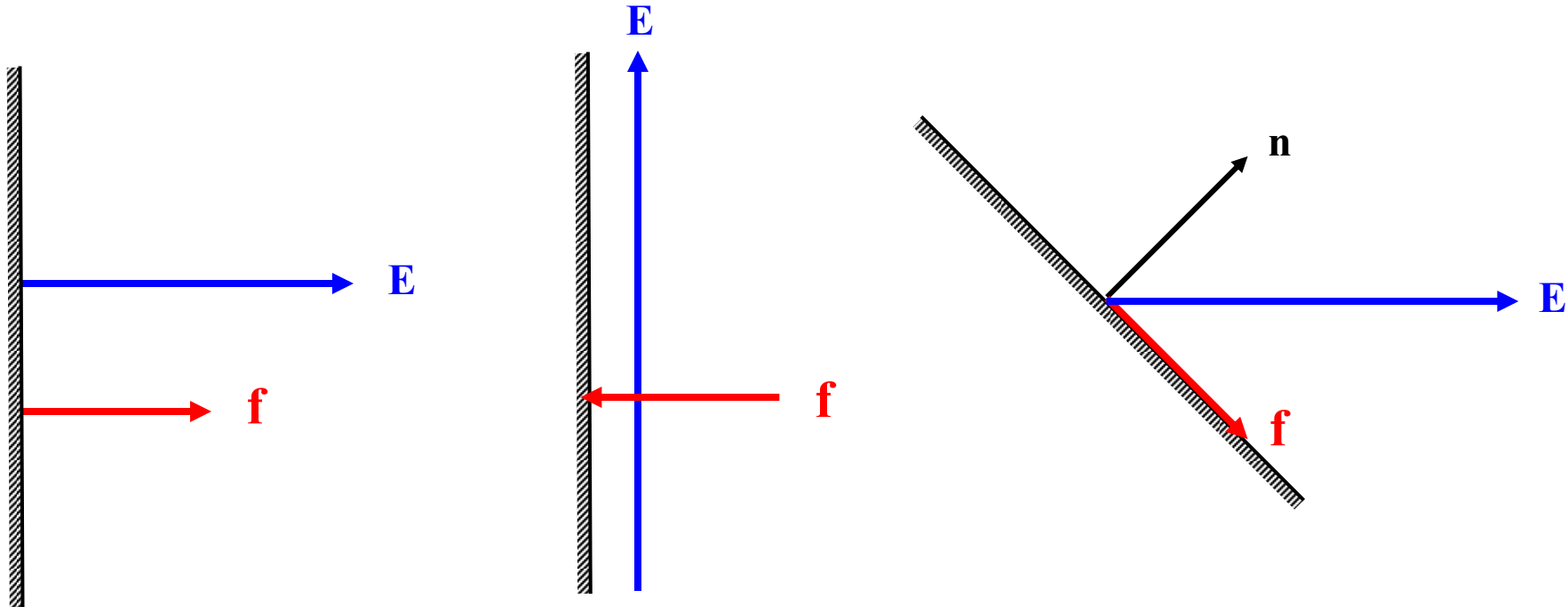
$$\mathbf{T}^e = \epsilon \begin{bmatrix} \frac{1}{2}E^2 & 0 & 0 \\ 0 & -\frac{1}{2}E^2 & 0 \\ 0 & 0 & -\frac{1}{2}E^2 \end{bmatrix}$$



**The electric field bisects the angle between the normal to the surface and the direction of the resultant force acting on the surface.**



# Examples of Maxwell Stresses



# Hydrodynamic Equations

**Conservation of mass (incompressible):**

$$\nabla \cdot \mathbf{v} = 0$$

**Conservation of momentum:**

$$\rho \frac{D\mathbf{v}}{Dt} = -\nabla p^* + \mu \nabla^2 \mathbf{v} + \nabla \cdot \mathbf{T}^e$$

$$\rho \frac{D\mathbf{v}}{Dt} = -\nabla p^* + \mu \nabla^2 \mathbf{v} + \rho_f \mathbf{E} - \frac{1}{2} E^2 \nabla \varepsilon$$

- $p^*$  may have absorbed forces of electric origin;
- $*$  dropped in future eqns

Electric force  
due to  
free charge

Polarization force  
due to  
pairs of charges

# Hydrodynamic Jump Conditions

$$\mathbf{n} \times \llbracket \mathbf{v} \rrbracket = 0$$

$$\mathbf{n} \cdot \llbracket \mathbf{v} \rrbracket = 0$$

$$\mathbf{n} \llbracket p \rrbracket = \mathbf{n} \cdot \llbracket \mathbf{T}^m + \mathbf{T}^e \rrbracket \quad \text{where,}$$

$$\mathbf{T}^m = \mu(\nabla \mathbf{v} + \nabla \mathbf{v}^T)$$

$$\mathbf{T}^e = \varepsilon \mathbf{E} \mathbf{E} - \frac{1}{2} \varepsilon E^2 \boldsymbol{\delta}$$

$$\llbracket \mathbf{T}^e \cdot \mathbf{n} \rrbracket \cdot \mathbf{n} = \frac{1}{2} \llbracket \varepsilon (\mathbf{E} \cdot \mathbf{n})^2 - \varepsilon (\mathbf{E} \cdot \mathbf{t}_1)^2 - \varepsilon (\mathbf{E} \cdot \mathbf{t}_2)^2 \rrbracket$$

$$\llbracket \mathbf{T}^e \cdot \mathbf{n} \rrbracket \cdot \mathbf{t}_i = q \mathbf{E} \cdot \mathbf{t}_i \quad \left( \text{using } \mathbf{n} \times \llbracket \mathbf{E} \rrbracket = 0, \mathbf{n} \cdot \llbracket \varepsilon \mathbf{E} \rrbracket = q \right)$$



# Consequence of Helmholtz Force Density

$$\mathbf{f} = \rho_f \mathbf{E} - \frac{1}{2} E^2 \nabla \varepsilon = \nabla \cdot \left( \varepsilon \mathbf{E} \mathbf{E} - \frac{1}{2} \varepsilon E^2 \boldsymbol{\delta} \right) = \nabla \cdot \mathbf{T}^e$$

- The electric field bisects the angle between the normal to the surface and the direction of the resultant force acting on the surface.
- **Leaky dielectric model is required to generate tangential shear stress at interfaces**
  - » Interface between perfect conductors ( $\sigma \rightarrow \infty$ ): supports no tangential electric stress (not even free charge)
    - **As far as electrostatics is concerned, the two regions can be joined together as a single “perfect” conductor with  $\sigma \rightarrow \infty$ .**
  - » Interface between perfect dielectrics ( $\sigma = 0$ ): force density is perpendicular to the surface (in the direction of  $-\nabla \varepsilon$ )



# Electromechanical Coupling

- **Surface**-coupled model

- » Electromechanical coupling is through **interfacial** electric stresses (i.e. jump conditions)
- » Volumetric force density is zero for piecewise homogeneous media

$$\mathbf{n} \llbracket p \rrbracket = \mathbf{n} \cdot \llbracket \mathbf{T}^m + \mathbf{T}^e \rrbracket$$

- **Bulk**-coupled model

- » Electromechanical coupling is through **volumetric** electric forces
- » Property variations are continuous (i.e. no jump conditions)

$$\mathbf{f} = \rho_f \mathbf{E} - \frac{1}{2} E^2 \nabla \varepsilon$$

# Electrohydrodynamics in Microsystems

- Instantaneous charge relaxation

- » Typically aqueous electrolyte (high-conductivity limit)

$$\cancel{\frac{D\rho_f}{Dt}} + \nabla \cdot (\sigma \mathbf{E}) = 0 \quad \rightarrow \quad \nabla \cdot (\sigma \mathbf{E}) = 0$$

- Diffusive processes become important

- » Small length scales

- » Bulk-coupled model for miscible interfaces

$$\frac{\partial \sigma}{\partial t} + (\mathbf{v} \cdot \nabla) \sigma = 0 \quad \rightarrow \quad \frac{\partial \sigma}{\partial t} + (\mathbf{v} \cdot \nabla) \sigma = D_{eff} \nabla^2 \sigma$$