

**AN ELECTROHYDRODYNAMIC ANALYSIS OF THE EQUILIBRIUM SHAPE AND STABILITY OF STRESSED CONDUCTING FLUIDS : APPLICATION TO LMIS**

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Résumé - Une analyse électrohydrostatique du modèle du cône de Taylor utilisant à la fois les critères de Taylor et de Zeleny fait ressortir plusieurs contradictions. On peut montrer qu'un traitement dynamique de la géométrie d'équilibre et de la stabilité peut résoudre les contradictions apparentes.

Et précisément, en utilisant l'équation électrodynamique linéarisée et des corrections au 1<sup>er</sup> ordre, on montre qu'au seuil d'instabilité le cône prend une forme, "cuspidale". La tension critique de seuil d'instabilité est obtenue pour le gallium à partir de la relation de dispersion. La valeur calculée de 5.8 kV correspond bien aux mesures expérimentales de ~4-7 kV. Enfin, le caractère très localisé de l'instabilité est en accord avec les observations expérimentales obtenues en TEM par Sudraud, et. al.

Abstract - An electrohydrostatic analysis of the Taylor cone model, using both the Taylor and Zeleny stability criteria has revealed several inconsistencies in the model. It is shown that a dynamical treatment of the equilibrium shape and stability can resolve these apparent contradictions in the Taylor model. Specifically, using the linearized electrohydrodynamic equations with corrections up to first-order, it is shown that, at the onset of instability, the cone deforms into a cuspidal shape. From the dispersion relations, the critical voltage for the onset of instability is obtained for liquid gallium. The calculated value of 5.8 kV compares well with experimental values of ~4-7 kV. Finally, the instability is predicted to be highly localized, which agrees with the experimental observations in the TEM images or Sudraud, et. al.

## I. INTRODUCTION

For the past several years, we have been investigating in a systematic and rigorous way some of the basic physics of conducting fluids in strong electric fields[5]. The ultimate objective of this study is to help explain the mechanism for ion (and neutral) emission from a field emission liquid metal ion source (LMIS).

In this study three specific questions are addressed:

1. What is the evolution of the dynamical equilibrium shape of the liquid metal electrode with applied field?
2. When and how does instability occur (i.e., breakdown or disintegration of the liquid electrode, resulting in emission)?
3. What is the specific mechanism for ion formation?

The answer to the first, and also to a large extent the second question, requires a macroscopic theory using fluid mechanics and electrostatics. The physical model(s) and analysis required to answer the last question are microscopic, and use quantum physics. It is obvious that a relationship exists between the quantum mechanism for ion formation and the 'macroscopic' fluid shape. Although this will not be discussed here, some initial results relating to the fluid shape and ion formation will be published elsewhere. In the current paper, we shall be concerned with the macroscopic part of the problem, stated in questions one and two. Specifically, we have used a full electrohydrodynamic (EHD) theory to calculate the dynamical shape of a three-dimensional electrified fluid at the onset of instability. The results are applied to a Ga LMIS and both qualitative and quantitative agreement are found with experimental observations. The dynamical analysis also resolves certain contradictions and inconsistencies in the Taylor cone model.

In Section II, we describe briefly the macroscopic hydrostatic and hydrodynamic concepts of instability. The results of the electrohydrostatic analysis are summarized in Section III. The electrohydrodynamic calculations are outlined in Section IV, and results and conclusions presented in Section V.

## II. MACROSCOPIC DESCRIPTION OF INSTABILITY

An exact theory for the onset of instability, or breakdown of a fluid surface, would treat the problem dynamically, using the complete set of EHD equations. In principle, the set of equations would be solved, for appropriate boundary conditions, from zero field to the critical voltage when instability in the fluid occurs. For convenience, we define two limiting regimes for an instability:

1. The electrohydrostatic limit: This is characterized by low fields and small fluid velocities. In this regime, a quasi-static equilibrium shape is assumed to exist. By quasi-static, we mean the fluid surface deforms in response to the electric field, but the fluid is static. In this limit, the equilibrium surface of the fluid satisfies the Laplace-Young stress balance condition (LY):

$$S \equiv \Sigma(\text{stresses}) = \Delta p + \frac{E^2}{8\pi} - T \left( \frac{1}{R_1} + \frac{1}{R_2} \right) = 0, \quad (1)$$

where the notation is standard, and  $\Delta p = \text{constant} \neq 0$ , in general. Physically, an instability occurs if the outward directed stresses are equal to or greater than the inward stresses. Equation 1 can be solved analytically in some cases[2], and numerically in others[1,2] for quasi-static equilibrium shapes. By applying a criterion for the stability of the solution (see the following discussion), one can solve for  $V_c$ , the critical or threshold voltage for breakdown in the static (or quasi-static) model. This is an important parameter, since it can be compared with experiments[3]. It is essential to know whether the shape determined from the solution of Equation 1 is stable.<sup>†</sup> We have used two criteria to analyze the stability of hydrostatic equilibrium shapes. Given in reverse chronological order in which they first appeared, they are the Taylor[5] and Zeleny[6] stability criteria. We briefly consider each of them:

- (i) Taylor stability criterion: In the Taylor model[5], it is assumed that for equilibrium across the fluid interface, the electrical stress is balanced by the mechanical or surface tension stress, so that Eq. 1 becomes:

$$S = \frac{E^2}{8\pi} - T \left( \frac{1}{R_1} + \frac{1}{R_2} \right) = 0 \text{ with } \Delta p = 0. \quad (2)$$

Although Eq. 2 has been used implicitly by Taylor[5] and others as a stability criterion in the analysis of LMIS[7], it is, in fact, only a limit-

As is well known, "...There must be an exact solution of the equations of fluid dynamics for any problem with given steady external conditions;...yet not every solution of the equation of motion, even if it is exact can actually occur in Nature. The flows that occur in Nature must not only obey the equations of fluid dynamics, but also be stable." [4]

ing case for an equilibrium condition[2]. Furthermore, it is valid only for one special shape of the fluid, with  $\Delta p=0$  and a special form of the counter-electrode, a result recognized by several other groups[8,9].

- (ii) Zeleny stability criterion: Formally, this criterion states that the first derivative of the stresses (S) with respect to a coordinate, say  $\beta$ , characterizing the fluid surface is zero at a point where the instability may arise. This is just the general condition for mechanical stability based on energy considerations. It is equivalent to the mathematical statement that  $\partial^2 U / \partial \beta^2 = 0$ , where U is the free energy of the system, and  $\beta$  is the surface coordinate[10].

Somewhat inexplicably, the Zeleny criterion for stability of fluid interfaces has generally been ignored or overlooked in the analysis of stressed conducting fluids and LMIS.

2. The electrohydrodynamic limit: In this dynamic limit, it is assumed there are pressure gradients which give rise to fluid flow. Hence, the pressure in the LY stress balance condition is treated as a time-dependent quantity. Fluid flow is now included in the description of the deformation of the fluid surface in response to the applied electric field. In the EHD limit, the instability is determined by solving boundary conditions for the velocity and electrostatic potentials, and the fluid surface deformation. Then, from the time-dependent LY stress balance condition, one obtains the dispersion relation

$$\omega^2 = \omega^2(k, T, E, \dots), \quad (3)$$

where  $\omega$  is the angular frequency of the perturbed eigenmode and  $k$  is the wavevector. A hydrodynamic instability occurs when  $\omega^2 < 0$ [4,11]. Physically, the amplitude of the eigenmode  $\omega$  increases without limit leading to an instability, because the fluid cannot follow the oscillation of that mode.

It is important to recognize the different physics, and criteria, necessary to describe the onset of fluid instability in the EHS and EHD cases.

Chung, et. al[2] have given a detailed analysis of an EHS interpretation of the Taylor cone model. We briefly summarize some of their conclusions.

Their results of the analysis of the Taylor model can be stated as follows: A conical interface between two fluids can exist in equilibrium in the presence of an electric field, only if i) the cone has a semi-vertex angle of  $49.3^\circ (\Xi\alpha)$ ; ii)  $\Delta p=0$  across the interface, and iii) the counter-electrode has a shape given by  $r=r_0 [P_{\frac{1}{2}}(\cos \theta)]^{-2}$ . [5,7] Furthermore, using Eq. 2 as a stability criterion, the critical voltage for onset of instability is found to be[5,7]

$$V_C^T = [4 \sin \theta_0 / P_{-\frac{1}{2}}(\cos \theta_0)] \sqrt{2 \pi r_0 T \cot \alpha} \quad (4)$$

where the notation is defined in Refs. 5 or 7.

Three difficulties, or contradictions, are apparent in the Taylor model.

1. The Taylor form of the stress balance condition, Eq. 2, is obtained assuming  $\Delta p=0$  in the general LY condition. What is the validity of assuming  $\Delta p=0$ ? This question has been addressed and discussed in Ref. 7, 8, and 9, and will not be treated further here other than to observe that in our dynamic treatment of the "conical" model, we find that  $\Delta p \neq 0$  after deformation (see Section IV).
2. The cone is treated as a hydrostatic or quasi-static equilibrium shape. However, the Taylor stress condition is regarded as simultaneously a criterion for stability, yielding  $V_C^T$ . If true (i.e., if Eq. 4 is valid), then the Taylor stability criterion predicts an overall - or global - breakdown simultaneously across the entire surface of the cone. This has never been observed experimentally in any study of stressed conducting fluids or LMIS[8,9,12,13].

3. The cone has a singularity at the apex, where the field becomes infinite. This is obviously incompatible with a static equilibrium shape. The significance of singularities on stability and breakdown was first discussed by Chung, et. al.[14] in the analysis of an ideal cuspidal model as a static equilibrium shape.

The results of our calculations suggest that the dynamical treatment of an equilibrium shape and stability can resolve the apparent contradictions and/or inconsistencies in the Taylor model. It is shown that at the onset of instability, the 'static' cone deforms into a cuspidal form, a highly localized instability is predicted and lastly that  $\Delta p \neq 0$ . It is also concluded that  $\Delta p = 0$  is a necessary but not sufficient condition for the onset of a dynamic instability. This is in agreement with Gabovich.[9]

To obtain these results, we have assumed that the most general form of the LY stress condition is

$$\Delta p(t) = \Delta p_0 + \rho \frac{\partial \Omega}{\partial t} = T \left( \frac{1}{R_1} + \frac{1}{R_2} \right) - \frac{E^2}{8\pi} \neq 0, \quad (5)$$

where subscript '0' refers to the static limit, and  $\Omega$  is the velocity potential. Since the fluid velocity  $\vec{u} = -\vec{\nabla}\Omega$ , flow is now introduced into the problem. Before discussing the EHD calculations, we briefly review the EHS analysis of the stability of the Taylor cone. Chung, et. al.[2] have given a complete treatment of the stability of both simple and arbitrary shaped fluid surfaces in the EHS limit.

### III. AN ELECTROHYDROSTATIC ANALYSIS OF THE STABILITY OF SIMPLE COORDINATE SURFACES: THE TAYLOR CONE.

The objective of this analysis is to determine the stability of some known static (or quasi-static) equilibrium shapes. It is necessary to know  $S$ , which requires a knowledge of the curvature and the electrostatic field appropriate for that shape when an external voltage is applied. In general, for arbitrary shapes, the curvature and fields must be obtained numerically[1,8], and the analysis for the onset of instability done by a special procedure of numerical simulation of the fluid surface as a function of applied voltage. This is described in the papers by Chung, et. al. [1,2].

Certain simple coordinate surfaces (or shapes) are especially useful because there are experimental images[5,13] to suggest that each of these shapes has been observed prior to or during the onset of instability. Furthermore, for each of these coordinate surfaces, the cone, the cusp and the hyperboloid of revolution, the curvature and the electrostatic field can be obtained analytically, so that  $S$  can be evaluated. Therefore, the stability analysis can also be done analytically, albeit sometimes approximately.

We here consider only the stability analysis of the cone using the Taylor and Zeleny criteria. The details are given in Ref. 2 for the cone and the other equilibrium shapes.

When the Taylor criterion (i.e., Eq. 2 or  $S=0$  with  $\Delta p=0$ ) is applied, Eq. 4 results. In addition, the following conclusions emerge:

- i) The Taylor cone is an isobaric surface, with, in fact,  $p=0$ .
- ii) Other cones with semi-vertex angle not equal to  $49.3^\circ$  are not isobaric surfaces. That is,  $\Delta p$  varies in value from point to point on the surface.

In the next conclusion, we anticipate results from the complete EHS analysis of stability.

- iii) The Taylor cone is unique in that it is the only shape of a simple coordinate system which is allowed hydrostatically.

One of the contradictions in the Taylor model becomes apparent when  $V_C^T$  (i.e., Eq. 4) is compared with experiment. For liquid Ga,  $T=718$  dynes/cm, so  $V_C^T \approx 12.1$  kV for  $r_0=1$ mm and  $\approx 17.1$  kV for  $r_0=2$ mm. The measured  $V_C^{\text{exp}} \approx 4-7$  kV[13].

When applied to the Taylor cone, the Zeleny stability criterion takes the form  $(\partial S/\partial \theta)_{\theta=\theta_0}=0$  where  $S$  is still given by Eq. 2 applied to the Taylor cone. This can be solved for the critical voltage

$$V_C^Z \approx \frac{4}{P_{-\frac{1}{2}}(\cos \theta_0)} \sqrt{\frac{\pi r_0 T}{(\partial V_0/\partial \theta)_{\theta_0} \ln(r/r_0)}} \quad (6)$$

where  $V_0$  is the non-integral index of the Legendre function and  $r$  is the position coordinate with origin at the apex of the cone. An analysis of Eq. 6 leads to the following conclusions:

- i)  $(\partial S/\partial \theta)_{\theta=\theta_0} < 0$  as  $r \rightarrow 0$ , so the cone is unstable near the apex. At  $r=0$  (i.e., apex), the cone spontaneously disintegrates.
- ii)  $V_C^Z = V_C^Z(r)$ , and therefore predicts local breakdown.

When a similar analysis is done for the cuspidal model as an equilibrium shape, it is found that[2]

- i) The cusp is not an isobaric surface, and therefore cannot be a static equilibrium shape.
- ii) The cusp is spontaneously unstable near and at the apex. The reason is that the high field stresses, due to the singular shape, exceed the mechanical stresses.

Several important conclusions emerge from the complete EHS analysis.

#### A. Taylor cone model:

1. The parameter  $V_C^T$ , obtained from the Taylor stability condition is the voltage necessary to form a static fluid cone with the Taylor angle. It is not the critical or threshold voltage for the onset of instability.
2. In the EHS limit the existence of a singularity at the apex (whether mathematically, or "physically" induced) is essential for the development of an instability there.
3. The quantity,  $V_C^Z$ , calculated from the use of Zeleny stability criterion,  $S'=0$ , is to be interpreted as the additional voltage, over and above  $V_C^T$  for the fluid to disintegrate in the EHS model. Furthermore,  $V_C^Z \rightarrow 0$  as  $r \rightarrow 0$  and no additional voltage beyond  $V_C^T$  is necessary for instabilities to arise at the apex.

#### B. Cuspidal shape:

1. At or near onset of instability where cuspidal shapes are observed experimentally[13], an EHD treatment is necessary to describe accurately the shape and critical voltage for the onset of instability.

C. It is conjectured that none of the simple coordinate surfaces, except the Taylor cone, are allowed static equilibrium shapes. However, according to the Zeleny criterion, the Taylor cone is spontaneously unstable.

D. Finally, arbitrary shaped fluid surfaces with axial symmetry exhibit hydrostatic equilibrium, but only for  $\Delta p \neq 0$ .

#### IV. ELECTROHYDRODYNAMIC ANALYSIS OF EQUILIBRIUM SHAPE AND STABILITY

It is well established that the Taylor cone model has been, and is useful for the study of some qualitative and quantitative features of LMIS. However, it is a static model, and as has been demonstrated, it has inherent inconsistencies (e.g., spontaneous disintegration at the apex). It is apparent that a dynamical theory is necessary for a correct and accurate description of the basic physics involved. Specifically, in order to explain the development and localization of instabilities, as well as the dynamical shapes observed experimentally, an EHD treatment is required. We have therefore used an EHD theory, valid to first-order in the linear approximation, to calculate the dynamical shape of a three-dimensional stressed fluid surface. In this analysis the zero-order (undistorted) surface corresponds to one of the coordinate surfaces of a separable coordinate system (e.g., cone, cusp, etc...). In the calculation reported below, the linearized EHD equations are applied to the exact Taylor cone model because it is a useful (i.e., tractable) zero-order approximation. As will be shown, using this dynamical theory, the cone deforms into approximately the cuspidal shape under the perturbing action of the external electric field.

Since the calculation is very lengthy and tedious, although straightforward, only the formal details are presented here. The mathematical analysis and calculational details will be published elsewhere.

The formal set of EHD equations to be solved are:

1. The Laplace Equations for the electric potential  $\Phi$ , and the velocity potential  $\Omega$ :

$$\nabla^2 \Phi = 0 \quad \text{Outside the conducting fluid} \quad (7a)$$

$$\Delta^2 \Omega = 0 \quad \text{Inside the conducting fluid} \quad (7b)$$

where  $\vec{E} = -\vec{\nabla}\Phi$  and  $\vec{u} = -\vec{\nabla}\Omega$  are the electric field and velocity of the field, respectively.

2. Bernoulli's Equation

$$\rho \frac{\partial \Omega}{\partial t} + \frac{1}{2} \rho (\nabla \Omega)^2 + P + \rho g z = P_0 = \text{constant} \quad (8)$$

This is the equation of motion for an incompressible and irrotational fluid of density  $\rho$ .

3. Boundary conditions on the potentials.

$$\Phi = V_0 \quad \text{on the free fluid surface.} \quad (9a)$$

$$\Phi = 0 \quad \text{on the rigid counter-electrode.} \quad (9b)$$

$$\vec{n} \cdot \vec{\nabla} \Omega = 0, \quad \text{the normal component of the fluid velocity is zero on the} \quad (9c)$$

axis of symmetry.

4. Let  $Z(\alpha, t)$  be a (shape) function which describes the deformation of the fluid surface, and  $\beta = \beta_0$  be the coordinate describing the undistorted fluid surface. We define a function  $F \equiv \beta - Z(\alpha, t) - \beta_0$ , where  $\beta$  defines the deformed surface. Then

$$F = (\beta - \beta_0) - Z(\alpha, t) = 0$$

describes the instantaneous shape of the fluid surface. The function  $F$  must satisfy [15]

$$\frac{dF}{dt} = \frac{\partial F}{\partial t} - \vec{\nabla} \Omega \cdot \vec{\nabla} F = 0. \quad (10)$$

5. The time-dependent LY stress balance condition,

$$\Gamma \left( \frac{1}{R_1} + \frac{1}{R_2} \right) - \frac{E^2}{8\pi} - \Delta P(t) = 0 \quad (11)$$

evaluated on the (deformed) free surface of the fluid,  $\beta = \beta_0 + Z(\alpha, t)$ . The "exact"

set of the coupled EHD eqs. and boundary conditions, given by Eqs. 7-11, are intrinsically nonlinear in the velocity term  $\vec{u}^2$ . Except in very special circumstances, direct solution of these equations is impossible. Linearization of the EHD Eqs. makes some problems tractable, by which we mean that we can solve the equations for approximate solutions that describe the perturbed motion on the fluid surface in terms of simple harmonic oscillations.

Instability in the fluid is a manifestation of developing non-linearity in the surface waves. Therefore, to find the instability, it is necessary to consider non-periodic motion described by the higher-order corrections to the linearized EHD eqs.

The procedure for obtaining and solving the linearized EHD eqs. to first-order is again straightforward, but very tedious. A detailed mathematical treatment is given by Chung[16] and will be published elsewhere. We here merely note that the procedure first entails expanding the deformation and potentials as sums of 'even' and 'odd' contributions. Assuming the deformation  $Z$  to be small, the potentials are expanded in a Taylor series about the undistorted surface, and substituted into the set of the EHD equations and boundary conditions. By equating terms of equal order in the resulting set of equations, a new set of equations is obtained for each of the successively higher-order corrections to the potentials and deformations, in terms of lower-order contributions. Therefore to solve the equations to first-order, we need the zeroth-order solutions for  $\Phi$  and  $\Omega$  on the undeformed surface. When the undeformed surfaces correspond to simple coordinate surfaces (e.g., cone, cusps, etc...), the solutions for  $\Phi_0(\Omega_0)$  are known, and the first-order equations can be solved for the potentials  $\Phi_1(\Omega_1)$  and deformation  $Z_1$ .

It can be shown[16] that the first-order stress condition yields the dispersion relation for  $\omega$ , the frequency (or energy) associated with a perturbed surface wave. From the condition  $\omega^2=0$ , the critical voltage for the onset of an EHD instability is obtained.

## V. RESULTS AND CONCLUSIONS

The above mathematical procedure was first applied to 2-dimensional hydrodynamic fluid geometries consisting of a planar surface with a normal electric field and a cylindrical fluid with a radial field[16], respectively. For liquid Ga,  $V_C \approx 50$  kV and 35 kV for the 2-dimensional geometries.

We used the Taylor cone model in the EHD calculations for three reasons: 1. It is a simple coordinate surface and therefore the zeroth-order solutions for the potentials are known. 2. It is an allowed EHS equilibrium shape, and hence a good zeroth-order model for a dynamical treatment. 3. It is a 'real' 3-dimensional model, and, to our knowledge, the first-order corrections have not been applied to any three-dimensional geometry.

Consider a small deformation  $\zeta(r,t)$  of the liquid electrode about the static Taylor cone. We describe the deformation by  $\Theta=\Theta_0+\zeta(r,t)$ ,  $\Theta_0=130.7^\circ$  for this model, Eqs. 9a and 11 are applied at  $\Theta=\Theta_0+\zeta$ , Eq. 9b at  $r=r_0 [P_{-1/2}(\cos \Theta_0)]^{-2}$  and Eq. 9c at  $\Theta=\pi$ . [17]. The technical details and solutions are discussed elsewhere. Here, we merely note that a rather complicated dispersion relation yields the following expression for the critical voltage:

$$V_C \approx \left[ \frac{2 \sin \Theta_0}{P_{-1/2}(\cos \Theta_0)} \right]^{3/2} \sqrt{4\pi r_0 T (r/a)^s} \quad (12)$$

which can be compared with experiment and the other theoretical expressions,  $V_C^T$  and  $V_C^Z$  (Eqs. 4 and 5).

$V_C$  is rather insensitive to the restricted values of the two parameters  $s$  and  $a$ , where  $r < a < r_0$  and  $0 < s < 0.2 + \eta$ ,  $\eta \ll 1$ . For liquid Ga,  $r = 2$  mm, and  $(r/a) = 0.5$ ,  $V_C = 5.5$  kV. It can be shown[16] that in the limit  $(r/a) \rightarrow 1$ , the critical voltage has the constant value  $V_C = 5.8$  kV, which is in very good agreement with  $V_C^{\text{exp}} \approx 4-7$  kV. For other

liquid metals,  $V_C$  scales as  $\sqrt{T}$ .

Since  $V_C \sim r^S/2$ , the instability is highly localized, which agrees with experimental observations[13].

Finally, the calculated deformation of the cone shows that it deforms into a cuspidal shape. This cuspidal shape agrees well with recent observations of Ben Assayag, et. al.[13], and predictions of Chung, et. al.[14] and of Kingham and Swanson[18].

In summary, the results of the present electrohydrodynamic calculations have demonstrated that an a priori treatment of the equilibrium shape and stability of a stressed conducting fluid is feasible. In a specific application, we have predicted the dynamic shape and critical voltage of an operating LMIS at the onset of instability that is in good agreement with experiment.

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