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The effect of temperature-induced variations in charge carrier mobility on a stationary electrohydrodynamic instability

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An externally applied d.c. electric field is known to produce instability in a plane layer of dielectric liquid heated from above. The stationary linear instability of a unipolar charge injection equilibrium for which carrier mobility depends linearly on temperature is investigated. It is found that the sign of the temperature gradient in relation to the emitting electrode determines whether or not such instability can occur, and that two types of instability can be distinguished: (i) a space charge modified Bénard mode; and (ii) a thermally modified space charge mode. It is shown that the critical voltage is highly dependent upon the absolute value of mobility and its variation across the layer.

1. Introduction

Considerable interest both experimental and theoretical has been shown in recent years in the augmentation of single phase heat transfer in dielectric liquids by employing strong d.c. or a.c. electric fields. For reviews of such investigations see, for example, Bergles (1969), Turnbull (1969), Turnbull & Melcher (1969) and Lazarenko, Grosu & Bologa (1975). Furthermore the possibility of utilizing electrohydrodynamic phenomena in laboratory models of geophysical processes has also been examined (Gross & Porter 1966; Smylie 1966; Gross 1967; Chandra & Smylie 1972).

However, most studies of the effects of d.c. electric fields, and of electrophoresis in particular, assume that the charge distribution in the bulk of the liquid results primarily from thermally induced variations in the small electrical conductivity of the liquid. When the temperature gradient across the liquid is reduced to zero the then constant electrical conductivity does not give rise to a net space charge. The conduction process in such a situation is then described by a 'conductivity' model in which the current is linearly proportional to the electric field when convection and charge diffusion are neglected.

On the other hand fundamental isothermal studies of the processes of electrical conduction in organic liquids, both polar and non-polar, carried out by several investigators (Atten & Gosse 1969; Schneider & Watson 1970; Watson, Schneider & Till 1970; Atten & Moreau 1972; Atten 1975) have concentrated on 'mobility' models of charge transport. This approach is supported by recent electrochemical arguments (Williams, Richardson & Poulter 1978; Williams & Richardson 1978) suggesting that electrical conduction in well-filtered liquid hydrocarbons (e.g. *n*-hexane, transformer oil, kerosene) is governed by the presence of neutral covalent electroactive impurities that undergo oxidation and reduction reactions at the electrodes. The charged impurities

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are then assumed to drift down an electrical potential gradient with a velocity that is linearly proportional to the local electric field strength, the constant of proportionality being called the carrier or ionic mobility.

This mobility approach allows us to consider the practical possibility of controlling the species of impurity, the intensity of injection and the position of the emitting electrode independently of any applied temperature gradient. Furthermore, there is the possibility of a charge-induced fluid dynamic instability even in an isothermal system. A conductivity model, however, does not permit consideration of such details nor does it admit the possibility of an isothermal instability.

Perhaps the simplest configuration exhibiting the destabilizing effect of an electrical field on an otherwise thermally stable layer is the parallel plate system considered by Gross & Porter (1966) and Turnbull (1968*b*). Even though analyses have been attempted by Turnbull (1968*a*), Roberts (1969), Takashima & Aldridge (1976) and Bradley (1978) their results still leave doubt as to the underlying mechanisms responsible for the observed fluid motions. Since we believe that impurities will have a profound effect on the behaviour of such an experimental system, we have chosen to investigate a model that is consistent with much better controlled electrochemical experiments.

2. Equilibrium configuration

Consider an incompressible dielectric liquid of density ρ, electrical permittivity ϵ, constant kinematic viscosity ν and constant thermal diffusivity κ contained between two perfectly conducting rigid horizontal planar electrodes of infinite extent that are distance d apart and maintained at constant though different temperatures and electrical potentials. Suppose also that the liquid contains space charge of density Q with which is associated a charge carrier mobility K. The electrode whose polarity has the same sign as this space charge is then regarded as the emitter, and defines the plane $z = 0$, so that in what follows variables evaluated at the emitter bear the suffix '0', and the collecting electrode corresponds to the plane $z = d$.

A dimensional analysis of the problem suggests that current density, applied voltage and thermal effects (i.e. buoyancy forces, variations in dielectric constant and carrier mobility) as well as the thermomechanical and electrical properties of the liquid be characterized by a set of non-dimensional parameters. It is convenient to choose therefore

$$\left.\begin{gathered} C = Q_0 d^2/\epsilon_0 \phi_0, \quad T_s = \epsilon_0 \phi_0/\rho_0 \nu K_0, \\ R = \alpha\beta g d^4/\nu\kappa, \quad \epsilon_1 = e_1 \beta d, \quad \kappa_1 = k_1 \beta d, \\ P = \nu/\kappa, \quad A = P\rho_0 K_0^2/\epsilon_0 \end{gathered}\right\} \tag{2.1}$$

respectively,† where ϕ_0, β, α, g, e_1 and k_1 are the applied potential difference, average temperature gradient, liquid thermal expansion coefficient, the acceleration due to gravity and the temperature coefficients of electrical permittivity and of charge carrier mobility. R is the Rayleigh number, P the Prandtl number, A the 'mobility parameter' for the liquid and C a measure of the magnitude of injected charge.

Once values have been assigned to these parameters the fluid velocity, temperature and electrical potential fields are determined by the governing electrohydrodynamic

† The quantity ϵ_0 is not to be confused with the permittivity of free space.

equations in which magnetic effects have been neglected. We then have the following reduced field equations:

$$\nabla.\mathbf{u} = 0, \quad \rho(\partial/\partial t + \mathbf{u}.\nabla)\mathbf{u} = -\nabla p + \rho g \mathbf{i}_z + \rho\nu\nabla^2\mathbf{u} + \mathbf{F}, \tag{2.2), (2.3}$$

$$(\partial/\partial t + \mathbf{u}.\nabla)T = \kappa\nabla^2 T, \quad \mathbf{E} = -\nabla\phi, \tag{2.4), (2.5}$$

$$\nabla.\mathbf{D} = Q, \quad \nabla.\mathbf{j} = -\partial Q/\partial t; \tag{2.6), (2.7}$$

where $\mathbf{u}$ denotes fluid velocity, p kinetic pressure, $\mathbf{F}$ the electrical body force per unit volume, T temperature, $\mathbf{E}$ electric field, ϕ electrical potential, $\mathbf{D}$ electric displacement vector and $\mathbf{j}$ current density. Viscous and electrical dissipation have been neglected in the energy equation (2.4). In addition we require constitutive equations in order to specify the particular model under consideration. Having already assumed the liquid to be Newtonian [cf. (2.3)] we further postulate that

$$\mathbf{D} = \epsilon\mathbf{E}, \quad \mathbf{j} = Q(K\mathbf{E} + \mathbf{u}), \tag{2.8), (2.9}$$

$$\mathbf{F} = Q\mathbf{E} - \tfrac{1}{2}E^2\nabla\epsilon + \tfrac{1}{2}\nabla\{\rho E^2(\partial\epsilon/\partial\rho)_T\}, \tag{2.10}$$

so that the liquid is a linear isotropic dielectric containing a single charged species and charge diffusion phenomena are negligible. Finally the equations of state for density, permittivity and mobility are assumed to take the form

$$\rho = \rho_0[1 - \alpha(T - T_0)], \tag{2.11}$$

$$\epsilon = \epsilon_0[1 + e_1(T - T_0)], \tag{2.12}$$

$$K = K_0[1 + k_1(T - T_0)], \tag{2.13}$$

where T_0 is a reference temperature defined as that of the emitter. The temperature dependence of mobility as measured isothermally is usually expressed by the Arrhenius-type equation

$$K(T) = K_0 \exp\left[-W_k\left(\frac{1}{T} - \frac{1}{T_0}\right)\right], \tag{2.14}$$

where W_k is the ratio of an activation energy to the Boltzmann constant (cf. Gallagher 1975, chapter 1), so that (2.13) is equivalent to a Taylor expansion of (2.14) to first order in $T - T_0$. A comparison of these equations with experimental and theoretical data for non-polar organic liquids near room temperature suggests that $k_1 \sim 10^{-2}$ (Gray & Lewis 1969; Adamczewski & Calderwood 1975) whereas $e_1 \sim -10^{-3}$ (Kaye & Laby 1973).

In general kinematic viscosity ν and thermal diffusivity κ will vary with temperature thus requiring two further equations of state. However, in this analysis we neglect such variations in order to highlight the effects of buoyancy and the temperature dependence of mobility.

The above system of equations (2.4)–(2.13) possesses a steady one-dimensional hydrostatic ($\mathbf{u} = 0$) equilibrium $\mathbf{j} = [0, 0, j(z)]$, $\mathbf{E} = [0, 0, E(z)]$ given by

$$T = T_0 + \beta z, \quad \epsilon = \epsilon_0(1 + e_1\beta z), \tag{2.15), (2.16}$$

$$K = K_0(1 + k_1\beta z), \quad j = j_0 = K_0 Q_0 E_0 = K(z)\,Q(z)\,E(z), \tag{2.17), (2.18}$$

$$E = \frac{\epsilon_0 E_0}{\epsilon}\left[1 + \frac{2Q_0}{\epsilon_0 E_0}H(z)\right]^{\frac{1}{2}}, \tag{2.19}$$

$$Q = K_0 Q_0 \epsilon \Big/ \left\{\epsilon_0 K\left[1 + \frac{2Q_0}{\epsilon_0 E_0}H(z)\right]^{\frac{1}{2}}\right\}, \tag{2.20}$$

and
$$\phi = \int_z^d E(z')\,dz', \tag{2.21}$$

where $H(z)$ is defined by

$$H(z) = \frac{K_0}{\epsilon_0}\int_0^z \frac{\epsilon(z')}{K(z')}dz' \tag{2.22a}$$

$$\simeq \frac{d}{\kappa_1}\ln\left(1+\frac{\kappa_1 z}{d}\right) \quad \text{for} \quad |e_1| \ll |k_1|, \tag{2.22b}$$

satisfying boundary conditions on electrical potential and temperature of the form

$$\phi(0) = \phi_0, \quad \phi(d) = 0, \quad T(0) = T_0, \quad T(d) = T_0 + \beta d, \tag{2.23}$$

provided that

$$\epsilon_0 E_0 \int_0^d \left\{\left[1+\frac{2Q_0}{\epsilon_0 E_0}H(z')\right]^{\frac{1}{2}} \Big/ \epsilon(z')\right\} dz' = \phi_0. \tag{2.24}$$

However for a complete specification we require a further condition relating Q_0 and E_0. This takes the form of a postulated injection law $Q_0 = f(E_0)$ describing the manner in which charge enters the liquid at the emitter. In the absence of a well-established electrochemical theory of charge injection at a solid/liquid interface we choose for simplicity an 'autonomous' injection following Atten & Moreau (1972) who argue that it is the least stabilizing of injection laws. In practice, of course, charge is removed from the liquid at the collector, a process which should in principle be described by an ejection law. That no such law is required in the present analysis results from neglecting charge diffusion. The equilibrium defined by (2.15)–(2.24) is then a mainstream solution of a boundary-layer problem, the matching exponential charge diffusion boundary layer being located at the collector (cf. Richardson & Poulter 1976). The limiting case of strong injection, known as that of space-charge-limited currents (SCLC) is defined by the limit $Q_0 \to \infty$ [or $C \to \infty$; cf. (2.1)] implying that $E_0 = 0$ [cf. (2.18)]. This means that the system is passing the largest current possible for given ϕ_0 and β. Again in practice charge diffusion will prevent Q_0 from becoming infinite. However, further boundary-layer analysis (cf. Richardson 1978) suggests that the equilibrium (2.15)–(2.24) must in this case be matched to an algebraic charge diffusion boundary layer at the emitter by using the condition $E_0 = 0$ on the mainstream solution. Hence for the case of infinite C we replace the autonomous injection law by $E_0 = 0$.

It is clear that there are four distinct cases of this equilibrium depending upon the position of the emitting electrode and its temperature relative to that of the collector (see figure 1). The possibility of these equilibria being dynamically unstable is suggested by an energy argument (cf. Gross 1969) implicitly neglecting the effects of buoyancy, viscosity, thermal diffusion and space charge modification of the electric field. A necessary condition for stationary instability is that

$$\nabla\phi.\nabla Q > 0, \tag{2.25}$$

somewhere within the liquid. This is equivalent, on neglecting the temperature-induced variations of ϵ, to

$$\kappa_1 + C\frac{\phi_0}{E_0 d}[1 + 2\ln(1+\kappa_1 z/d)] > 0. \tag{2.26}$$

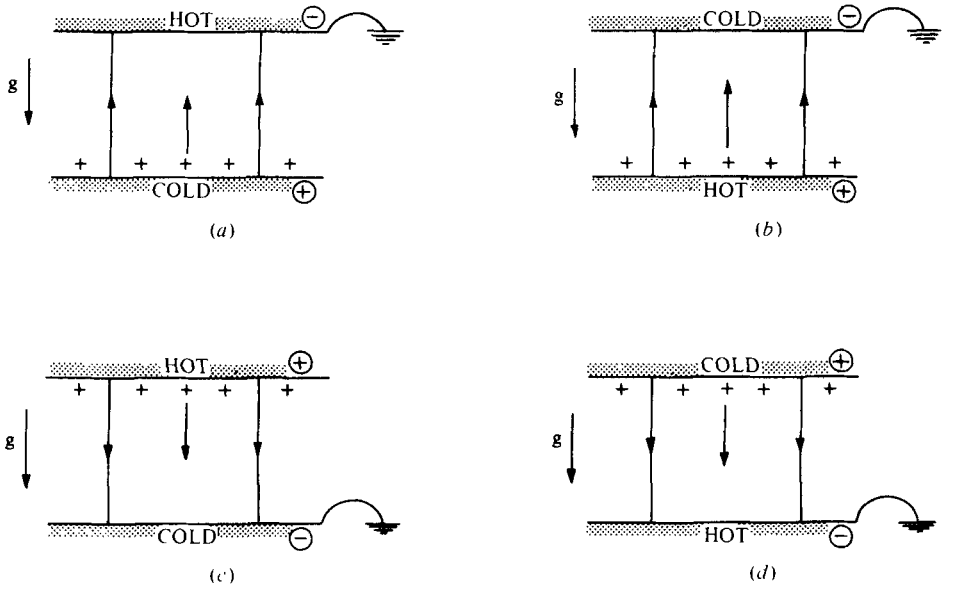

FIGURE 1. Equilibrium configurations: (*a*) cold emitter $\beta > 0$, heating from above $R < 0$; (*b*) hot emitter $\beta < 0$, heating from below $R > 0$; (*c*) hot emitter $\beta < 0$, heating from above $R < 0$; (*d*) cold emitter $\beta > 0$, heating from below $R > 0$.

Since C and E_0/ϕ_0 are intrinsically positive, this implies that (*a*) if $\kappa_1 > 0$ (i.e. the cold emitter case) the system is potentially unstable, and (*b*) if $\kappa_1 < 0$ (the hot emitter case) the system is not susceptible to stationary instability when

$$C(\phi_0/E_0 d) + \kappa_1 < 0. \tag{2.27}$$

We thus expect a system with a cold emitter to be generally more unstable than one with a hot emitter in otherwise similar circumstances; thus, for instance, configuration (*a*) in figure 1 should be more unstable than (*c*) for the same values of C and $|\beta|$.

3. Linear stability analysis

3.1. *Perturbation equations and boundary conditions*

Let $\mathbf{u} = (u_x, u_y, u_z)$, δT and $\delta\phi$ be perturbations, whose squares and products are negligible, in the velocity, temperature and electrical potential respectively of the one-dimensional equilibrium state of § 2. If these perturbations are analysed into normal modes so that

$$u_z = V(z)\exp[i(k_x x + k_y y) + st], \tag{3.1}$$

$$\delta T = \theta(z)\exp[i(k_x x + k_y y) + st], \tag{3.2}$$

$$\delta\phi = F(z)\exp[i(k_x x + k_y y) + st], \tag{3.3}$$

where $\mathbf{k} = [k_x, k_y]$ is a two-dimensional vector wavenumber of magnitude k and s is the growth rate, which can be complex, and we introduce non-dimensional variables

$$x^* = x/d, \quad y^* = y/d, \quad z^* = z/d, \quad t^* = \nu t/d^2,$$

$$u_z^* = du_z/\kappa, \quad \delta T^* = \delta T/\beta d, \quad E^* = Ed/\phi_0,$$

$$\delta\phi^* = \delta\phi/\phi_0, \quad Q^* = Q/Q_0, \quad \epsilon^* = \epsilon/\epsilon_0, \quad K^* = K/K_0, \tag{3.4}$$

the resulting equations governing linear perturbations reduce to

$$(D^2-k^2-Ps)\,\theta = V, \tag{3.5}$$

$$(D^2-k^2)(D^2-k^2-s)\,V = k^2[-R\theta+\epsilon_1 AT_s^2 ED(E\theta)]+k^2\epsilon_1 AT_s^2(EDE)\,\theta + k^2AT_s^2C(DQ)\,F-k^2AT_s^2\epsilon E(D^2-k^2)\,F, \tag{3.6}$$

$$\begin{aligned} C(DQ)\,V = {} & Ps\epsilon(D^2-k^2)\,F+AT_sCKQ(D^2-k^2)\,F \\ & +AT_sD[\epsilon EK(D^2-k^2)\,F]+AT_sC[D(KQ)]\,DF \\ & +\epsilon_1 PsDF+\epsilon_1 AT_sD[KEDF]-\epsilon_1 AT_sD[KED(E\theta)] \\ & -\kappa_1 AT_sCD[QE\theta]-\epsilon_1 PsD(E\theta). \end{aligned} \tag{3.7}$$

Here D denotes differentiation with respect to z, asterisks have been omitted, and we have made the Boussinesq approximation for density variations. The Rayleigh number R is positive (negative) for heating from below (above) provided that the sign of g is taken to be positive (negative) when the upper (lower) electrode is the emitter.

The three ordinary differential equations (3.5)–(3.7), whose coefficients are functions of z, constitute a ninth-order eigenvalue problem upon specification of nine boundary conditions. At the rigid perfectly conducting electrode/liquid interfaces clearly the no-slip conditions and fixed temperature and potential criteria must hold, so that

$$V = DV = \theta = F = 0 \quad \text{at} \quad z = 0, 1. \tag{3.8}$$

The ninth condition is obtained from an injection law, and reduces to

$$D^2F - C_1 DF = 0 \quad \text{at} \quad z = 0, \tag{3.9}$$

where $C_1 = (d/\epsilon_0)(df/dE_0)$. In the case of autonomous injection we choose $C_1 = 0$ giving $D^2F = 0$ whereas in the case of space-charge-limited currents we let $C_1 \to \infty$ giving $DF = 0$ [cf. Atten & Moreau 1972].

In the isothermal limit $\epsilon_1 = \kappa_1 = R = 0$ (3.6) and (3.7) reduce after some manipulation to the seventh-order system of Atten & Moreau (1972) as expected. In the case of a perfectly insulating liquid ($C = 0$) it is possible from (3.5)–(3.7), after assuming $|\epsilon_1| \ll 1$ and that θ and $D\theta$ are order one quantities, to recover the equations of the first model considered by Roberts (1969), which contains the classical Bénard problem as a special case. For the non-polar liquids under consideration we have $|e_1| \ll |k_1|$ and so restrict our attention to the special case $\epsilon_1 = 0$. As a first attempt we further restrict our analysis to stationary instability for which the marginal stability equations are

$$(D^2-k^2)\,\theta = V, \tag{3.10}$$

$$(D^2-k^2)^2 V = -k^2R\theta - k^2AT_s^2E_0\,q(z)\left\{(D^2-k^2)\,F+\frac{\gamma[\gamma+\kappa_1 q^2(z)]}{K^2(z)\,q^4(z)}F\right\}, \tag{3.11}$$

$$\begin{aligned} [\gamma+\kappa_1 q^2(z)]\,V = {} & \kappa_1 AT_sE_0\,q^3(z)\,[K(z)\,D\theta-\kappa_1\theta] \\ & -AT_sK(z)\left\{\begin{array}{l} -\gamma DF \\ +\dfrac{1}{\gamma}K(z)\,q^2(z)\,D[K(z)\,q^2(z)\,(D^2-k^2)\,F] \end{array}\right\}, \end{aligned} \tag{3.12}$$

where

$$\gamma = C/E_0, \quad K(z) = 1+\kappa_1 z, \tag{3.13), (3.14}$$

$$q(z) = \begin{cases} [1+2\gamma\ln(1+\kappa_1 z)/\kappa_1]^{\frac{1}{2}} & \text{for} \quad \kappa_1 \neq 0, \qquad (3.15a) \\ (1+2\gamma z)^{\frac{1}{2}} & \text{for} \quad \kappa_1 = 0, \qquad (3.15b) \end{cases}$$

and together with boundary conditions (3.8) and (3.9) define the linear instability problem whose solution is to be investigated numerically. Once specific values have been assigned to the four parameters A, R, C and κ_1, thus defining a particular physical situation, solving the eigenvalue problem for the voltage parameter T_s for a range of values of wavenumber k produces marginal stability curves in the k, T_s plane corresponding to the various modes of instability. We first however consider certain limiting cases.

3.2. *Small parameter approximations*

In this section we consider the equations resulting from (3.10)–(3.15) on taking the limits $C \to 0$ and $\kappa_1 \to 0$. Each coefficient is expanded in terms of C and κ_1 and on further assuming that $DV \sim V$, etc., only the leading-order terms are retained. We find that $\gamma \simeq C$, $E_0 \simeq 1$ and $K(z) \simeq q(z) \simeq 1$ and, on defining $G(z) = (D^2 - k^2) F(z)$, the system factorizes into second- and seventh-order differential equations. Eliminating V and G from the latter produces

$$D[(D^2-k^2)^3 + k^2 R]\,\theta - k^2 T_s C(C+\kappa_1)(D^2-k^2)\,\theta + k^2 \kappa_1 A T_s^2 C D\theta = 0, \qquad (3.16)$$

which is subject to the four no-slip conditions, the two fixed temperature conditions and the constraint $G(0) = 0$.

If we now postulate that $\kappa_1 = \kappa_0 C^m$ ($m \geqslant 0$) as $C \to 0$ and write

$$T_s = C^\mu \sum_{n=0}^{\infty} T_n C^n,$$

a leading-order analysis shows that: (i) for $0 \leqslant m \leqslant 3$, $\mu = -\frac{1}{2}(m+1)$ and we obtain

$$D[(D^2-k^2)^3 + k^2 R^*]\,\theta = 0, \qquad (3.17)$$

where $$R^* = R + \lim_{C \to 0} \kappa_1 A T_s^2 C;$$

(ii) for $m = 3$, $\mu = -2$ and we obtain

$$D[(D^2-k^2)^3 + k^2 R]\,\theta - k^2 T_l (D^2-k^2)\,\theta + k^2 \kappa_0 A T_l^2 D\theta = 0, \qquad (3.18)$$

where $$T_l = \lim_{C \to 0} T_s C^2;$$

and (iii) for $m > 3$, $\mu = -2$ we obtain

$$D[(D^2-k^2)^3 + k^2 R]\,\theta - k^2 T_l (D^2-k^2)\,\theta = 0, \qquad (3.19)$$

where again $$T_l = \lim_{C \to 0} T_s C^2.$$

Applying the boundary condition $G(0) = 0$ after a single integration of (3.17) leads to the classical Bénard equation with R replaced by R^*. The solution for the most unstable mode is therefore

$$R_c^* \simeq 1708 \quad \text{and} \quad k_c \simeq 3{\cdot}117. \qquad (3.20)$$

If for example $\kappa_1 > 0$ and $R = -20$ we then have

$$T_c \simeq (1728/\kappa_1 AC)^{\frac{1}{2}}, \qquad (3.21)$$

whereas for κ_1 and R both negative we have no solution, i.e. no stationary instability. This special case (i) we term the space charge modified Bénard regime. On remembering that $(D^2-k^2)\,\theta = V$ [cf. (3.10)], we note that (3.19) is clearly equivalent to the fifth-order small injection equations of Atten & Moreau (1972) with the addition of the

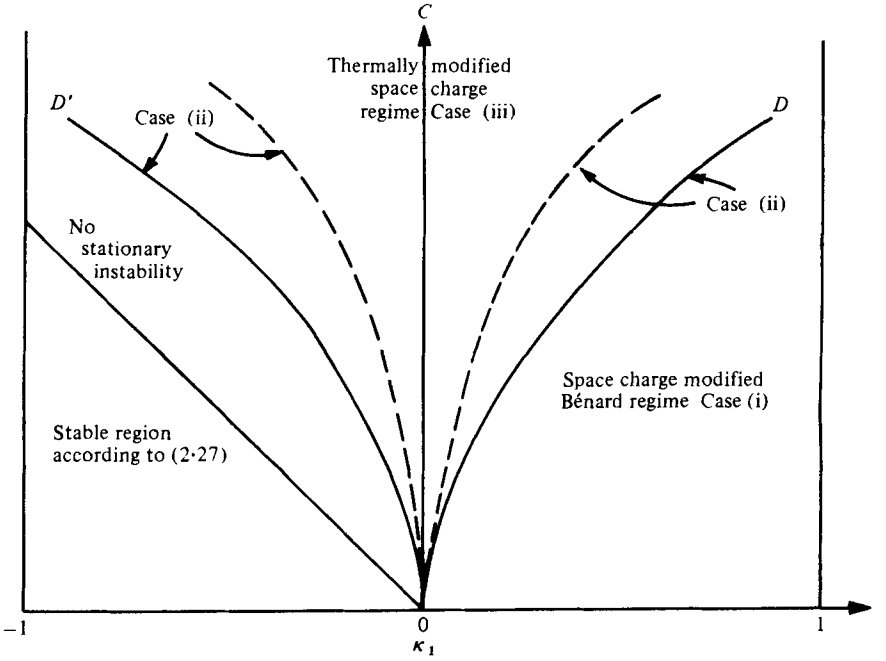

FIGURE 2. Schematic representation of the limiting small C regimes in the κ_1, C plane. The dashed line denotes the smaller extent of case (iii) as A increases.

buoyancy term $k^2R\,D\theta$. This special case (iii) we term the thermally modified space charge regime. The regions in the κ_1, C plane where these cases apply are illustrated in figure 2. Notice that the larger the value of A the smaller the region enclosed by branches OD, OD' (see §4.2).

3.3. *Space-charge-limited currents*

In the limiting case $C\to\infty$ we see from (2.18) to (2.24) that the space-charge-limited current equilibrium has a vanishing electric field E_0 and an associated singularity in space charge density Q_0 at the emitter. An analysis of the perturbation equations (3.11) and (3.12) in this limit indicates that the perturbation δQ in space charge density in the vicinity of $z=0$ behaves like $z^{-\frac{1}{2}}$ in the same way as the equilibrium Q. In order to improve the convergence of the numerical method of solution we therefore filter out from the governing equations the singular behaviour of δQ by introducing a function $F_1(z)$, related to $F(z)$ (cf. Atten & Moreau 1972) and defined by

$$F(z) = F_1(z) - [h(z)/h(1)]^{\frac{3}{2}}F_1(1), \tag{3.22}$$

where

$$h(z) = \begin{cases} \ln(1+\kappa_1 z)/\kappa_1 & \text{for} \quad \kappa_1 \neq 0, \quad (3.23a) \\ z \quad \text{for} \quad \kappa_1 = 0. & (3.23b) \end{cases}$$

In terms of V, θ and F (3.11) and (3.12) become

$$\begin{aligned}(D^2-k^2)^2V = {} & -k^2R\theta \\ & -\frac{k^2[h(z)]^{\frac{1}{2}}}{B}AT_s^2\left\{(D^2-k^2)F_1+\frac{1+2\kappa_1 h(z)}{4K^2(z)\,h^2(z)}F_1\right\} \\ & +\frac{k^2AT_s^2}{B[h(1)]^{\frac{3}{2}}}\left\{\frac{1-\kappa_1 h(z)}{K^2(z)}-k^2h^2(z)\right\}F_1(1), \end{aligned} \tag{3.24}$$

$$[1+2\kappa_1 h(z)]\,V = \frac{2\kappa_1}{B}[h(z)]^{\frac{3}{2}} A T_s [K(z)\,D\theta - \kappa_1\theta]$$
$$- A T_s K(z) \begin{Bmatrix} -DF_1 + 4K^2(z)\,h^2(z)\,D(D^2-k^2)\,F_1 \\ +4K(z)\,h(z)\,[1+\kappa_1 h(z)]\,(D^2-k^2)\,F_1 \end{Bmatrix}$$
$$- A T_s \left[\frac{h(z)}{h(1)}\right]^{\frac{3}{2}} \begin{Bmatrix} 6\kappa_1[2-\kappa_1 h(z)] \\ +k^2K^2(z)\,h(z)\,[10+4\kappa_1 h(z)] \end{Bmatrix} F_1(1), \quad (3.25)$$

where B is a constant depending on κ_1 given by

$$B(\kappa_1) = \int_0^1 [h(z)]^{\frac{1}{2}}\,dz. \quad (3.26)$$

The boundary conditions on velocity and temperature remain unchanged, and those on potential at the emitter are satisfied by the function F_1. However, since the definition of $F_1(z)$ [cf. (3.22)] implies $F(1) = 0$ we must replace this latter condition by an implicit one obtained by differentiating (3.25) once and applying it at $z = 0$. We then have the nine constraints

$$V = DV = \theta = 0 \quad \text{at} \quad z = 0, 1; \quad F_1 = DF_1 = D^2F_1 = 0 \quad \text{at} \quad z = 0. \quad (3.27)$$

4. Numerical analysis

4.1. *Method of solution*

The two ordinary differential eigenvalue problems (3.8)–(3.12) and (3.10), (3.24)–(3.27) were solved numerically on a CDC 7600 machine using a Chebyshev collocation technique providing the critical values of T_s and its associated eigenfunctions $V(z)$, $\theta(z)$ and $F(z)$ for the lowest mode of instability. Each variable is represented by a finite series of Chebyshev polynomials of the same degree N, e.g.

$$V(z) = \sum_{n=0}^{N} V_n T_n(z),$$

so that both systems of equations together with their associated boundary conditions are equivalent to linear algebraic eigenvalue problems whose solutions provide the Chebyshev coefficients. After applying the differential equations at collocation points and appropriately adding in the equations obtained from the boundary conditions we have

$$\mathbf{A}_1\mathbf{V} + \mathbf{B}_1\boldsymbol{\theta} = 0, \quad \mathbf{A}_2\mathbf{V} + \mathbf{B}_2\boldsymbol{\theta} + T_s^2\mathbf{C}_2\mathbf{F} = 0, \quad (4.1), (4.2)$$

$$\mathbf{A}_3\mathbf{V} + T_s\mathbf{B}_3\boldsymbol{\theta} + T_s\mathbf{C}_3\mathbf{F} = 0, \quad (4.3)$$

where $\mathbf{A}_i$, $\mathbf{B}_i$ $(i = 1, 2, 3)$ and $\mathbf{C}_i$ $(i = 2, 3)$ are $(N+1)\times(N+1)$ matrices and $\mathbf{V}$, $\boldsymbol{\theta}$ and $\mathbf{F}$ are vectors containing the respective Chebyshev coefficients. By construction matrices $\mathbf{A}_2$, $\mathbf{B}_1$ and $\mathbf{C}_3$ possess inverses so that eliminating $\boldsymbol{\theta}$ and $\mathbf{F}$ from the system (4.1)–(4.3) gives

$$(\lambda_s^2\mathbf{I} + \lambda_s\mathbf{M}_1 + \mathbf{M}_2)\mathbf{V} = 0, \quad (4.4)$$

where $\lambda_s = T_s^{-1}$, $\mathbf{M}_1 = -\mathbf{A}_2^{-1}(\mathbf{B}_2\mathbf{B}_1^{-1}\mathbf{A}_1 + \mathbf{C}_2\mathbf{C}_3^{-1}\mathbf{A}_3)$, $\mathbf{M}_2 = \mathbf{A}_2^{-1}\mathbf{C}_2\mathbf{C}_3^{-1}\mathbf{B}_3\mathbf{B}_1^{-1}\mathbf{A}_1$ and $\mathbf{I}$ is the $(N+1)\times(N+1)$ identity matrix.

The $2N+2$ eigenvalues λ_s of (4.4) are identical to those of the linear algebraic system

$$(\mathbf{M} - \lambda_s\mathbf{I})\mathbf{X} = 0, \quad (4.5)$$

where

$$\mathbf{M} = \begin{bmatrix} 0 & \mathbf{I} \\ -\mathbf{M}_2 & -\mathbf{M}_1 \end{bmatrix} \quad \text{and} \quad \mathbf{X} = \begin{bmatrix} \mathbf{V} \\ \lambda_s\mathbf{V} \end{bmatrix}. \quad (4.6)$$

For any fixed wavenumber k the lowest positive real value of T_s ($= \lambda_s^{-1}$) then corresponds to the fundamental mode of instability. On constructing the velocity eigenfunction $V(z)$ from the first $N+1$ elements of the corresponding eigenvector $\mathbf{X}$, we find that this mode has a single-cell character. The most unstable mode is then obtained by using a quadratic minimization routine to determine the minimum value of T_s as a function of k.

In most cases the solution was computed using a degree of Chebyshev polynomial representation $N = 25$ and the process repeated for $N = 40$. It was found that the number of significant figures to which corresponding values of T_s agreed was dependent primarily on the value of C, but to a lesser extent on κ_1 and A. Typically for values of $C \lesssim 2$ agreement was to seven figures, for $C = 10$ it was only to three figures, whereas for $C = \infty$ five or six figure agreement was achieved. As a further check on the validity of this numerical technique the solution to the isothermal case $R = \kappa_1 = 0$ was found to be consistent with the results of Atten & Moreau (1972).

The present method of solution of the thermal problem involved solving three equations in three variables. However, it was found that eliminating one of these variables to reduce the amount of algebraic manipulation in the programme resulted in very poor convergence for large C. This was accredited to the application of higher-order boundary conditions.

4.2. *Numerical results*

Applying a weakly stabilizing temperature difference of 1 °C across a layer of chlorobenzene or n-hexane of depth 0·5 mm corresponds to a Rayleigh number $R \simeq -20$. Using measured values of ion mobility leads to values of the mobility parameter A of 0·2 for chlorobenzene ($K_0 = 4{\cdot}5 \times 10^{-8}\,\mathrm{m^2/V\,s}$; cf. Lacroix, Atten & Hopfinger 1975) and 1·5 for n-hexane ($K_0 = 10^{-7}\,\mathrm{m^2/V\,s}$; cf. Gallagher 1975). Most numerical results were obtained by choosing $R = -20$, $A = 0{\cdot}2$ and 20 and κ_1 lying in the ranges $10^{-4} \leqslant \kappa_1 \leqslant 1$ and $-0{\cdot}1 \leqslant \kappa_1 \leqslant -10^{-4}$.

Considering first the case of a cold emitter ($\kappa_1 \geqslant 0$) we see from figure 3 that the critical voltage parameter T_c decreases with increasing $\kappa_1 (= k_1 \beta d = k_1 \Delta T)$, i.e. with increased temperature difference. Furthermore this reduction is greater for (i) smaller values of injection strength C: thus for the case $A = 20$, $C = 10^{-3}$, T_c is reduced from 29404·27 ($\kappa_1 = 10^{-4}$) to 432·2923 ($\kappa_1 = 1{\cdot}0$) whereas for $C = \infty$, T_c is only reduced from 160·3389 ($\kappa_1 = 10^{-4}$) to 17·917 ($\kappa_1 = 1{\cdot}0$); and (ii) for larger values of the mobility parameter A whenever C is large: thus when $C = \infty$, $A = 20$, T_c is reduced from 160·3389 ($\kappa_1 = 10^{-4}$) to 17·917 ($\kappa_1 = 1{\cdot}0$) whereas for $A = 0{\cdot}2$ the reduction is from 162·180 ($\kappa_1 = 10^{-4}$) to 106·451 ($\kappa_1 = 1{\cdot}0$). In addition we notice that in accordance with (3.21) for the space charge modified Bénard regime the curves for small C have gradient approximately $-0{\cdot}5$ (cf. $\ln T_c \sim -\frac{1}{2} \ln \kappa_1 +$ constant). From tables 1 and 2 it is clear that the product $\kappa_1 A$ in the cases computed here is important in determining the nature of the solution not only for small C [cf. (3.21)] but also for larger values.

From figure 4 it is apparent that in general the critical wavenumber k_c decreases with increasing κ_1. It also increases as C becomes larger, though for moderate values of κ_1 this latter effect is more marked the smaller the value of A. Alternatively we can say that the larger the value of A the more extensive is the region of the κ_1, C plane essentially characterized by the space charge modified Bénard instability. This is also evident from plots of T_c against C (figure 5) for various values of κ_1. The curves for small C and $\kappa_1 \neq 0$ have slope $-0{\cdot}5$ [cf. (3.21)], whereas the limiting curve $\kappa_1 = 0$ (for

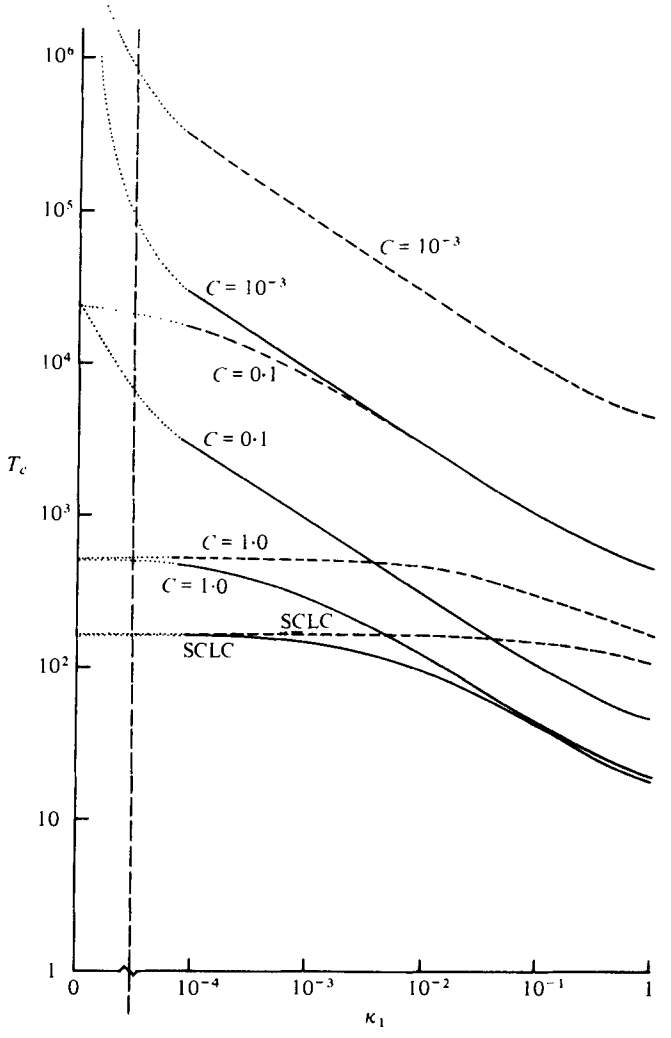

FIGURE 3. Variation of critical voltage parameter T_c with κ_1 for $R = -20$ and $C = 0{\cdot}001$, $0{\cdot}1$, 1, ∞. The solid lines correspond to $A = 20$ and the dashed lines to $A = 0{\cdot}2$.

small C and any value of A) has a slope tending to -2. This corresponds to the thermally modified space charge regime [cf. (3.19)] where, for $R = -20$,

$$\lim_{C\to 0} T_c C^2 = 223{\cdot}0984 \quad \text{and} \quad k_c = 4{\cdot}591,$$

compared with the isothermal results of Atten & Moreau (1972)

$$\lim_{C\to 0} T_c C^2 = 220{\cdot}7621 \quad \text{and} \quad k_c = 4{\cdot}5715.$$

We next consider the case of a hot emitter ($\kappa_1 < 0$) and present some numerical results in table 3. For small values of C the space charge modified Bénard equation [cf. (3.17)] implies, for stationary instability to be possible, that

$$R + \kappa_1 A T_s^2 C \gtrsim 1708. \tag{4.7}$$

C	$\kappa_1 = 0$	$\kappa_1 = 10^{-3}$	$\kappa_1 = 10^{-2}$	$\kappa_1 = 10^{-1}$	$\kappa_1 = 1{\cdot}0$
10^{-3}	$2{\cdot}232906 \times 10^8$	92997·41	29532·41	9742·552	4310·94
	(4·590)	(3·117)	(3·118)	(3·120)	(3·129)
10^{-2}	$2{\cdot}251118 \times 10^6$	29343·47	9346·941	3079·522	1358·338
	(4·590)	(3·131)	(3·125)	(3·130)	(3·144)
10^{-1}	24402·27	8060·134	2896·402	983·843	432·8282
	(4·591)	(3·507)	(3·255)	(3·193)	(3·201)
1	516·2250	509·5479	461·5147	298·352	161·3287
	(4·700)	(4·677)	(4·514)	(3·988)	(3·556)
10	165·6	165·4	164	151	120·7
	(5·138)	(5·135)	(5·113)	(4·945)	(4·493)
∞	162·199	162·0046	160·303	146·964	106·451
	(5·164)	(5·162)	(5·141)	(4·982)	(4·561)

TABLE 1. Values of the critical voltage parameter T_c and associated wavenumber k_c for $\kappa_1 \geqslant 0$, $R = -20$, $A = 0{\cdot}2$.

C	$\kappa_1 = 0$	$\kappa_1 = 10^{-3}$	$\kappa_1 = 10^{-2}$	$\kappa_1 = 10^{-1}$	$\kappa_1 = 1{\cdot}0$
10^{-3}	$2{\cdot}232906 \times 10^8$	9302·888	2954·967	975·7901	432·2923
	(4·590)	(3·116)	(3·116)	(3·117)	(3·123)
10^{-2}	$2{\cdot}251118 \times 10^6$	2951·63	937·8324	309·6255	137·0432
	(4·590)	(3·118)	(3·117)	(3·118)	(3·124)
10^{-1}	24402·27	953·7064	306·5797	101·4390	44·61840
	(4·591)	(3·158)	(3·131)	(3·125)	(3·133)
1	516·2250	293·6654	122·6839	43·81576	18·7739
	(4·700)	(3·954)	(3·466)	(3·276)	(3·220)
10	165·6	149	96	42	19·6
	(5·138)	(4·930)	(4·338)	(3·792)	(3·551)
∞	162·199	146·63	96·079	42·382	17·917
	(5·164)	(4·967)	(4·384)	(3·824)	(3·561)

TABLE 2. Values of the critical voltage parameter T_c and associated wavenumber k for $\kappa_1 \geqslant 0$, $R = -20$, $A = 20$.

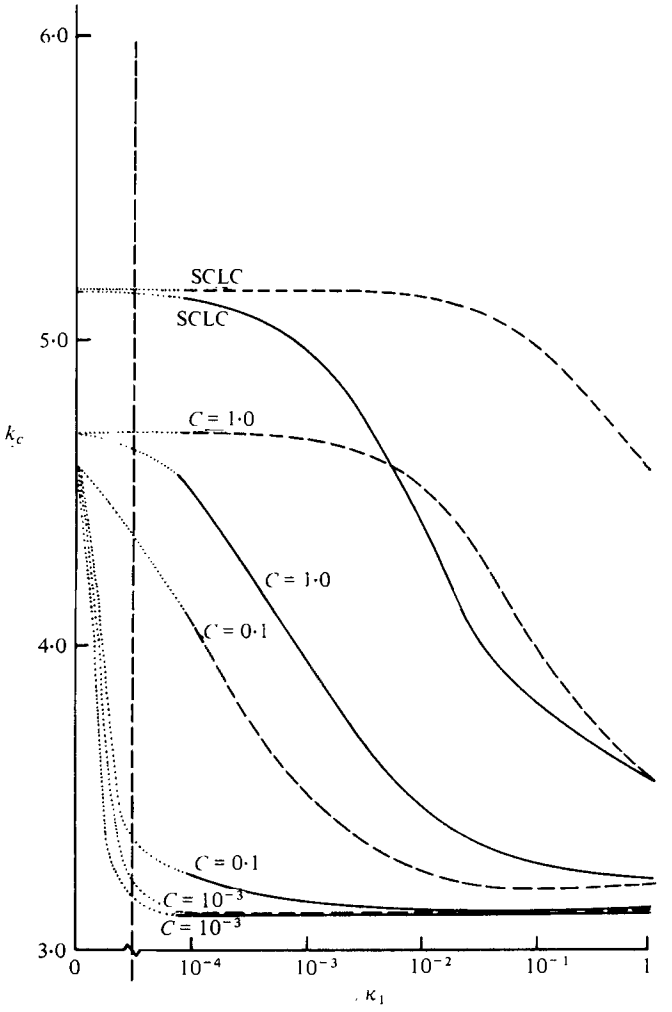

FIGURE 4. Variation of critical wavenumber k_c with κ_1 for $R = -20$ and $C = 0{\cdot}001$, $0{\cdot}1$, 1, ∞. The solid lines correspond to $A = 20$ and the dashed lines to $A = 0{\cdot}2$.

This suggests that for negative values of κ_1, since A and C are inherently positive, the electric field exerts a stabilizing influence in that it reduces the effective Rayleigh number. Indeed for $R \lesssim 1708$ no stationary instability is possible. As before the parameter $\kappa_1 A$ is important in characterizing the solution even for large values of C. Thus for $C = \infty$ the solution for $A = 0{\cdot}2$, $\kappa_1 = -10^{-2}$ is $T_c = 164{\cdot}2095$ and $k_c = 5{\cdot}189$, whereas for $A = 20$, $\kappa_1 = -10^{-4}$ we have $T_c = 164{\cdot}1482$ and $k_c = 5{\cdot}190$. As $|\kappa_1 A|$ increases the critical voltage parameter T_c and corresponding wavenumber k_c increase. The convergence of the Chebyshev series approximations becomes progressively poorer suggesting the approach to a stability boundary (cf. figure 2). Where, however, a solution has been obtained the character of the instability is of a thermally modified space charge kind.

Finally in the case of space-charge-limited currents ($C = \infty$) we find that as R

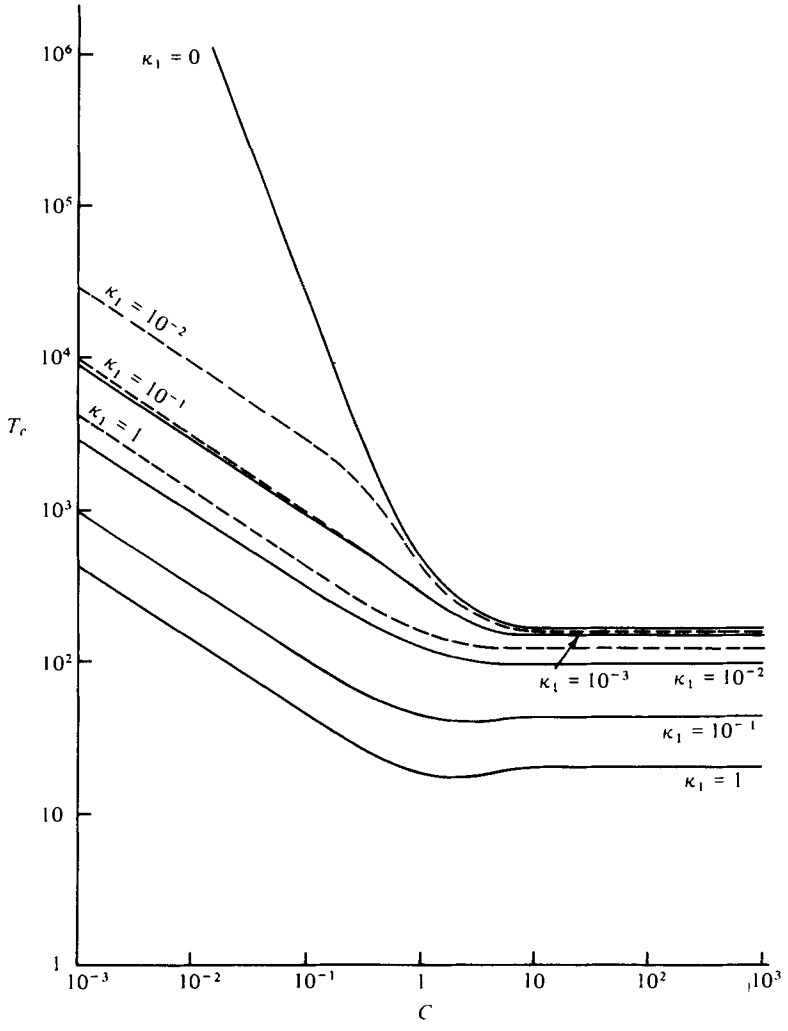

FIGURE 5. Variation of critical voltage parameter T_c with injection strength C for $R = -20$. (*a*) $A = 0{\cdot}2$, $\kappa_1 = 0$, 0·01, 0·1, 1 (dashed line); (*b*) $A = 20$, $\kappa_1 = 0$, 0·001, 0·01, 0·1, 1 (solid line).

	$\kappa_1 = 0$	$\kappa_1 = -10^{-4}$	$\kappa_1 = -10^{-3}$	$\kappa = -10^{-2}$	$\kappa = -10^{-1}$
$A = 0{\cdot}2$					
$C = 1$	516·2250	516·9121	523·2660	612·3832	—
	(4·700)	(4·702)	(4·724)	(5·033)	—
$C = \infty$	162·199	162·2187	162·3949	164·2095	190·41
	(5·164)	(5·164)	(5·167)	(5·189)	(5·539)
$A = 20$					
$C = 1$	516·2250	609·153	—	—	—
	(4·700)	(5·027)	—	—	—
$C = \infty$	162·199	164·1482	187·512	—	—
	(5·164)	(5·190)	(5·511)	—	—

TABLE 3. Values of critical voltage parameter T_c and associated wavenumber k_c for $\kappa_1 \leqslant 0$, $R = -20$, $A = 0{\cdot}2$ and 20 in the cases $C = 1$ and $C = \infty$. A line indicates that no solution was found.

	$R = 20$	$R = -20$	$R = -2 \times 10^3$	$R = -2 \times 10^4$
$\kappa_1 = 0$	159·3003	162·199	276·2265	658·623
	(5·113)	(5·164)	(7·282)	(13·518)
$\kappa_1 = 0{\cdot}1$				
$A = 0{\cdot}2$	144·504	146·964	242·710	590·06
	(4·937)	(4·982)	(6·802)	(12·721)
$A = 20$	41·841	42·382	64·112	No
	(3·808)	(3·824)	(4·566)	solution

TABLE 4. Values of the critical voltage parameter T_c and associated wavenumber k_c for different values of the Rayleigh number R in the case of space-charge-limited current. Note that the results do not depend on A when $\kappa_1 = 0$.

becomes more negative (cf. table 4), corresponding to a more strongly stable thermal stratification, the critical values T_c and k_c both increase. They also increase as the mobility parameter A is reduced.

5. Concluding remarks

Perhaps the most striking feature of the above analysis is that it demonstrates the importance of the sign of the temperature gradient in relation to the emitting electrode. It suggests that a system with an emitter cooler than the collector is more susceptible to stationary instability than one with the opposite temperature gradient. Indeed, in the hot emitter case the electric field may for sufficiently large $|\kappa_1 A|$ fail to destabilize an otherwise thermally stable layer, although oscillatory instabilities have not been ruled out. This may exert a considerable influence on the practical design of a heat exchanger using electrohydrodynamic principles.

The second feature of note is the effect of the mobility variation in the two types of instability according to the values of injection strength C, mobility variation parameter κ_1 and mobility parameter A. In the Bénard type mode the electric field, depending upon the sign of κ_1, acts to increase or reduce the effective Rayleigh number, i.e. it tends to counteract or reinforce gravity. In the other, which is essentially a space charge mode of the Atten & Moreau type modified by buoyancy, its effect is of secondary importance; this latter mode will be dominant in the case of small $|\kappa_1 A|$ and large C. When $C \ll 1$ we notice that the dependence of the critical voltage parameter T_c upon C is quite different for these two modes of instability.

It is evident that the magnitude of $\kappa_1 A$ plays a central role in characterizing the precise form of the instability. This may be exemplified by an argument analogous to that used by Felici (1971) with reference to Bénard convection (see figure 6). Ignoring for the moment buoyancy effects and the positive feedback mechanism involving circulations and gradients in space charge, we consider the effect of thermally induced variations in mobility across the layer. The constancy of current [cf. (2.18)] implies that as mobility K varies the electrical body force QE varies reciprocally. An imbalance in the body force per unit volume $\Delta(QE) \simeq j_0 \kappa_1 / K_0$ is then established between emitter and collector regions, which must, in steady convection, be balanced by viscous drag $\sim \rho_0 \nu U / d^2$, where U is a characteristic velocity, so that $U \sim j_0 \kappa_1 d^2 / \rho_0 \nu K_0$. The cool rising columns of liquid will gain heat by conduction from the adjacent falling

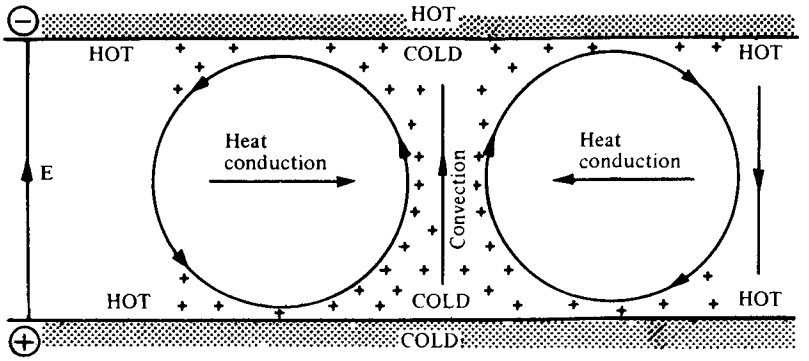

FIGURE 6. Thermally induced electroconvection: the cold liquid rises owing to the electrical body force imbalance and after gaining heat falls back having lost its electrical buoyancy. The process is limited by heat conduction between falling and rising liquid columns.

columns so that the imbalance in body force driving the motion is reduced. Growth of liquid circulations will be limited when the heat conducted into a single cell $\sim \lambda\beta d^2$, where λ is the thermal conductivity, becomes comparable with the convective heat flux $\sim \rho_0 c_p \beta U d^3$, c_p being the specific heat at constant pressure. This balance condition gives $U \sim \lambda/\rho_0 c_p d = \kappa/d$ and combined with the balance of forces implies

$$j_0 \kappa_1 d^3/\rho_0 \nu\kappa K_0 \sim 1. \tag{5.1}$$

In the case of small injection for which $E_0 \simeq \phi_0/d$ so that $j_0 \simeq K_0 Q_0 \phi_0/d$ condition (5.1) becomes

$$\kappa_1 d^2 Q_0 \phi_0/\rho_0 \nu\kappa = \kappa_1 A T_s^2 C \sim 1, \tag{5.2}$$

[cf. (3.17) and the definition of R^*], whereas for strong injection $j_0 \sim \epsilon_0 K_0 \phi_0^2/d^3$ so that (5.1) becomes

$$\kappa_1 \epsilon_0 \phi_0^2/\rho_0 \nu\kappa = \kappa_1 A T_s^2 \sim 1. \tag{5.3}$$

Whilst this is not a rigorous analysis it nevertheless suggests the importance of the parameter $\kappa_1 A$. In the case of negative κ_1, i.e. a hot emitting electrode, the imbalance in the electrical body force is reversed, thus tending to stabilize the system in a manner similar to that of the body force in a Bénard layer heated from above. This is consistent with the energy argument leading to (2.27), once again highlighting the distinction between the cases of hot and cold emitters.

The applicability of the above stability analysis to the experiments of Gross & Porter (1966) and Gross (1967) on a layer of transformer oil of depth 1 mm, across which was applied a stabilizing temperature difference of 12 °C, depends critically on the value assumed for ion mobility within the oil. Taking the values $R \simeq -20$, $|\kappa_1| \simeq 0{\cdot}25$ and assuming that the threshold voltage for instability was 180 V we find that (i) if $K_0 \simeq 3 \times 10^{-10}$ m^2/V s then $A \simeq 2 \times 10^{-3}$ and the experimental value of $T_c \simeq 340$. The present model provides a similar value for T_c on choosing $C \simeq 2$ regardless of the sign of κ_1. This would imply that the thermal effects are of little significance and the instability is essentially of the space charge type; (ii) if $K_0 \simeq 10^{-9}$ m^2/V s then $A \simeq 2 \times 10^{-2}$ and the experimental value of $T_c \simeq 100$. Then even for $\kappa_1 > 0$ and assuming SCLC conditions the model does not appear to account for the instability. However, the degree to which the test liquid was filtered would have affected any experimental results. Nevertheless, the effect of temperature dependent mobility

should be better exhibited if the test liquid is chosen to have a higher value of A (e.g. n-hexane).

Finally we note that whilst we have isolated thermally induced variations of the charge carrier mobility we have ignored variations of kinematic viscosity, thermal conductivity and electrical permittivity, the effects of charge diffusion and the underlying electrochemical electrode reactions. We have also only considered unipolar injection. Undoubtedly many situations arise for which a bipolar injection model including recombination and dissociation effects would be more appropriate. Once again neither the possibility of overstability nor the effects of nonlinearities have been considered.

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