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PHYSICS

Electrohydrodynamic Instability of Dielectric Liquids in High Electric Fields and Decay into an Anisotropic Two-Phase Vapor–Liquid System

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Stable states of matter are off the curve of coexistence of phases; moreover $\left(\frac{\partial p}{\partial \rho}\right)_T > 0$. A homogeneous state below the coexistence curve can be either metastable $\left(\frac{\partial p}{\partial \rho}\right)_T > 0$ or unstable $\left(\frac{\partial p}{\partial \rho}\right)_T < 0$. Metastable states may exist for a relatively long time and then decay into liquid and vapor through the process of heterogeneous and homogeneous nucleation [1, 2]. Unstable states are thermodynamically prohibited and matter decays very fast through spinodal decay [3]. Experiments [4] revealed that an electric field influences the region of liquid stability. Earlier [5], it was theoretically demonstrated that the critical point and, hence, the curve of phase coexistence are shifted under the action of electrostriction forces.

In this paper, we reveal a previously unknown mechanism of instability of dielectric liquids in strong electric fields, namely, anisotropic decay into a two-phase system of vapor channels in the liquid oriented along the field.

The volume force acting upon a charged dielectric liquid in an electric field E is given by the Helmholtz formula [5]

$$\mathbf{F} = q\mathbf{E} - \frac{E^2}{8\pi}\nabla\varepsilon + \frac{1}{8\pi}\nabla\left[E^2\rho\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T\right]. \quad (1)$$

For gases and liquids with weakly polarizable molecules, the permittivity is a linear function of the density:

$$\varepsilon = 1 + 3\alpha\rho, \quad (2)$$

where $\alpha = \frac{4\pi\beta}{3m}$, β is the polarizability, and m is the molecular mass. Nonpolar liquids obey the Clausius–Mosotti law [6]

$$\varepsilon = 1 + \frac{3\alpha\rho}{1-\alpha\rho} \quad (3)$$

polar liquids obey the Onsager–Kirkwood–Fröhlich relation [6], though experimental values of quantities $\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T$ and $\left(\frac{\partial^2\varepsilon}{\partial\rho^2}\right)_T$, which are usually positive, are more reliable.

We have performed a linear stability analysis of the Euler equations [7] for homogeneous dielectric liquids in a uniform electric field in the simplest isothermal case (obviously, accompanied by the corresponding supply and removal of heat).

Let a uniform field be applied between two plane horizontal electrodes, so that only the vertical projection of the electric field vector E_z is nonzero. We consider the growth of small one-dimensional density and velocity perturbations corresponding to stratification along the field

$$\rho = \rho_0 + A_0\exp(\gamma t)\exp\frac{i\cdot 2\pi x}{\lambda},$$
$$u_x = B_0\exp(\gamma t)\exp\frac{i\cdot 2\pi x}{\lambda} \quad (4)$$

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and to the stratification across the electric field

$$\rho = \rho_0 + A_0 \exp(\gamma t) \exp\frac{i \cdot 2\pi z}{\lambda},$$
$$u_z = B_0 \exp(\gamma t) \exp\frac{i \cdot 2\pi z}{\lambda}, \quad (5)$$

where λ is the wavelength, A_0 and B_0 are the initial amplitudes of perturbations, γ is the instability increment, and ρ_0 is the average density of the fluid.

In the case of perturbations (4), the volume force can be written as

$$F_x = \frac{E_0^2 \rho}{8\pi}\left(\frac{\partial^2 \varepsilon}{\partial \rho^2}\right)_T \frac{\partial \rho}{\partial x} = K_x \frac{\partial \rho}{\partial x}, \quad (6)$$

where E_0 is the electric field strength. For perturbations (5), we have

$$F_z = \frac{D_0^2 \rho}{8\pi\varepsilon^2}\left(\left(\frac{\partial^2 \varepsilon}{\partial \rho^2}\right)_T - \frac{2}{\varepsilon}\left(\frac{\partial \varepsilon}{\partial \rho}\right)_T^2\right)\frac{\partial \rho}{\partial z} = K_z \frac{\partial \rho}{\partial z}, \quad (7)$$

where D_0 is the electric displacement, which is in this case constant in space. In both cases, the instability increment and the equation of the spinodal are given as

$$\gamma = \frac{2\pi}{\lambda}\sqrt{-\left(\frac{\partial p}{\partial \rho}\right)_T + K}, \quad (8)$$

$$\left(\frac{\partial p}{\partial \rho}\right)_T = K. \quad (9)$$

For the growth of two-dimensional perturbations (like cylindrical channels parallel to the z axis, provided that K_y is described by a relation analogous to Eq. (6)),

$$\rho = \rho_0 + A_0 \exp(\gamma t) \exp\frac{i \cdot 2\pi x}{\lambda} \cdot \exp\frac{i \cdot 2\pi y}{\lambda},$$
$$u_x = B_0 \exp(\gamma t) \exp\frac{i \cdot 2\pi x}{\lambda} \cdot \exp\frac{i \cdot 2\pi y}{\lambda}, \quad (10)$$
$$u_y = Q_0 \exp(\gamma t) \exp\frac{i \cdot 2\pi x}{\lambda} \cdot \exp\frac{i \cdot 2\pi y}{\lambda},$$

the increment is greater by a factor of $\sqrt{2}$ than the value for the perturbations (4).

The instability increment (8) exhibits infinite growth with decreasing λ. However, for viscous fluids (in considering the stability of one-dimensional Navier–Stokes equations [7]), the instability increment is limited

$$\gamma = \frac{2\pi}{\lambda}\left(\sqrt{\left(\frac{\pi b}{\lambda}\right)^2 + K - \left(\frac{\partial p}{\partial \rho}\right)_T} - \frac{\pi b}{\lambda}\right), \quad (11)$$

i.e., it tends to a constant value $\gamma_{\max} \approx \left(K - \left(\frac{\partial p}{\partial \rho}\right)_T\right) b^{-1}$ for $\lambda < \lambda_* \sim 2\pi b\left(\sqrt{K - \left(\frac{\partial p}{\partial \rho}\right)_T}\right)^{-1}$. Here, $b = \frac{4/3\mu + \xi}{\rho_0}$, μ is the dynamic viscosity, and ξ is the second viscosity coefficient [7]. Thus, the regard for viscosity does not change the equation of spinodal (9).

Equations (6) and (7) yield that $K_z < K_x$; therefore, the instability increment depends on the orientation of perturbations.

For both polar and nonpolar liquids, we have $K_x > 0$, which implies that the electric field increases the instability increment of perturbations of the type described by Eq. (4). At the same time, the stability of fluid with respect to stratification across the field increases, because $K_z < 0$. Thus, for $K_x > \left(\frac{\partial p}{\partial \rho}\right)_T$, electrostriction forces cause decay of a homogeneous fluid into an anisotropic two-phase system of vapor filaments located in the liquid and oriented along the electric field.

For gas-type law (2), we have $K_x = 0$. Therefore, instability is possible only in the region of forbidden states $\left(\frac{\partial p}{\partial \rho}\right)_T < 0$, as well as in the absence of an electric field. Note that, even in this case, the instability is anisotropic because $K_z = \frac{-D_0^2(\varepsilon - 1)^2}{4\pi\varepsilon^3\rho} < 0$.

As is known, an electric field shifts the critical point both in temperature and density [5]. The equation of spinodal $\left(\frac{\partial p}{\partial \rho}\right)_T = K_x$ obtained from the condition of hydrodynamic stability exactly coincides with the boundary of thermodynamic stability for dielectric liquids as determined in [5]:

$$\left(\frac{\partial p}{\partial \rho}\right)_T = \frac{E_0^2 \rho}{8\pi}\left(\frac{\partial^2 \varepsilon}{\partial \rho^2}\right)_T. \quad (12)$$

At the same time, monograph [5] did not take into account the possibility that the instability and decay of a dielectric can be anisotropic.

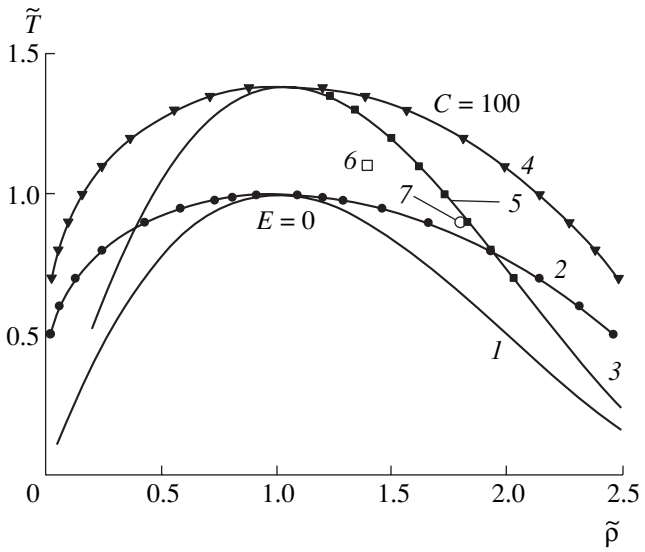

Fig. 1. Diagrams showing (*2*, *4*) the coexistence curves and (*1*, *3*) theoretical spinodals (15) for the van der Waals equation calculated (*1*, *2*) in the absence of an electric field and (*3*, *4*) in a uniform electric field; (*5*) points of the spinodal obtained in hydrodynamic calculations; (*6*, *7*) states which are initially above the critical point ($\tilde{\rho}_0 = 1.4$; $\tilde{T} = 1.1$) and in the region of stable liquid ($\tilde{\rho}_0 = 1.8$; $\tilde{T} = 0.9$), respectively.

Let us consider a liquid obeying the van der Waals equation of state (written in dimensionless variables)

$$\tilde{p} = \frac{8\tilde{T}\tilde{\rho}}{3-\tilde{\rho}} - 3\tilde{\rho}^2. \quad (13)$$

For nonpolar liquids, relation (3) yields

$$K_x = \frac{E_0^2}{36\pi}(\varepsilon - 1)^2\frac{\varepsilon + 2}{\rho}, \quad (14)$$

and an expression for the spinodal can be written in an explicit form as

$$\tilde{T} = \frac{\tilde{\rho}(3-\tilde{\rho})^2}{4}\left(1 + C\frac{(\alpha\rho_{cr})^2}{(1-\alpha\rho_{cr}\tilde{\rho})^3}\right), \quad (15)$$

where the second term in parentheses describes the temperature shift of the critical point. Here, $C = \frac{E_0^2}{8\pi p_{cr}}$ is the dimensionless magnitude of the electric field squared. The shift of the critical point in density $\Delta\tilde{\rho}_{cr} \sim 2(\alpha\rho_{cr})\Delta\tilde{T}$ is small due to the smallness of $\alpha\rho_{cr} \ll 1$. For the values of parameters corresponding to argon ($T_{cr} = 151$ K, $\rho_{cr} = 531$ kg/m^3, $p_{cr} = 4.86$ MPa, and $\alpha\rho_{cr} = 0.057$), the spinodals for $E = 0$ and $C = 100$ are shown in Fig. 1.

The evolution (electrohydrodynamics) of a homogeneous dielectric liquid that is initially at rest in a uniform electric field was simulated using the method of lattice Boltzmann equations with phase transitions [8] with allowance for the action of electric forces on the liquid [9]. The boundary conditions of the x axis direction were periodic. Random density fluctuations $\frac{\Delta\rho}{\rho_0} \sim 10^{-6}$ were set. The electrodes were characterized by neutral wetting (contact angle was set to $\frac{\pi}{2}$). The electric field distribution was determined by solving the system of equations

$$\mathrm{div}(\varepsilon\nabla\varphi) = 0, \quad \mathbf{E} = -\nabla\varphi \quad (16)$$

with the corresponding boundary conditions $\varphi = 0$ and $\varphi = E_0 L_y$ on the lower and upper electrodes, respectively. The size of the computational area was 150×150 nodes.

For a dielectric liquid with the values of the parameters corresponding to argon, the results of numerical calculations were used to plot the coexistence curves in the absence of a field (Fig. 1, curve *2*) and in the initial vertical uniform electric field (curve 4), as well as a part of the spinodal (points *5*). A sufficiently strong field makes possible the anisotropic instability for metastable and even stable states (for instance, states *6* and *7* in Fig. 1). The same effects will be observed for polar dielectric liquids.

Indeed, the results of computer simulation showed evidence of stratification along the uniform electric field ($C = 100$) for a fluid that was initially in the states both above the critical point (Figs. 2a, 2b) and in the liquid state (Figs. 2c, 2d). It is important that the less dense phase is developed in the form of channels of an approximately cylindrical shape oriented along the field. They expand generating compression waves (Fig. 2d) up to the values corresponding to the shifted curve of coexisting phases; this is a cooperative effect in the theory of nucleation [2].

In preceding investigations, the possibility of nucleation was considered only for spherical or ellipsoidal bubbles, but not for vapor channels (see [10, 11], etc.). The mechanism suggested in [10] involves the development of cracks (as in solids) in a liquid containing a population of spherical microbubbles. However, for the case of linear dependence $\varepsilon(\rho)$ considered in [10], the anisotropic instability is possible only for initially unstable states. Therefore, the mechanism [10] is fun-

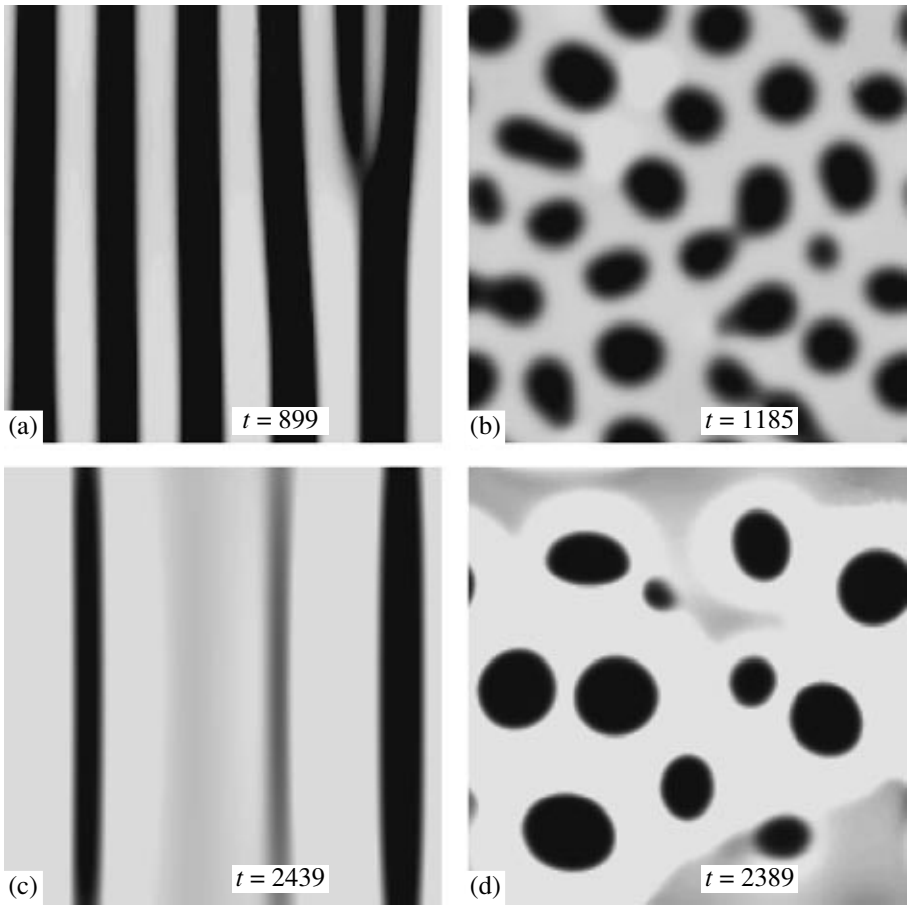

Fig. 2. Anisotropic decay of a dielectric liquid under the action of a uniform electric field: (a, c) stratification along the initial vertical field; (b, d) development of instability in the *xy* plane perpendicular to the field. Dark regions correspond to lower density of the fluid; (a, b) $\tilde{\rho}_0 = 1.4$; $\tilde{T} = 1.1$ (state *6* in Fig. 1); (c, d) $\tilde{\rho}_0 = 1.8$; $\tilde{T} = 0.9$ (state *7* in Fig. 1).

damentally different from the mechanism of anisotropic instability.

In the process of breakdown of dielectric liquids in strong electric fields (with local field strengths reaching 1–100 MV/cm for various liquids), the anisotropic instability described above is probably the key mechanism of inception of streamer structures and their ultra-fast propagation (at a velocity exceeding 100 km/s [12]) in the form of thin branching filaments oriented on average along the local electric field (Figs. 2a, 2c). In the process of formation of a gas phase, an electric breakdown occurs in a certain channel. As a result, the electric field in the neighboring channels decreases and these channels disappear. The electric field ahead of the conductive filament, on the contrary, is enhanced, and the anisotropic instability is developed in a new region of the dielectric liquid, where the states are below the local spinodals. This process can propagate very rapidly step by step.

Thus, an earlier unknown mechanism of instability of dielectric liquids in electric fields of extreme strengths, namely, the mechanism of anisotropic decay into a two-phase system of vapor filaments in a liquid, has been revealed, investigated, and illustrated by computer simulation. The described mechanism of the gas phase formation is probably the key point in the inception of streamer structures and their ultra-fast propagation.

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