Anisotropic instability of a dielectric liquid in a strong uniform electric field: Decay into a two-phase system of vapor filaments in a liquid

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The linear stability analysis of dielectric liquid placed in uniform electric field with respect to perturbations of density of an initially uniform state was carried out. The electric field increases the instability increment for the stratification along the field and decreases it for the transversal stratification. Thus, anisotropic separation into liquid and vapor phases is possible in high electric fields for a liquid that is initially in unstable state, as well as in metastable or stable states. Computer simulations of electrohydrodynamics confirm the theoretical calculations. It is important that new regions of low density phase appear as narrow cylindrical channels oriented along the field. This mechanism of generation of gaseous phase in locally high electric field can play a key role in processes of inception and ultrafast propagation of streamers during breakdown of liquid dielectrics in nanosecond range.

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I. INTRODUCTION

For most substances, liquid (high-density) and saturated vapor (low-density) phases can coexist in an interval of temperatures between the triple point and the critical point. The density of a liquid decreases with an increase of temperature, the density of a vapor increases. These densities become equal at the critical temperature. A homogeneous state below the coexistence curve with a density intermediate between equilibrium values for a liquid and a vapor can be either metastable [if $(\partial p/\partial\rho)_T>0$] or unstable [$(\partial p/\partial\rho)_T<0$]. A metastable state can exist for a relatively long time, and finally it decays into a two-phase system of pure liquid and vapor through the process of homogeneous or heterogeneous nucleation [1,2]. Unstable states are thermodynamically prohibited, a homogeneous matter cannot exist and decay very fast through a spinodal decomposition [3]. Experiments [4,5] revealed that electric field influences the region of liquid stability. The shift of critical point in temperature was theoretically calculated earlier in Ref. [6]. Hence, the position of regions of different stability can be changed by external fields acting on a matter. The influence of an electric field is of particular theoretical and practical interest.

II. ANISOTROPIC INSTABILITY

The body force acting on charged dielectric liquid in electric field is given by the Helmholtz formula [6]

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

The second and third terms represent the action of electric field on polarization charges in nonuniform dielectric and the electrostriction force.

We focus our attention on ideal dielectrics with zero free charge, hence, the first term in (1) falls out. If, moreover, the temperature is constant, we can rewrite the body force as

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

For gases and liquids with weakly polarizable molecules, the permittivity depends linearly on fluid density,

$$\varepsilon=1+3\alpha\rho, \tag{3}$$

where $\alpha=4\pi\beta/(3m)$, β is the molecular polarizability, m is the mass of a molecule.

For nonpolar liquids, the density dependence of permittivity is given by the Clausius -Mosotti law [7]

$$\varepsilon=1+3\alpha\rho/(1-\alpha\rho). \tag{4}$$

For polar liquids, the analytical formula of Onsager, Kirkwood, and Fröhlich is rather complicated, and it is more reliable to use the experimental values of $\varepsilon,(\partial\varepsilon/\partial\rho)_T,(\partial^2\varepsilon/\partial\rho^2)_T$ that are usually positive.

Linear stability analysis of the Euler equations [8]

$$\frac{\partial\rho}{\partial t}+\frac{\partial\rho u_i}{\partial x_i}=0,\quad \frac{\partial\rho u_i}{\partial t}+\frac{\partial(p\delta_{ij}+\rho u_i u_j)}{\partial x_j}=F_i \tag{5}$$

for dielectric liquid under the action of electric field in the simplest isothermal case was carried out. Obviously, the isothermality of liquid should be imposed externally by forced heat inflow and removal.

A constant voltage was applied between two plane horizontal electrodes. In this case, only the vertical component of electric field E_z is nonzero. Let us consider a growth of small one-dimensional harmonic perturbations of density and velocity corresponding to the stratification of matter along the electric field,

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$$\rho = \rho_0 + A_0 \exp(\gamma t)\exp(i2\pi x/\lambda),$$

$$u_x = C_0 \exp(\gamma t)\exp(i2\pi x/\lambda), \quad (6)$$

and to the stratification across the electric field

$$\rho = \rho_0 + A_0 \exp(\gamma t)\exp(i2\pi z/\lambda),$$

$$u_z = C_0 \exp(\gamma t)\exp(i2\pi z/\lambda), \quad (7)$$

where λ is the wavelength, A_0, C_0 are the initial amplitudes of perturbations, γ is the instability increment, ρ_0 is the mean density of a matter.

In the case of the perturbation (6), the magnitude of uniform electric field E_0 is constant. Hence, the body force (2) acting on a matter, has the form

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

For the perturbation (7), layers with different permittivity are perpendicular to the electric field. Therefore, electric displacement D_0 is constant everywhere in space due to well-known boundary conditions of continuity of the normal component of electric displacement. Hence, the electric field is $E=D_0/\varepsilon$, and we have

$$F_z = \frac{D_0^2\rho}{8\pi\varepsilon^2}\left[\left(\frac{\partial^2\varepsilon}{\partial\rho^2}\right)_T - \frac{2}{\varepsilon}\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T^2\right]\frac{\partial\rho}{\partial z} = K_z\frac{\partial\rho}{\partial z}. \quad (9)$$

In both cases, the instability increment is given by

$$\gamma = \frac{2\pi}{\lambda}\sqrt{-\left(\frac{\partial p}{\partial\rho}\right)_T + K}. \quad (10)$$

From (8) and (9), the inequality $K_z < K_x$ always follows, hence, the equation of spinodal is given by

$$(\partial p/\partial\rho)_T = K_x. \quad (11)$$

Similarly, the linear stability analysis of the one-dimensional Navier-Stokes equations [8],

$$\frac{\partial\rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} = 0,$$

$$\frac{\partial(\rho u)}{\partial t} + \frac{\partial(p+\rho u^2)}{\partial x} = F + (4/3\mu + \xi)\frac{\partial^2 u}{\partial x^2} \quad (12)$$

was carried out. Here, μ is the dynamic viscosity of a fluid, ξ is the coefficient of second viscosity.

The instability increment (10) increases as wavelength of perturbations decreases and tends to infinity as wavelength tends to zero. However, taking into account the fluid viscosity gives the following value of instability increment for small perturbations:

$$\gamma = \frac{2\pi}{\lambda}\left[\sqrt{(\pi b/\lambda)^2 + K - (\partial p/\partial\rho)_T} - \pi b/\lambda\right], \quad (13)$$

where the quantity $b=(4/3\mu+\xi)/\rho_0$. For viscous fluid, the divergence of the instability increment for short waves does not take place, and its value is almost constant $\gamma_{\max} \approx [K-(\partial p/\partial\rho)_T]/b$ for wave lengths smaller than $\lambda_* \sim 2\pi b/\sqrt{K-(\partial p/\partial\rho)_T}$. Hence, the increase of instability increment is limited by viscous forces. The viscosity does not change the boundary of instability on the $\tilde{T}$-$\tilde{\rho}$ diagram obtained from (11).

The expressions for instability increment for small perturbations along and across the electric field for a specific liquid are determined by the form of the density dependence of permittivity. The values for K are different for stratifications along and across the field. Hence, the instability increment depends on the orientation of initial perturbations.

For both the polar and nonpolar dielectric liquids $K_x > 0$, hence, the electric field increases the instability increment for perturbation of type (6). In all cases considered, we have $K_z < 0$, hence, the stability of a matter with respect to the stratification across the field is increased. Thus, for $K_x > (\partial p/\partial\rho)_T$, the anisotropic decay of homogeneous fluid into system of vapor filaments in a liquid parallel to the field occurs.

Considering growth of two-dimensional harmonic perturbations in the x-y plane, we obtain the instability increments which are $\sqrt{2}$ times greater than (10) and (13) and a round cross section of growing channels.

For the "gas" law (3) we have $K_x=0$, hence, the instability is possible only in the region of forbidden states $(\partial p/\partial\rho)_T < 0$, the same as in the case without electric field. However, even in this case, the instability is anisotropic. Indeed, the stability for stratification across the field is increased due to the negative coefficient $K_z = -D_0^2(\varepsilon-1)^2/(4\pi\varepsilon^3\rho)$.

III. SHIFT OF THE SPINODAL

It is well known, that in an electric field the critical point is shifted both in temperature and density [6]. The equation of the spinodal curve

$$(\partial p/\partial\rho)_T = \frac{E_0^2\rho}{8\pi}\left(\frac{\partial^2\varepsilon}{\partial\rho^2}\right)_T \quad (14)$$

follows immediately from Eqs. (8) and (11). This boundary of hydrodynamic stability exactly coincides with the boundary of thermodynamic stability of dielectric liquids obtained in Ref. [6].

However, the possibility of anisotropic instability and, consequently, the possible stratification of a matter were not considered in Ref. [6].

As an example, we considered a fluid with the van der Waals equation of state in reduced variables

$$\tilde{p} = 8\tilde{T}\tilde{\rho}/(3-\tilde{\rho}) - 3\tilde{\rho}^2. \quad (15)$$

For nonpolar liquids (4) we have

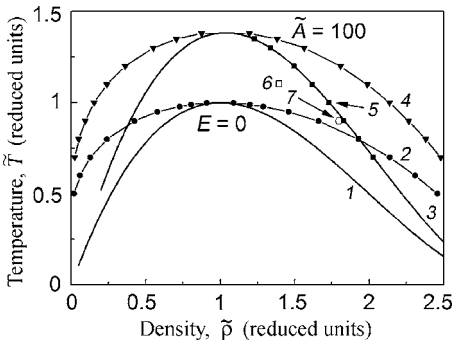

FIG. 1. Coexistence curves (2 and 4) and theoretical spinodals (17) (curves 1 and 3) for the van der Waals equation of state (15). Curves 1, 2, without electric field; curves 3, 4, in uniform electric field at $\tilde{A}=100$. Points 5, the spinodal obtained in hydrodynamic simulations by LBE method. Points 6 and 7 are the states initially above the critical point ($\tilde{\rho}_0=1.4$, $\tilde{T}=1.1$) and in the region of stability of liquid phase ($\tilde{\rho}_0=1.8$, $\tilde{T}=0.9$), respectively.

$$K_x = \frac{E_0^2}{12\pi}(\varepsilon-1)^2\frac{(\varepsilon+2)}{3\rho}. \tag{16}$$

In this case, the formula of the spinodal curve can be written in an explicit form

$$\tilde{T} = \frac{\tilde{\rho}(3-\tilde{\rho})^2}{4}\left(1+\tilde{A}\frac{(\alpha\rho_{cr})^2}{(1-\alpha\rho_{cr}\tilde{\rho})^3}\right). \tag{17}$$

The second term in parentheses corresponds to the shift of the critical point in temperature. Here $\tilde{A}=E_0^2/(8\pi p_{cr})$ is the dimensionless magnitude of electric field squared. For parameters corresponding to argon $T_{cr}=151$ K, $\rho_{cr}=531$ kg/m^3, $p_{cr}=4.86$ MPa, $\alpha\rho_{cr}=0.057$, the spinodal curves were calculated and are shown in Fig. 1 for $E=0$ (curve 1) and for $\tilde{A}=100$ (curve 3). The shift of the critical point in density $\Delta\tilde{\rho}_{cr}\sim 2(\alpha\rho_{cr})\Delta\tilde{T}$ is very small due to the small value of parameter $\alpha\rho_{cr}$.

The critical density and the value of $\alpha\rho_{cr}$ for rare gases increase with the increase of atomic mass, hence, the shift of a spinodal in an electric field becomes more pronounced. Thus, for xenon, the shift of the critical point in reduced temperature is approximately twice as much as the shift for argon.

IV. NUMERICAL SIMULATIONS

We performed the simulations of the evolution of homogeneous dielectric fluid that was initially at rest in uniform electric field (initial random density perturbations in the nodes of the lattice were of order of $(\Delta\rho/\rho_0\sim 10^{-6})$. The dynamics of continuous media was simulated using the lattice Boltzmann equation (LBE) method [9,10] modified to enable an arbitrary equation of state with possible phase transition [11,12] and to take into account an action of electric forces (1) on a fluid [13]. Periodic boundary conditions in x direction were used. The neutral wetting of electrodes was assumed (the wetting angle was set equal to $\pi/2$). The distribution of electric field was obtained by solving the equations

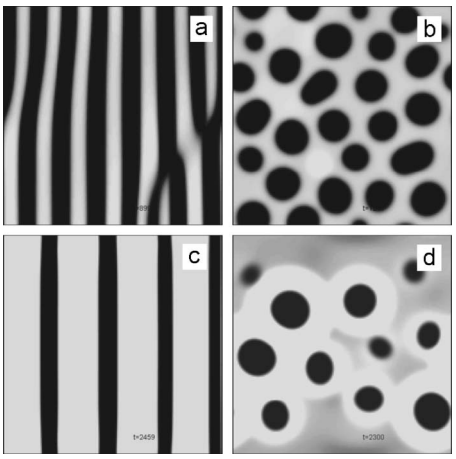

FIG. 2. Anisotropic stratification of fluid under action of uniform electric field. Stratification along the initially vertical field (a), (c). Development of instability in the plane x-y perpendicular to the field (b), (d). The lower density is shown by dark color. In (a) and (b) $\tilde{\rho}_0=1.4$, $\tilde{T}=1.1$ (state 6 in Fig. 1); in (c) and (d) $\tilde{\rho}_0=1.8$, $\tilde{T}=0.9$ (state 7 in Fig. 1). $\tilde{A}=100$.

$$\mathrm{div}(\varepsilon\,\nabla\,\varphi)=0, \quad \mathbf{E}=-\nabla\varphi \tag{18}$$

with corresponding boundary conditions $\varphi=0$ and $\varphi=E_0L_y$ at the lower and upper electrodes, correspondingly. The simulations were performed on a 150×150 lattice.

For nonpolar dielectric (4) the coexistence (binodal) curves were obtained in computer simulations both without electric field (Fig. 1, curve 2) and in an initially vertical uniform electric field (curve 4). The high-density part of the spinodal curve (points 5) was also calculated. Both the spinodal and coexistence curves are shifted upward the more the larger is the value of an electric field squared.

The diagram obtained shows that the anisotropic decay of liquid along the field is possible for matter lying initially in metastable and even in stable states (states 6 and 7 in Fig. 1) not far from the initial coexistence curve in sufficiently high electric field. The same effect will be observed for polar dielectrics.

Predicted stratification along a uniform electric field was indeed observed in computer simulations for matter that was initially both in a state above the critical point [Figs. 2(a) and 2(b)] and in a stable liquid state [Figs. 2(c) and 2(d)]. Simulations cover both the linear and nonlinear stages of instability. Instability arose in form of channels of approximately circular cross-section that generated compression waves during expansion [Fig. 2(d)]. This is a cooperative effect in the theory of nucleation [2]. The phase separation proceeded until the densities of phases reached the equilibrium values corresponding to the states on coexisting curve in presence of an electric field. In Fig. 2(a), branching and reconnection of vapor channels can be seen. This phenomenon is of stochastic nature, hence, it was observed not at every run for the size of computation area which was used. A frame that demon-

strates this phenomenon was specially chosen. For larger regions of uniform electric field, such reconnections will be more frequent.

V. DISCUSSION AND CONCLUSIONS

The main drawback of all previous works on the behavior of dielectric liquids in high electric fields (Refs. [14,15] and others) is that only the possibility of generation of spherical or ellipsoidal vapor bubbles was considered. The possibility of anisotropic instabilities already at the initial stage after voltage applied was not considered at all. Moreover, in Ref. [14], only the case of dielectric liquids with the linear density dependence of permittivity (3) was considered for which anisotropic instability is impossible for initially stable and metastable states as mentioned above. Hence, in Ref. [14], another mechanism of streamer growth was proposed that assumed the fast propagation of Griffith crack (similar to brittle material) in a liquid containing population of initial submicroscopic spherical holes (bubbles). The mechanism considered in Ref. [14] is very interesting but fundamentally different from the mechanism of anisotropic instability proposed here.

In the process of breakdown of liquid dielectrics in strong electric fields that can locally reach the values of $\sim 1-100$ MV/cm (for different liquids), the proposed anisotropic instability is possibly the key mechanism of inception of streamer structures, their ultrafast propagation in a form of thin filaments (the velocity can exceed 100 km/s [16]) oriented on average along the local electric field [Figs. 2(a) and 2(c)], and also of their branching during propagation. Since the electric strength of low-density fluid (vapor) is lower, electric breakdown can occur in some of the vapor channels produced by the anisotropic instability. When a filament becomes conductive, electric field in neighbor nonconductive channels decreases, and these channels disappear if their states leaves the region below the coexistence curve. Electric field ahead of the conductive filament is on the contrary enhanced. Hence, the instability conditions are fulfilled in a new region of dielectrics. This process can propagate very fast step by step in the space between electrodes.

So far, no experiments on the observation of the anisotropic instability were carried out, because the phenomenon was not known. Related experimental results are also rather scarce, nevertheless, several indirect confirmations of the model proposed can be listed: ultrafast propagation of streamer tips in form of filamentary channels with average velocity of order of 100 km/s (the "mode 4" in terms of the Grenoble group, LEMD CNRS [16]); stepwise propagation of streamer structure; pulsed character of light emission of streamer branches and of electric current in external circuit; loss of transparency of liquid in whole visible spectral range just before the inception of streamer channels [17]; appearance of a sheaf of thin radial filaments in highly nonuniform field (point-plane electrode geometry) [18].

Experimental observation of the proposed electrohydrodynamic instability of dielectric liquids would be complicated because the electric breakdown of a low-density phase in strong electric field can occur simultaneously with the development of instability. The easiest way to separate these effects is, possibly, to investigate the initial states with temperature slightly higher than the critical one. In this case, relatively low electric fields are sufficient for the anisotropic instability, hence, a process of breakdown could be avoided.

A related process was investigated in Ref. [19], namely a partial separation of binary mixture in a nonuniform electric field. The moving force of this separation is a minimization of electrostatic energy, hence, a component with higher ε is pulled into a region of stronger electric field (in itself, a well-known effect). However, possible anisotropic decay was not considered. Our theory can be extended to the case of binary mixtures, and an anisotropic demixing is possible even in a uniform field if $\left(\frac{\partial^2 \varepsilon}{\partial \phi^2}\right)_T \neq 0$ (ϕ is a concentration of one component). The consideration of binary mixtures is however more complicated and can be a subject of a future paper.

In summary, we discovered a anisotropic instability of a dielectric liquid in a uniform electric field and a segregation into a system of vapor filaments in a liquid parallel to the field. The shift of spinodal and binodal curves in an electric field was obtained from the linear stability analysis and in the computer simulations of electrohydrodynamics for a van der Waals fluid. Theoretical prediction of the decay of dielectric liquid into a system of filamentary vapor channels in a liquid was confirmed in computer simulations.

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