Hexagonal convection patterns and their evolutionary scenarios in electroconvection induced by a strong unipolar injection

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A regular hexagonal pattern of three-dimensional electroconvective flow induced by unipolar injection in dielectric liquids is numerically observed by solving the fully coupled governing equations using the lattice Boltzmann method. A small-amplitude perturbation in the form of a spatially periodic pattern of hexagonal cells is introduced initially. The transient development of convective cells that undergo a sequence of transitions agrees with the idea of flow seeking an optimal scale. Stable hexagonal convective cells and their subcritical bifurcation together with a hysteresis loop are clearly observed. In addition, the stability of the hexagonal flow pattern is analyzed in a wide range of relevant parameters, including the electric Rayleigh number $\mathcal{T}$, nondimensional mobility $M$, and wave number $k$. It is found that centrally downflowing hexagonal cells, which are characterized by the central region being empty of charge, are preferred in the system.

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I. INTRODUCTION

A hexagonal pattern is a well-known dissipative structure formulated in a nonlinear open system. The formation process of hexagonal convection cells exhibits dynamic self-organization manifested as an increase in coherence and a decrease in statistical entropy. Two typical hexagonal convection phenomena have been experimentally observed: Rayleigh-Bénard convection (RBC) and electrohydrodynamics (EHD) convection under the input of thermal energy and electric energy, respectively. In the field of EHD, four kinds of hexagonal patterns have been observed in different EHD convective systems [1–6], namely a charge injection hexagonal pattern [1,2], a corona-induced hexagonal pattern [3], a cell pattern in the formation of liquid crystals [4], and an electro-osmotic hexagonal pattern near charge-selective surfaces [5,6]. In this study, we are concerned with the electroconvection (EC) in a dielectric liquid layer lying between two parallel planar electrodes and subjected to unipolar injection of charges. This is one of the most classical problems studied in this field [7,8].

Electroconvective flow induced by unipolar charge injection into an insulating liquid is a fundamental problem in EHD [7,8]. Ions are generated by the electrochemical reaction at the interface between a liquid and an electrode, and then they enter into the bulk of the liquid due to the effect of an electric field. When the applied voltage is high enough, the Coulomb force gives rise to the development of an instability that puts the liquid into motion, in the form of two-dimensional electroconvective rolls or three-dimensional cellular patterns such as a hexagonal pattern. This type of flow motion plays the central role in several industry applications, such as heat-transfer enhancement

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[9] and flow control [10], EHD drag pump [11], EHD turbulent mixer [12], electrostatic precipitators [13], and atomization technology [14].

Similar hexagonal patterns have also been reported in other electrokinetic systems, for instance the electro-osmotic instability near a charge-selective surface (ion-exchange membranes, electrodes, or a system of micro- or nanochannels). This instability was theoretically predicted by Rubinstein and Zaltzman [5]. Then, direct numerical simulations are successfully conducted based on the fully coupled Nernst-Planck-Poisson-Stokes equations (NPPS) after long-term efforts [6,15–17]. Although the electro-osmotic instability is a related problem, its nature is different from the unipolar injection instability considered in this paper. Electro-osmotic instability appears in electrolytes in water solutions, which are orders of magnitude more conducting than the dielectric insulating liquids used in unipolar injection. As a result, the Debye length is much greater than the liquid layer thickness in the dielectric case, whereas it is much smaller in the water solution case. On the other hand, the diffusion term, although important in the detailed analysis of the injection process, is negligible in the liquid bulk for very insulating liquids, again a very different situation from the aqueous solution case. As a consequence, the NPPS model is not applicable in EC in very insulating liquids.

The EC problem (in the rest of this paper, EC refers in particular to the injection-induced electroconvection in dielectric liquids) [2] presents a clear analogy with classical RBC [18]. In both cases, flows are driven by body forces, i.e., the Coulomb force with EC and the thermal buoyancy force with RBC. Beyond a certain threshold, the fluid layer becomes unstable and breaks up into regular patterns of convective cells. However, the EC problem is specific and cannot be simply reduced to RBC. The differences may be ascribed to the nonlinear character of the electrical equations in EC as opposed to the linear energy equation in RBC [19]. Specifically, at a rest state the charges are transported via the drift and diffusion mechanisms, whereas the heat is transferred by diffusion alone. In addition, the bifurcation of the EC problem is of subcritical type featured by a linear stability criterion $T_{c}$, a nonlinear one $T_{f}$, and a hysteresis loop between them, while the RBC problem under the Boussinesq approximation exhibits supercritical bifurcations. As a classic case in nonlinear dynamics, systematically experimental, theoretical, and numerical studies have been devoted to RBC. By contrast, the EC has not been well understood, especially from a numerical standpoint. For example, the hexagonal cells observed in experiments have not been numerically reproduced, and some predictions by theoretical analysis [19] have not been proven.

Some basic features of EC, such as subcritical bifurcation and regular hexagonal cells, have been extensively investigated both in experimental studies and stability analysis [1,19–21]. However, few numerical results based on the fully coupled three-dimensional (3D) EHD model are available for these interesting phenomena. There are two main reasons for the lack of 3D simulations. One is the high computational cost due to the stiffness of the problem, which represents high gradients in charge-density distribution, and the other is the high complexity of nonlinear coupling between the charges, the electric field, and the flow field. Kourmatzis et al. [22] conducted a three-dimensional simulation of EC between two plates using commercial software. In that study, the authors mainly focused on the fully turbulent regime but ignored the EC phenomena close to the stability threshold, and the typical hexagonal pattern and subcritical bifurcation in EC were not mentioned.

Early research on the EC focused mainly on experimental studies and theoretical analysis. Previous experimental results are obtained with circular plane electrodes and with a very large aspect ratio $\Gamma = D/2d \geq 25$ [23] ($D$ is the diameter of the cylindrical cavity and $d$ is the distance between electrodes), in which regular hexagonal patterns in EC were clearly observed. The stability analysis simply assumed an infinite domain, and the flow is always fully developed and takes a self-similar form [21]. As for 3D numerical simulations, the large/infinite domain dramatically increases the computational cost. An effective skill is to use a finite computational domain with an appropriate size (usually, several times the critical wavelength) together with periodic boundary conditions applied to the wall-parallel direction, similar to the technique adopted in some research on RBC [24]. Since the domain’s size is limited, it naturally restricts the development of some modes of perturbation, and the flow may not tend to the fully developed state. Therefore, some artificial perturbations are subsequently introduced in the initial condition of simulations, and then different flow patterns arise.
These patterns can be viewed as good approximations of the fully developed flow pattern (e.g., the hexagonal cells) in the infinite domain. Therefore, the idea of a finite domain with periodic horizontal boundaries and special initial perturbations is adopted in our simulations.

Several conventional methods based on the partial differential equation (PDE) have been introduced to solve EC. These include the finite-difference method (FDM) [6,25], the finite-element method (FEM) [26], and the finite-volume method (FVM) [27,28]. In our recent work [29], we developed a unified lattice Boltzmann model (LBM) based on three lattice Boltzmann equations (LBEs) to calculate the fluid flow, the electric potential, and the charge-density distribution. Due to its kinetic particulate nature, in comparison with the conventional computational fluid dynamics (CFD) solvers, the LBM exhibits some attractive advantages, such as the simplicity of the calculation procedure, ease in programming and the boundary treatment of complex geometry, intrinsic parallelism, and so on [30–33]. In addition, as a transient solver, the LBM can naturally track the bifurcations and unsteady flow in the EC problem. Later, our LBM was extended to electrothermoconvection [34] and EC in complex geometries [35].

In this work, the regular hexagonal patterns and their subcritical bifurcation phenomena in 3D EC are numerically investigated. The main purpose of the present investigation is threefold: (i) to numerically validate the existence of a regular hexagonal pattern in EC, and to provide a detailed comparison between the features of hexagonal cells in RBC and EC; (ii) to exhibit the subcritical feature of the bifurcation in 3D EC, and to investigate the effect of mobility $M$ on the nonlinear stability criterion; and (iii) to determine the parameter scope $(T,k)$ in which stable hexagons with wave number $k = k_0$ can be developed from the initial perturbation.

II. MATHEMATICAL FORMULATION AND LATTICE BOLTZMANN MODEL

A. Physical problem and governing equations

Consider an insulating liquid layer of thickness $H$ enclosed between two planar electrodes of area $= L_x \times L_y$ normal to the $z$ axis, as shown in Fig. 1(a). A potential difference $\Delta \phi = \phi_0 - \phi_1$ is

![FIG. 1. (a) Sketch of the hexagonal cells in the electroconvection problem; (b) top view of the periodic hexagonal cells with dotted lines showing Voronoi polygon subdivision; (c) and (d) numerically obtained isosurfaces of charge-density distribution $q$ and vertical velocity component $w$ within a periodic unit.](image-url)
applied between the two electrodes. Free charges are first generated at the electrode-liquid interface and then enter the bulk of liquid under the effect of an electric field, which gives rise to a Coulomb force responsible for the fluid motion. The fluid layer in the marginal state will be tessellated into regular polygons. Among the polygonal cells, however, only the periodic hexagonal cells have been experimentally observed, which can be explained by the fact that for an isotropic liquid where no favored direction exists in the horizontal plane, a hexagonal pattern has a higher symmetry than other distributions, such as squares or rectangles patterns [19].

Figure 1(b) presents the top view of the regular hexagonal pattern, which has a honeycomb symmetry. The hexagonal cells have two typical features: (i) symmetry, i.e., invariance for rotation by 60° about the hexagonal center as expressed in Eq. (1a); and (ii) periodicity, i.e., the wavelength in the y direction is seen to be $\sqrt{3}$ times the wavelength in the x direction, as expressed in Eqs. (1b) and (2),

$$w(r,\theta) = w(r,\theta + 60^\circ),$$  \hspace{1cm} (1a)  
$$w(x + n\lambda_x, y + m\lambda_y) = w(x,y),$$  \hspace{1cm} (1b)

where $w$ is the vertical velocity component, $r$ and $\theta$ are axes in the cylindrical coordinate system (the origin of coordinates being the center of the maps), $m$ and $n$ are arbitrary integers, $\lambda_x$ and $\lambda_y$ are the wavelengths of the hexagonal pattern in the $x$ and $y$ directions, respectively, and $k$ is the corresponding wave number, defined as

$$\lambda_x = \sqrt{3}L, \quad \lambda_y = 3L, \quad k = 2\pi \sqrt{\frac{1}{\lambda_x^2} + \frac{1}{\lambda_y^2}} = \frac{4\pi}{3L},$$  \hspace{1cm} (2)

where $L$ is the side length of a hexagon. In addition, $k_c$ is defined to be the wave number of the most unstable mode, and the value of $k_c = 5.1$ is obtained by stability analysis for the injection strength $C = 10$ [36]. The corresponding critical wavelengths in the $x$ and $y$ directions are $\lambda_{cx} = 4\pi/\sqrt{3}k_c$ and $\lambda_{cy} = 4\pi/k_c$, respectively. In Figs. 1(c) and 1(d), a periodic unit structure of charge-density distribution $q$ and vertical velocity $w$ isosurfaces is plotted; it exhibits regular hexagonal features at the eventual steady state.

The complete set of macroscopic governing equations of EHD flows includes the mechanical equations and the electrical equations. Considering an incompressible, Newtonian, and linear isotropic fluid, mathematical formulations are given as [8,37]

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0,$$  \hspace{1cm} (3a)  
$$\frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = -\nabla \hat{p} + \nabla \cdot (\mu \nabla \mathbf{u}) + q \mathbf{E},$$  \hspace{1cm} (3b)  
$$\nabla^2 \phi = -q/\varepsilon,$$  \hspace{1cm} (3c)  
$$\mathbf{E} = -\nabla \phi,$$  \hspace{1cm} (3d)  
$$\frac{\partial q}{\partial t} + \nabla \cdot \mathbf{j} = 0, \quad \mathbf{j} = (K \mathbf{E} + \mathbf{u})q - D \nabla q,$$  \hspace{1cm} (3e)

where $\mathbf{u} = [u,v,w]$ is the fluid velocity; $\mathbf{E} = [E_x,E_y,E_z]$ is the electric field; and $\phi$, $q$, $\varepsilon$, $K$, $D$, $\rho$, $\mu$, and $\nu$ are the electric potential, charge density, permittivity, ion mobility, charge-diffusion coefficient, fluid density, dynamic viscosity, and kinematic viscosity, respectively. In addition, $\hat{p}$ denotes the generalized pressure including the hydrostatic pressure and the extra electrostrictive contribution. The system is essentially governed by the following four nondimensional parameters:

$$T = \frac{\varepsilon \Delta \phi}{\mu K}, \quad C = \frac{q_0 H^2}{\varepsilon \Delta \phi}, \quad M = \frac{1}{K} \left( \frac{\varepsilon}{\rho} \right)^{1/2}, \quad \alpha = \frac{D}{K \Delta \phi}.$$
where $q_0$ is the charge density at the injecting electrode; $T$ is the electric Rayleigh number, which is the ratio between the Coulomb force and the viscous force; $C$ represents the injection strength; $M$ is the nondimensional mobility parameter, which depends only on the properties of the fluid and the ion; and $\alpha$ is the nondimensional charge-diffusion number, with a typical value in the range between $10^{-3}$ and $10^{-4}$ [2].

Three LBEs are formulized, corresponding to the following macroscopic equations: the Navier-Stokes equations [Eqs. (3a) and (3b)], Poisson’s equation [Eq. (3c)], and the charge-conservation equation [Eq. (3e)]:

$$f_j(x + c_j \Delta t, t + \Delta t) - f_j(x, t) = -\frac{1}{\tau_v} [f_j(x, t) - f_j^{\text{eq}}(x, t)] + \Delta t \times F_j,$$

$$g_j(x + c_j \Delta t, t + \Delta t) - g_j(x, t) = -\frac{1}{\tau_\phi} [g_j(x, t) - g_j^{\text{eq}}(x, t)] + \Delta t \times S_j,$$

$$h_j(x + c_j \Delta t, t + \Delta t) - h_j(x, t) = -\frac{1}{\tau_q} [h_j(x, t) - h_j^{\text{eq}}(x, t)],$$

where $f_j$, $g_j$, and $h_j$ are the distribution functions for the flow field, the electric potential, and the charge density, respectively. The single-relaxation-time (SRT) model [38] is adopted for all LBEs because of its simplicity, and the relaxation times in Eqs. (4a)–(4c) are given as

$$\tau_v = \frac{3v}{c^2 \Delta t} + \frac{1}{2}, \quad \tau_\phi = \frac{3\gamma}{c^2 \Delta t} + \frac{1}{2}, \quad \tau_q = \frac{3D}{c^2 \Delta t} + \frac{1}{2},$$

where the constant $\gamma$ is used to control the evolution speed of the electric potential field, and $\gamma = 0.3$ is adopted in our simulation. In Eqs. (4a)–(4c), the equilibrium distributions for the flow field, the electric potential, and the charge-density distribution can be computed using the macroscopic quantities, given as

$$f_j^{\text{eq}} = \rho \omega_j \left(1 + \frac{c_j \cdot u}{c_s^2} + \frac{(c_j \cdot u)^2}{2c_s^4} - \frac{u^2}{2c_s^4}\right),$$

$$g_j^{\text{eq}}(x, t) = \omega_j \phi,$$

$$h_j^{\text{eq}}(x, t) = q \omega_j \left(1 + \frac{c_j(KE + u)}{c_s^2} + \frac{(c_j(KE + u))^2}{2c_s^4} - \frac{c_j^2(KE + u)^2}{2c_s^4}\right),$$

where $c_s = c/\sqrt{3}$ in the D3Q19 lattice is the lattice sound speed. The split-forcing scheme [39] is used to accurately account for the body force. In this approach, the force term in Eq. (4a) and the source term in Eq. (4b) are formulated as

$$F_j = \omega_j \left(1 - \frac{1}{2\tau_v}\right) \frac{c_j \cdot (qE)}{c_s^2},$$

$$S_j = \omega_j \gamma q / \varepsilon.$$
B. Initial and boundary conditions

After initializing the distributions of different fields, three-dimensional EC flow tends to manifest itself in the form of polygonal cells after a certain amount of time. Based on the stability analysis reported in [19], perfect hexagonally shaped convection is preferred in an infinite domain. However, in 3D numerical simulations, a finite computational domain is always adopted, and two or more attractors corresponding to different flow forms (irregular hexagons, squares, rolls) can coexist in general under given parameter values and initial states. To reproduce the regular hexagonal pattern, a special initialization technique is adopted in our simulations. Specifically, at initial time, a spatially periodic small perturbation described by the Chandrasekhar function [40] with wave number $k_0$ is introduced to vertical velocity $w$, expressed as

$$w = \frac{1}{3} W(z) \left[ \cos k_0 y + 2 \cos \left( \frac{\sqrt{3} k_0 x}{2} \right) \cos \left( \frac{k_0 y}{2} \right) \right],$$

where $W$ is the amplitude of the perturbation.

To close the system, boundary conditions for the velocity field, the electrical potential, and the charge density need to be added. The periodic boundary conditions in the wall-parallel conditions are assumed in our problem. For the velocity field, the no-slip and no-penetration conditions are assumed for two solid electrodes,

$$u = 0 \quad \text{at} \quad z = 0, H.$$

For the electric potential, since a DC potential difference is applied, the Dirichlet conditions are required for both electrodes, given as

$$\phi = \phi_0 \quad \text{at} \quad z = 0, \quad \phi = \phi_1 \quad \text{at} \quad z = H.$$

The boundary for charge density is given with reference to the work of Pérez [41]. In the injection electrode, the assumption of autonomous and homogeneous injection is adopted, while a completely open electrode is assumed in the collection electrode,

$$q = q_0 \quad \text{at} \quad z = 0; \quad \partial q / \partial z = 0 \quad \text{at} \quad z = d.$$

The mesoscopic boundary conditions can be determined with the help of macroscopic boundary conditions. In the present work, the second-order nonequilibrium extrapolation scheme (NEES) proposed by Guo et al. [42] is employed for all LBEs at the solid electrodes. The advantage of the NEES for the coupled multifield problems lies in the fact that the same functional module can be applied to all independent variables. In the implementation of NEES, the unknown distribution function at $x_b$ is divided into an equilibrium component and a nonequilibrium one [43],

$$R^\text{eq}_j(x_b, t) = R^\text{eq}_j(x_b, t) + R^\text{neq}_j(x_b, t),$$

$$R^\text{neq}_j(x_b, t) = R_j(x_f, t) - R^\text{eq}_j(x_f, t),$$

in which the equilibrium part $R^\text{eq}_j(x_b, t)$ in the boundaries can be computed with the help of macroscopic boundary conditions, while the nonequilibrium component $R^\text{neq}_j(x_b, t)$ can be obtained from neighboring fluid nodes $x_f$ as given in Eq. (10b).

For the lateral boundaries, the periodic boundary condition, which assumes that a particle leaves the computational domain from one side and reenters the domain from the opposite side [44], is employed for all fields.

A complete numerical solution procedure by LBM is given as follows: At $t = 0$, a weak velocity perturbation expressed by Eq. (9) is introduced to vertical velocity $w$. Then, a global iteration is conducted in the following order: (i) Do the collision and streaming steps of Eq. (4a) and calculate the density $\rho$ and velocity $\mathbf{u}$ field by Eq. (8); (ii) solve the LBE for the electric potential [Eq. (4b)]
and calculate $\phi$ and the electric field $E$ by Eq. (8); (iii) solve the LBE for the charge density [Eq. (4c)] and calculate $q$ by Eq. (8); (iv) go back to step (i) unless the convergence criterion is satisfied.

Numerical results are presented by isosurfaces and streamlines, as well as the maximum fluid velocity ($V_{\text{max}}$) and the electric Nusselt numbers ($Ne$), given as

$$V_{\text{max}} = \max(\sqrt{u^2 + v^2 + w^2}), \quad Ne = I/I_0,$$

where $I$ is the electric current and $I_0$ is the electric current without flow motion [45]. The electric current can be computed either by integrating the current density on the entire emitter or by collecting electrodes.

### III. RESULTS AND DISCUSSIONS

#### A. Code and results validations

In our previous two-dimensional works [29,34], the unified lattice Boltzmann model for EC problems was confirmed. Here we further validate our LBM code for 3D hexagonal electroconvective flow. Results validations are conducted from three aspects: (i) a hydrostatic solution under weak, moderate, and strong injections; (ii) a comparison of the electric Nusselt number $Ne$ at the nonlinear stability criterion ($T = T_f$) with experimental results; (iii) a comparison of the linear and nonlinear stability criteria ($T_c$ and $T_f$) with stability analysis data; and (iv) validation of the critical wavelength $k_c$.

By setting $T$ smaller than the linear stability criterion $T_c$, or directly turning off the module for fluid velocity and then starting the computation with zero initial distributions, we can obtain the hydrostatic solution where fluid remains a motionless state. Figure 2 compares the numerical and analytical profiles of charge density at $C = 10$, 1, and 0.1. A fairly good agreement is readily seen, with the maximum difference between the analytical solution [36] and the numerical one being less than 0.86%.

The value of $Ne$ at $T = T_f$ represents the minimum dimensionless electric current of a hexagonal pattern. In our LBM simulations, the $Ne$ at $T = T_f$ is determined in the following way: restarting the computation from a steady convection obtained with $T$ slightly higher than $T_c$, then gradually decreasing $T$ by a small amount until a critical value (i.e., $T_f$), and recording the $Ne$ value at which the system suddenly jumps from a convective state of finite-amplitude strength to the rest state.

![FIG. 2. Comparison of charge-density distributions between numerical and analytical solutions at the hydrostatic state under different injection strengths.](image-url)
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As shown in Fig. 3(a), the LBM-obtained Ne corresponding to $T_f$ is 1.089, which is close to the experimental result $Ne = 1.060$ and the theoretical result $Ne = 1.071$.

The subcritical bifurcation is a representative characteristic of EC, featured by a linear stability criterion $T_c$, a nonlinear stability criterion $T_f$, and a hysteresis loop between these two criteria [1,21]. A brief explanation about these phenomena is as follows: the nonlinear coupling between the charge, the electric field, and liquid motion results in a sudden jump between the rest state and the motion state of the liquid, with a maximum velocity that exceeds the ion drift velocity [see Fig. 3(a)]. Once the motion takes place for $T > T_c$, decreasing the value of $T$ back to $T_c$ does not result in recovering the rest state. The motion is sustained until we reach a second criterion (nonlinear or finite-amplitude criterion) $T_f$, and the hysteresis loop links the linear and nonlinear criteria, which is a characteristic of the subcritical bifurcation.

As we have seen, previous numerical works are limited to the reproduction of bifurcations about two-dimensional roll flow in EC [46]. Therefore, here we extend the LBM results to a three-dimensional hexagonal pattern. Figure 3(a) shows the subcritical bifurcation of the hexagonal pattern expressed by the electric Nusselt number Ne versus $T$. The calculation procedure of the two critical values has been well documented in Refs. [29,47]. In 3D simulations, our numerical predictions of $T_c$ and $T_f$ are LBM 162.6 and 111.1, respectively. These two values are very close to those obtained by stability analysis ($T_c = 164.1$ and $T_f = 111.7$) [1,21]. The consistency between numerical results and stability analysis quantitatively justifies the accuracy of our 3D lattice Boltzmann method.

As mentioned in the theoretical works in Refs. [19,36], the critical wavelength $k_c$ corresponding to the most unstable mode at the linear stability criterion $T_c$ can be minimized. The values of $k_c$ of the hexagonal pattern predicted by stability analysis [19,36] and experimental observation [19] are $k_c(\text{theory}) = 5.1$ and $k_c(\text{exp}) \approx 5$, respectively. Here we conduct a series of numerical tests to confirm these values by changing the wave number $k$ and calculating the corresponding $T_c$. In numerical simulations, changing $k$ leads to a variation of the wavelength in $\lambda_x$ and $\lambda_y$ [see Eq. (2)], and then it affects the dimensionless computation domain size ($L_x \times L_y \times L_z = \lambda_x \times \lambda_y \times 1$). As shown in Fig. 3(b), seven different values of $k = 4, 4.5, 5.1, 5.5, 6, 7,$ and 8 are considered, and the calculated $T_c$ values are 172.93, 164.39, 162.58, 163.52, 168.79, 180.31, and 184.73, respectively. It is seen that $T_c$ indeed reaches its minimum at $k = 5.1$ in the range of the numerical test, which is consistent with the theoretical and experimental results [19,36].

B. Results and discussions

In this subsection, systematic simulation results for the 3D EC are presented and discussed. Figure 4 shows the transient development of hexagonal cells and a steady-state flow field and charge-
FIG. 4. (a) Time histories of the peak velocity for the case of \( T = 200 \). Steady-state results of hexagonal patterns for (b) velocity isosurfaces at \( w = 1 \) with streamlines, and (c) charge-density isosurfaces at \( q = 0.2 \) and 0.03; the computational domain of two times of the critical wavelength and the initial perturbation of the wave number is \( k_0 = k_c \).

density distributions. The domain size is set to be twice the critical wavelength \( 0 \leq x \leq 2\lambda_{cx} \) and \( 0 \leq y \leq 2\lambda_{cy} \). As shown in Fig. 4(a), the evolution of peak velocity \( V_{\text{max}} \) experiences two different stages after the initial disturbance. The first one is the exponential growth stage, where \( V_{\text{max}} \) follows a law \( f = f_0 e^{\sigma t} \) with a growth rate of \( \sigma \) [see the inset in Fig. 4(a)], which can be used to determine the linear stability criterion [47]. In detail, different \( \sigma \) can be extracted from the time evolution curves of different \( T \), and then extrapolated to obtain the linear stability criterion \( T_c \) corresponding to the zero-growth rate \( \sigma = 0 \). For example, in this case the values of \( \sigma \) corresponding to \( T_1 = 175 \) and \( T_2 = 185 \) are \( \sigma_1 = 0.89 \) and \( \sigma_2 = 1.61 \), respectively, and then \( T_c \) can be linearly approximated to be \( T_c = T_1 - \sigma_1(T_2 - T_1)/\sigma_1 = 162.58 \). The second stage in Fig. 4(a) is the flow transition stage in which the flow field adjusts itself to the regular hexagonal pattern, accompanied by the formation of a charge void region.

In Fig. 4(b), vertical velocity isosurfaces and streamlines at steady state are plotted. The flow field exhibits the features of basic hexagonal cells: (i) invariance for rotation by \( 60^\circ \) about the hexagonal center, and (ii) the wavelength of the periodicity in the \( y \) direction is seen to be \( \sqrt{3} \) times the wavelength in the \( x \) direction. In Fig. 4(c), charge-density isosurfaces (\( q = 0.2 \) and 0.02) are presented: the isosurfaces at \( q = 0.2 \) show the hexagonal cells, while the isosurfaces at \( q = 0.02 \) exhibit a “bun-shape” cells with the inner region being empty of charge. The formulation of this kind
FIG. 5. Comparison of RBC and EC: (a) upflowing in RBC at $Ra = 3000$, (b) downflowing in RBC at $Ra = 3000$, and (c) downflowing in EC at $T = 200$. In each subfigure, isosurfaces of vertical velocity with streamlines (top) and isosurfaces of temperature or charge density (bottom) are provided.

of charge void region can be explained by the competition between the migration and convection mechanisms for charge transport [19].

The analogy with nonlinear RBC may offer some guidance in the study of EC problems. As reported in several significant works by Busse [48] and Bodenschatz [49], a hexagonal pattern in RBC is preferred in a nonsymmetric layer; depending on the sign of the asymmetry, either the *l-hexagons* (fluid ascends in the center of the cells) or the opposite *g-hexagons* (fluid descends in the center of the cells) are optimal. In experimental and theoretical studies [48,50], a hexagonal pattern can be caused by the asymmetry of the fluid properties, the non-Boussinesq effect, or the asymmetry of the boundary conditions. However, in numerical studies, a hexagonal flow pattern can be obtained in a relatively simple way, as mentioned by Getling [24], i.e., a spatially asymmetric initial perturbation with a finite computational domain and periodic boundary conditions. Based on this idea, an LBM code for RBC is also programed and prevalidated for the purpose of comparison with EC phenomena. For a more detailed discussion about RBC, please refer to Ref. [18].

In both RBC and EC systems, flow can manifest itself in the form of polygonal cells when the governing parameter $T$ or $Ra$ exceeds the linear stability criterion ($T_c$ or $Ra_c$). According to the theoretical results [36,47], the critical governing parameters and critical wave numbers of EC with strong injection $C = 10$ and RBC are $T_c = 164.1$, $k_{c1} = 5.113$ and $Ra_c = 1708.7$, $k_{c2} = 3.117$, respectively. The domain size of one periodic unit $0 \leq x \leq \lambda_{cx} = 4\pi/\sqrt{3}k_c$ and $0 \leq x \leq \lambda_{cy} = 4\pi/k_c$ is adopted in both simulations. As shown in Fig. 5, there are two significant differences between the hexagonal convection of EC and RBC. One is the type of hexagon; the RBC shows *l-hexagons* or the opposite *g-hexagons* corresponding to upward or downward initial perturbations, as presented in Figs. 5(a) and 5(b), respectively. However, the EC is always of *g-hexagons* type (fluid descends
FIG. 6. (a) The subcritical bifurcation diagram of a hexagonal pattern of 3D electroconvection, and (b) the electric Nusselt number $N_e$ as a function of $T/T_c$ with different mobility $M$; the analytical curves are taken from Ref. [19]. (b) Different flow patterns in the $T$-$k_0$ plane from present numerical simulations.

in the center of the cells) since the viscous dissipation for a centrally downflowing hexagonal cell is less than that of an upflowing one [19]. Another difference is that the EC charge-density distribution shows high gradients in the peripheral region of hexagonal cells due to the convection-dominant feature of the charge-transport equation [see Fig. 5(c)]. In contrast, temperature distribution in RBC is much smoother.

Figure 6(a) shows the variation of the electric Nusselt number $N_e$ as a function of $T/T_c$ under different mobility numbers $M = 10$ and 20; the analytical curves from Ref. [19] based on 1-mode, 2-mode, and 4-mode are also provided. Similar to the situation of two-dimensional roll flow, the increase of $M$ from 10 to 20 slightly increases the $T_f$ and $N_e$ [46]. Moreover, with the increase of the order of expansions (from 1-mode to 4-mode), the analytical curves are closer to our LBM results, which is consistent with the fact that the real flow field corresponds to $\infty$-mode.

The existence and stability of the hexagonal pattern in the EC problem depends not only on electric Rayleigh number $T$ and mobility $M$, but also on the wave number $k$ of the initially specified perturbation. Therefore, a $(k,T)$ diagram is drawn in Fig. 6(b) for different $k_0$ and $T$ ($3 \leq k \leq 8$, $160 \leq T \leq 300$). The computational domain is set to be one periodic unit ($L_x \times L_y \times L_z = \lambda_x \times \lambda_y \times 1$). We first consider the values of $k_0$ close to the critical wave number $k_c$ ($= 5.14$), i.e., $k_0 = 5$ or 6, at which point stable hexagons with a wave number $k = k_0$ can develop from initial perturbation when $T$ exceeds $T_c$; see the 1-cell hexagon in Fig. 7(a). When decreasing the wave number of the initial disturbance to $k_0 = 3$, as shown in Fig. 7(b), the EC system will break into a stable $\sqrt{3}$-cell hexagonal flow with an actual wave number $k = \sqrt{3}k_0 = 5.196$ close to $k_c$, which meets the requirement of the system seeking its optimal mode. For $k_0 = 4$, a quasiperiodic unsteady flow is observed due to the competition of several modes. As presented in Figs. 7(c), 7(c'), and 7(c''), the transition regularly occurs between three quasistable states, i.e., a butterfly pattern, an inverse butterfly pattern, and a rectangles pattern, respectively.

With the increase of $k_0$ ($=7$ or 8), the restriction on the wavelengths of the perturbations would prevent the formation of hexagonal cells; in that situation, patterns with a smaller wavelength could be activated, such as a rectangles pattern or a rolls pattern. At $k_0 = 7$, the flow maintains a motionless state when $T < T_c$ (with $T_c$ calculated to be 180.3 in this case). When increasing $T$ exceeds its linear critical point $T_c$, the 1-rectangle pattern can be obtained as illustrated in Fig. 7(d). Moreover, a transition from the 1-cell rectangle pattern to the 2-cells rectangles pattern can be observed in Fig. 7(d') with $T = 300$. This phenomenon is similar to the flow breakdown in two-dimensional roll convective flow as reported in Refs. [28,46,51]. Finally, at $k_0 = 8$, flow field exhibits a quasiroll flow pattern as shown in Fig. 7(e), which can be viewed as a combination of roll and rectangle patterns.
FIG. 7. Types of final-state structures in 3D electroconvection for different $k$: (a) a one-cell hexagon at $k_0 = 5$ or 6, (b) a $\sqrt{3}$-cell hexagon at $k_0 = 3$, (c),(c') three quasi-stable states at $k_0 = 4$, (d) a one-cell rectangle pattern at $k_0 = 7$ breaking into (d') a two-cell rectangle pattern, and (e) a quasi-rolls flow pattern at $k_0 = 8$.

As plotted in the phase diagram Fig. 6(b), the star symbol represents the unsteady flow. Two types of unsteady flow can be obtained in numerical simulations, namely the quasiperiodic pattern and the chaotic pattern. The first scenario happens at wave number $k = 5$ when $T_c < T < 300$ due to the competition of several flow modes, while the chaotic patterns happen at a relatively higher control parameter $T \geq 300$, explained by the transition of the flow regime from laminar flow to chaotic flow. To fulfill the results discussion, a case at relatively high $T$ is considered, and other parameters are kept the same as those of Fig. 6.

Figure 8(a) plots the evolution in time of the maximum velocity magnitude for $T = 500$. The evolution of peak velocity $V_{\text{max}}$ also experiences two different stages after the initial disturbance. The first one is the exponential growth stage, which is the same as the situation of the smaller $T$ cases [see Fig. 4(a)]. However, the second stage in Fig. 8(a) is quite different, that is, the hexagonal pattern occurs [see point A in Fig. 8(a)] and then quickly becomes strongly unsteady and appears to be chaotic. The chaotic nature of the flow motion is confirmed by the spectral analysis shown in Fig. 8(b), which shows a broadband spectrum with an exponential decay. In other words, the flow passes from a motionless state to hexagonal convective flow and finally to turbulence once the system loses its linear stability, which can be explained by the fact that the system with high $T$ is essentially in the inertial state of motion. The charge-density distribution at three different nondimensional times $t^* = 0.72$, 3.8, and 5.6 [marked as A, B, and C in Fig. 8(a)] is presented in Figs. 8(c)–8(e), respectively. It is seen that the flow field shows a regular hexagonal pattern in an earlier stage; see the first map in Fig. 8(c). However, the flow will never reach a steady state with dramatic fluctuations around some average values.
IV. CONCLUSIONS

In this paper, the finite-amplitude EC in a layer of insulating liquid subjected to unipolar charge injection is numerically investigated by solving fully coupled governing equations using the lattice Boltzmann method. Three-dimensional hexagonal flow patterns as well as their subcritical bifurcations are numerically analyzed.

First, the 3D LBM code is validated by four carefully chosen cases, namely the hydrostatic solution, the value of electric Nusselt number Ne at \( T_f \), linear and nonlinear stability criteria (\( T_c \) and \( T_f \)), and critical wavelength \( k_c \). All results obtained with the LBM are found to be highly consistent with stability analysis results or experimental data. For example, numerically predicted linear stability and nonlinear stability criteria are 162.58 and 111.13, respectively, close to the values of stability analysis (164.1 and 111.7).

Then, the well-known honeycomb motion in 3D EC is successfully reproduced by numerical simulations. In the transient evolution of EC after an initial perturbation is analyzed, the flow field is found to adjust itself into a regular hexagonal pattern after several transient stages. The symmetry and periodicity in hexagonal cells is clearly observed; in particular, a charge-density distribution exhibits a buns-shaped charge void region. In addition, a comparison between the hexagonal pattern in EC and the well-known Rayleigh-Benard convection is conducted. Two remarkable differences are noted: (i) there are centrally upward and downward flows in RBC while merely downward flows in EC due to the ion drift mechanism, and (ii) there are high gradients in the charge-density distribution of EC due to the convection-dominant feature of the charge transport equation.
Finally, a \((k,T)\) diagram is drawn to study the stability of a hexagonal flow pattern when varying the wave number \(k\) and the electric Rayleigh number \(T\). It is found that a one-cell hexagon can be developed at \(k = 5\) or 6 (close to the critical wave number \(k = 5.14\)) when \(T\) exceeds \(T_c\). In addition, a stable \(\sqrt{3}\)-cell hexagon, rectangles, and quasiroll patterns can be observed at \(k = 3, 7,\) and 8, respectively. For \(k = 4\), a quasiperiodic unsteady flow is observed due to the competition of several quasistable states. In addition, at a relatively higher control parameter \(T = 500\), the flow passes from a motionless state to hexagonal convective flow and finally to turbulence once the system loses its linear stability.

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HEXAGONAL CONVECTION PATTERNS AND THEIR …


