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Influence of liquid bridge volume on the floating zone convection

W. R. Hu and Z. M. Tang*

National Microgravity Laboratory/CAS, Institute of Mechanics, Chinese Academy of Sciences, Beijing 100080, China

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The liquid bridge volume is a critical geometrical parameter in addition to the aspect ratio for onset of oscillation in the floating zone convection. The oscillatory features are generally divided into two characteristic regions: slender liquid bridge region and fat liquid bridge region. The oscillatory modes in two regions are discussed in the present paper.

Dedicated to Professor K. W. Benz on the occasion of his 65th birthday.

1 Introduction

Floating zone technique is an advanced method for high quality crystal growth and requires the studies on the convection in the liquid bridge. The gravity on the ground limits diameter of the grown crystal by the floating zone technique, however, crystals with larger diameter can be performed easily in microgravity environment [1]. The temperature non-uniform on the free surface of a liquid bridge induces the thermocapillary convection driven by the surface tension gradient. The thermocapillary convection is important for the processing of floating zone technique even on the ground condition, and dominates the process in the microgravity environment. Main research interesting related to the materials processing concentrates in the transition from a steady state to an oscillatory state of the convection, because the onset of oscillatory convection depends closely on appearance of striations in the solidified crystal.

The model of floating half zone was suggested for mechanism studies on the thermocapillary convection since the late of 1970's [2,3]. The schematic of a floating half zone is shown in Fig. 1. Studies have been performed by the methods of experiment, numerical simulation, and instability analysis (see for example [4]). The space experiment in the microgravity environment is of course beneficial most to understanding mechanism on floating zone convection, however, the opportunity is limited. Based on the Bond number simulation, the space processes, where gravity effect is negligible in comparison with thermocapillary effect, are simulated by the ground processes with small typical scale. The thermocapillary effect may be relatively important than the gravity effect in this case.

The process of floating zone convection is very complex, and many parameters are critical for the onset of oscillatory thermocapillary convection, such as the Marangoni number $Ma = |\sigma'_T| l\Delta T/\rho\nu\kappa$, Prandtl number $Pr = \nu/\kappa$, geometrical aspect ratio $A = l/d$, the parameter describing the heat process, and others. Where ρ, σ, ν and κ are respectively the liquid density, surface tension, kinematic viscous and thermal diffusion coefficients; l

* Corresponding author: e-mail: tang@nml.imech.ac.cn

and d are the height and rod diameter of the liquid bridge; and ΔT is the applied temperature difference. Most liquid bridges on the floating zone convection are analyzed as a cylindrical liquid bridge, and the typical geometrical parameter is aspect ratio A. It is noted that another geometrical parameter, the liquid bridge volume, should also be an additional critical parameter of floating zone convection on the ground, where the configuration of a liquid bridge likes a calabash in shape, and in microgravity environment where the configuration of liquid bridge deviates usually from the special case of a cylinder. The liquid bridge volume V is a sensitive critical parameter for onset of oscillation, and the marginal curve in case of a fixed aspect ratio A has two branches: slender liquid bridge and fat liquid bridge as shown in Fig. 2, where V_o is the typical volume of a cylinder $V_o = \pi l d^2/4$ [5,6]. The critical Marangoni number can change one or two times depending on liquid bridge volume in cases of a fixed aspect ratio A, and there is a small region between two branches with peak distribution of critical Marangoni number, related to a more stable region [7-9, 10-13].

Recently, two bifurcation transitions for onset of oscillatory thermocapillary convection have been studied. There is a bifurcation transition from steady and axial symmetric to steady and axial asymmetric state before the onset of oscillation. The studies was firstly given for a floating half zone with small Prandtl number Pr = 0.01 [14], and then for ones with large Prandtl number [15,16]. Two bifurcation transitions appear in relative fat liquid bridge $V/V_o > (V/V_o)_c$ for a fixed aspect ratio [17] and in relative small aspect ratio $A < A_c$ for a fixed liquid bridge volume [18] in cases of large Prandtl number. Two bifurcation transitions imply the existence of two critical Marangoni numbers. The first critical Marangoni number relates to the first bifurcation transition from steady and axial symmetric to steady and axial asymmetric convection, and the second critical Marangoni number relates to the second bifurcation transition from steady and axial asymmetric to oscillatory convection. Two bifurcation transition means that the onset of oscillatory thermocapillatory convection cannot be explained by mechanism of the hydrothermal instability.

In the present paper, the modes of oscillatory thermocapillary convection depending on the liquid bridge volume are discussed by the numerical simulation method for large Prandtl number fluid in detail. The oscillatory mode for slender liquid bridge is different from that for fat liquid bridge, and the modes for liquid bridge of large Prandtl number fluid is also different from that of small Prandtl number fluid.

2 The physical model

A floating half zone between parallel and co-axial two copper rods as shown in Fig. 1 is analyzed by the numerical simulation method. The temperature T_2' at the upper rod is higher than temperature T_1' at the lower rod, and the temperature difference is $\Delta T' = T_2' - T_1'$. The cylindrical coordinate system is used. The free surface shape of the liquid bridge is determined by the static condition of an isothermal liquid bridge, and is not changed during the computational process.

Dimensionless quantities are introduced as follows:

$$r = \frac{r'}{\ell},\quad z = \frac{z'}{\ell},\quad u = \frac{u'}{U_r},\quad v = \frac{v'}{U_r},\quad w = \frac{w'}{U_r},\quad t = \frac{t'}{\ell / U_r},$$

$$T = \frac{T'}{(T_* - T_1')},\quad Rs = \frac{U_r \ell}{\nu},\quad Ma = \frac{U_r \ell}{\kappa},\quad G_r = \frac{g\beta(T_* - T_1')\ell^3}{\nu^2}, \tag{2.1}$$

where superscript prime denotes the dimensional quantities, for examples, (u', v', w') is the dimensional velocity vector. A constant temperature $T_* = T_1' + 100^oC$ is adopted as reference temperature in the present paper. The typical velocity $U_r = |\partial\sigma/\partial T'|(T_* - T_1')/\rho\nu$ is defined by the condition of tangential momentum conservation on the free surface. The R_s, G_r, and M_a are respectively the Reynolds number, the Grashof number and the Marangoni number, and they are connected by the relationship Ma = Rs Pr, where $Pr = \nu / \kappa$ is the Prandtl number.

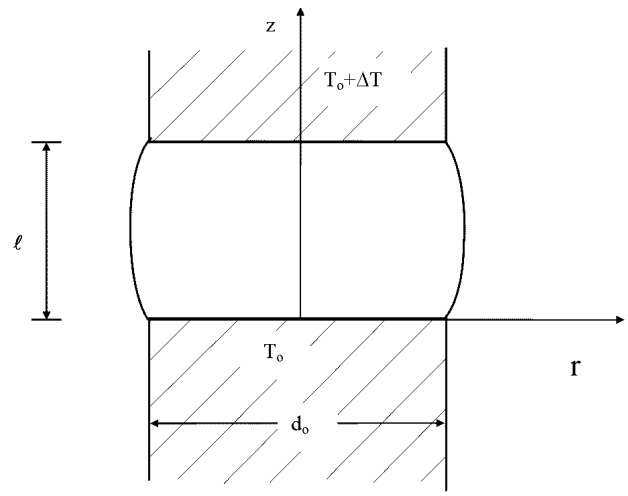


Fig. 1 Schematic of a floating half zone.

We introduce dimensionless vectors of the stream function y and the vorticity w

$$\nabla \times y = V \tag{2.2}$$

$$\nabla \times V = w \tag{2.3}$$

Unsteady and three-dimensional Navier-Stokes equations and energy equation can give the non-dimensionl equations of y, w and T as follows [12].

$$\nabla \times \nabla \times y = w \tag{2.4}$$

$$\frac{\partial w}{\partial t} + V \cdot \nabla w - w \cdot \nabla V = \frac{1}{\mathrm{Rs}}(\nabla^2 w + \frac{\ell^2}{??\mathrm{Ur}} \nabla \times \boldsymbol{F}), \tag{2.5}$$

$$\frac{\partial \mathrm{T}}{\partial t} + V \cdot \nabla \mathrm{T} = \frac{1}{\mathrm{Ma}} \nabla^2 \mathrm{T}. \tag{2.6}$$

F is body force, such as gravity, and is omitted in the present paper for microgravity environment.

The boundary conditions are z = 0 and z = 1:

$$\psi_r = \psi_\phi = 0, \qquad \frac{\partial \psi_z}{\partial z} = 0, \tag{2.7}$$

$$\omega_r = -\frac{\partial v}{\partial z}, \qquad \omega_\theta = \frac{\partial u}{\partial z}, \qquad \omega_z = 0, \tag{2.8}$$

at both the upper and lower boundaries;

$$\begin{cases} T(r,\theta,0,t) = 0, \\ T(r,\theta,1,t) = f(t); \end{cases} \tag{2.9}$$

where dimensional parameter α_T is a constant heating rate, and function $f(t) = \dfrac{\alpha_T\, t\, \ell}{(T_* - T_1^{'})U_r}$; $r = f_1(z)$:

$$\Psi_s = \Psi_f = 0, \quad \nabla \cdot \Psi = 0, \tag{2.10}$$

$$\omega_\phi = \left\{ \frac{(1+f'^2)}{(1-f'^2)} \frac{\partial T}{\partial S} + \frac{2f'}{(1-f'^2)} \left(\frac{\partial u}{\partial r} - \frac{\partial w}{\partial z} \right) + 2\frac{\partial u}{\partial z} \right\}, \tag{2.11}$$

$$\omega_z = \frac{(1+f'^2)^{1/2}}{r} \frac{\partial T}{\partial \phi} + 2\frac{\partial v}{\partial r} - f'(\omega_r + 2\frac{\partial v}{\partial z}), \tag{2.12}$$

$$\omega_r = \frac{1}{r}\frac{\partial w}{\partial \phi} - \frac{\partial v}{\partial z}, \tag{2.13}$$

$$\frac{\partial T}{\partial n} = 0, \tag{2.14}$$

at the free surface of the liquid bridge, which is described by r = f (z) as a rotational surface and symmetric to the z axis.

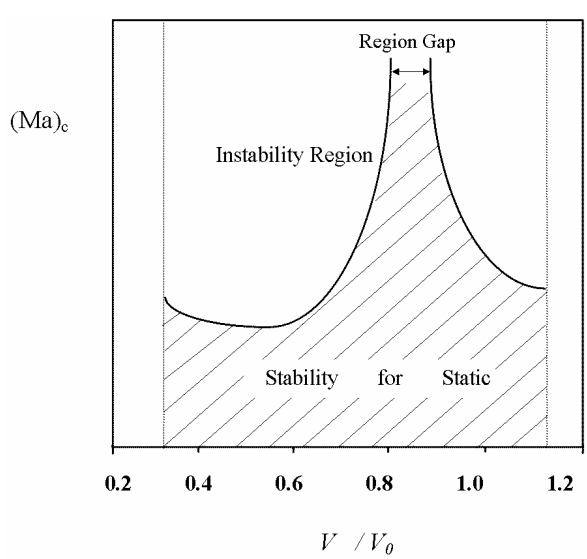


Fig. 2 Typical marginal curves depending on liquid bridge volume for onset of oscillatory thermocapillary convection.

3 Typical transition regions

In the present paper, the hybrid method of fractional step is used [19]. The numerical meshes are 12×16×12 in r, θ, z direction respectively, and the floating half zone is divided into 10758 quadrfaces-volume elements associated with totally 2064 nodes and 172 nodes in each layer. In order to consider the non-liner convective effects, the vorticity and energy equations are classified into the convection and the diffusion parts. The characteristic line method and the FEM method are applied respectively to the convective terms and the diffusion operators.

The onset of thermocapillary convection is related to the grid adopted in the calculation. For checking the three-dimensional, axi-symmetric program of present paper, the numerical calculation on the thermocapillary convection for a cylindrical liquid bridge with g = 0 and $\ell/d_0 = 10$ is compared with those obtained by linear stability analysis for an infinite length and cylindrical liquid bridge in microgravity environment [20]. The results coincide quite well, excepted near the free surface. It shows also that the numerical calculation on core

velocity profile for buoyancy convection on the ground in a horizontal cylinder of present program agrees well with the results of Bejan et al [21].

In the present paper, geometrical configuration of calculated liquid bridge is given by d = 15mm and l = 10.5mm, and the aspect ratio is A = 0.7. The liquid medium is 10 cSt silicon oil (Pr = 105.6). The applied temperature difference ΔT increases gradually from zero at the beginning, and the heating rate is 0.1°C/sec. The initial values of the temperature and the velocity are a constant and zero respectively in the liquid bridge. Then, the evolution of floating zone convection can be obtained step by step during the heating process, and the transition from steady and axial symmetric convections can be determined. Typical transition process depending on the liquid bridge volume is given by Fig. 3, and the marginal curves show three typical transition region. Curve 1 gives the transition from steady and axial symmetric convection to oscillatory convection in the slender liquid bridge. Curve 2 gives respectively the transition from steady and axial symmetric to steady and axial asymmetric convection, and curve 3 gives the transition from steady and axial asymmetric convection to oscillatory convection in the fat liquid bridge. Typical regions 1, 2, and 3 can be defined respectively just after curves 1, 2 and 3, and the typical patterns of velocity and temperature are different in different region. The convection will transit to turbulence if the applied temperature difference is increased further.

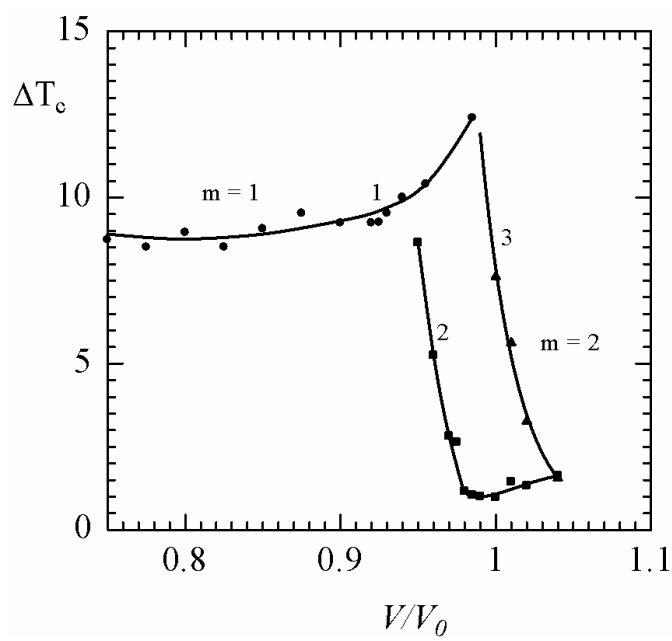


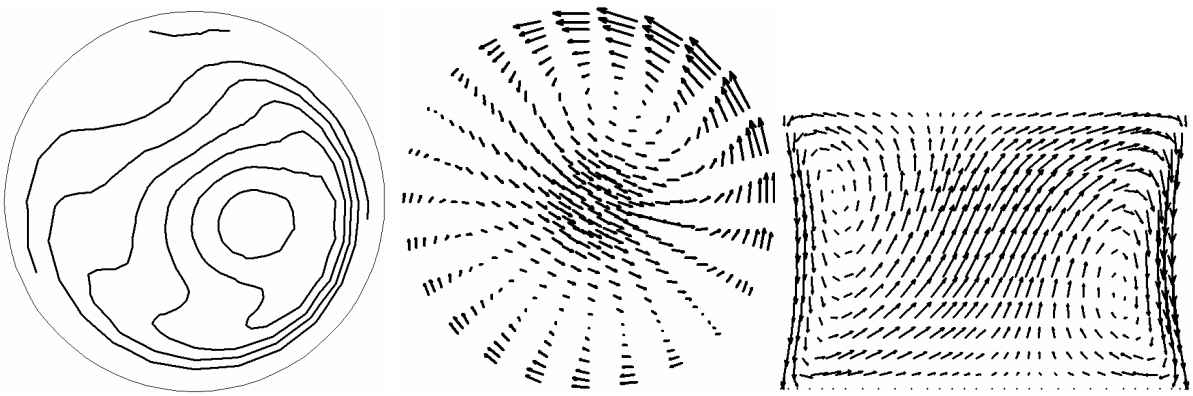
Fig. 3 Curves of regions of bifurcation transitions.

4 Pattern modes

The transition passed cure 1 in region 1 for the slender liquid bridge has only one bifurcation for onset of oscillation, and the steady and axi-symmetric convection transits directly to the oscillatory convection. The oscillatory mode, as an example, for V/V_o = 0.9 in region 1 relates to m = 1 as shown in Fig. 4 in case of temperature difference Δ T = 23.8°C, and temperature and velocity profiles in a horizontal cross-section at z = 0.55 are given respectively in Figs. 4a and 4b. Fig. 4c gives the velocity profiles in a vertical cross-section of liquid bridge. All patterns rotate with a same frequency f = 0.051 Hz. Both velocity profiles Figs. b and c show axial asymmetric distributions.

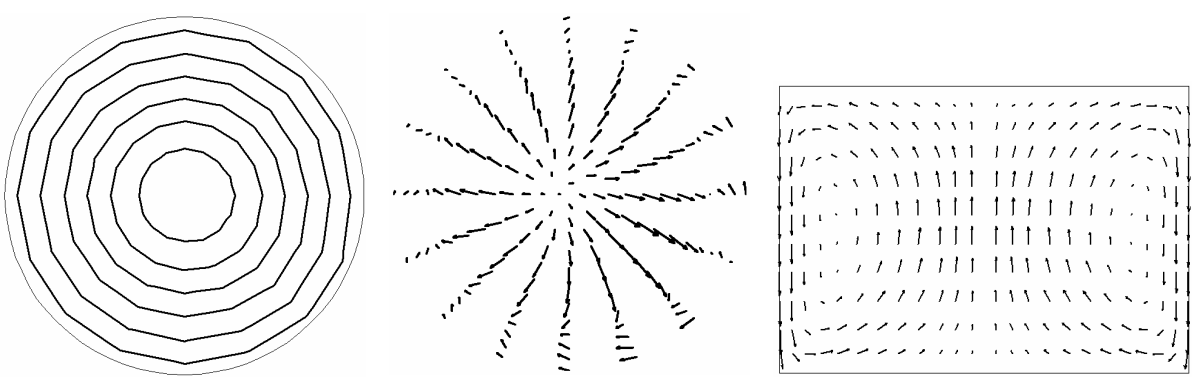
The transition passed cure 2 in region 2 for the fat liquid bridge associates with the transition from steady and axi-symmetric convection to steady and axial asymmetric convection. Fig. 5a and b show the patterns of temperature and velocity in a horizontal cross-section at z = 0.55 for volume ratio of liquid bridge V/V_o = 1.01 in case of temperature difference ΔT = 3 °C, and the patterns are closed curves with centers closing to the symmetric center of liquid bridge. Velocity patterns at a vertical cross-section as shown clearly in Fig. 5c are

axial symmetric. All patterns are time independent, and these features are seen in Fig. 5d, where the time evolution of temperature at 4 points of 90º separated each other at the boundary of cross section z = 0.55 for a temperature difference ΔT = 3 ºC. The evolutionary curves keep different constants and relate to mode m = 1.



a) b) c)

Fig. 4 Profiles of temperature in a horizontal cross-section (a), velocity in a horizontal cross-section (b) and velocity in a vertical cross-section (c) in region 1 for ΔT = 23.8 ^{O}C and V/V_o = 0.9.



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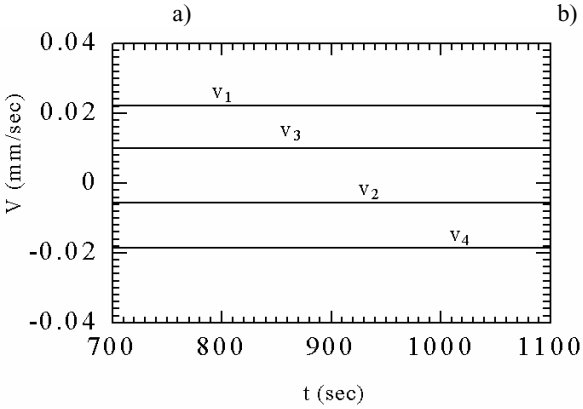
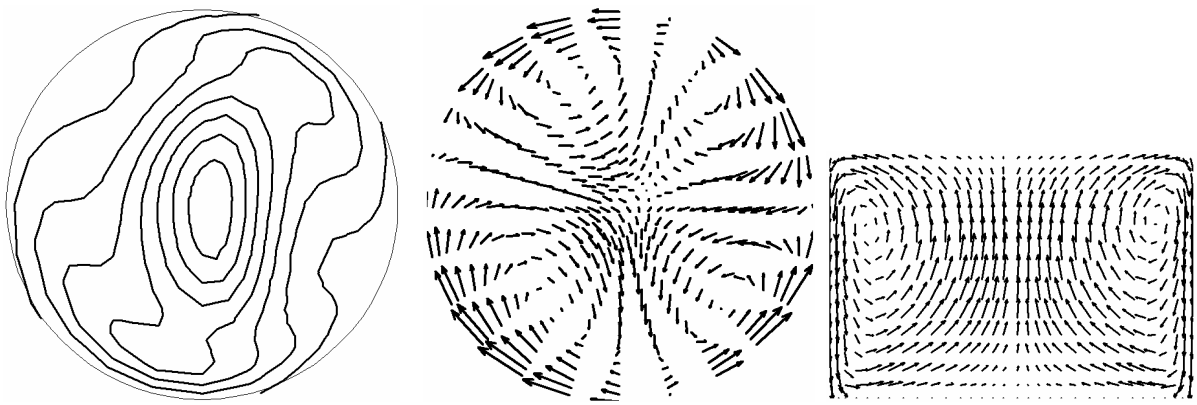


d)

Fig. 5 Profiles of temperature in a horizontal cross-section (a), velocity in a horizontal cross-section (b) and velocity in a vertical cross-section (c) in region 2 for ΔT = 3 ºC and V/V_o = 1.01

The transition passed cure 3 in region 3 for the fat liquid bridge relates to onset of oscillation from the steady and axial. Fig. 6a and b are temperature and velocity profiles in a horizontal cross-section z = 0.55 of a fat liquid bridge V/V_o = 1.02, in case of temperature difference ΔT = 15.8ºC. Fig. 6c gives the velocity

distributions in a vertical cross-section. All patterns rotate with the same frequency f = 0.035 Hz, and relate to oscillatory mode m = 2. Fig.6 b shows the axial asymmetric distribution, and Fig. 6 c shows the nearly symmetric distribution in a vertical cross-section but the symmetric distributions change in different cross-section.



a) b) c)

Fig. 6 Profiles of temperature in a horizontal cross-section (a), velocity in a horizontal cross-section (b) and velocity in a vertical cross-section (c) in region 3 for $\Delta T = 15.8$ °C and $V/V_o = 1.02$.

Two bifurcation transitions for fat liquid bridge are obtained as shown in Figs. 5 and 6 for a floating half zone with aspect ratio A = 0.7. The steady pattern mode is m = 1 for first bifurcation transition, and the oscillatory mode for second bifurcation transition is m = 2. These mode features on floating zone of larger Prandtl number is different from ones of small Prandtl number, where the modes of first and second bifurcation transitions relate respectively to m = 2 and m = 1 in a fat liquid bridge of cylindrical floating half zone [14]. On the other hand, oscillatory mode of slender liquid bridge is m = 1. These conclusions based on the numerical simulation are confirmed by the ground-based experiments in a small liquid bridge, which will be discussed elsewhere.

5 Conclusion

In the present paper, the numerical simulation method is used to discuss the onset of bifurcation transition depending on the liquid bridge volume, and discussions are emphasized in the pattern and oscillatory modes associated with the transition process. The oscillatory mode for onset of oscillatory thermocapillary convection in the slender floating half zone is m = 1; and the pattern modes and oscillatory mode for onset of first and second bifurcation transitions are respectively m = 1 and m = 2. The pattern modes in the floating zone convection of large Prandtl number fluid discussed in the present paper are different from that of small Prandtl number.

Onset of oscillatory convection will induce the striations in the grown crystal, and should be avoid in general. The conclusions of this paper show that the liquid bridge volume is a sensitive parameter for the transition process in the floating zone convection. It is beneficial to controlling the liquid bridge volume in the peak region of Figs. 2 and 3, where relate to more steady state. The influence of different oscillatory mode on the crystal growth is a subject, which needs to be studied in future.

Process of two bifurcation transitions in a fat floating half zone is an interesting subject, especially associated with the mechanism of transitions. The so-called hydrothermal instability was used broadly to explain the onset of thermocapillary convection, and required a traveling wave as the oscillatory mode. In the process of two bifurcation transitions, it is obvious that the first bifurcation requires the transition from steady and axi-symmetric state to another steady state, but not a traveling wave. The conclusion on two bifurcation transitions needs to be explained by a mechanism, which is different from the hydrothermal instability.

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