Transitions between steady states, traveling waves, and modulated waves in the system water-isopropanol heated from below

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Using high-resolution laser Doppler velocimetry, we have experimentally obtained comprehensive results concerning the oscillatory onset of free convection (relevant critical Rayleigh numbers and the associated periods of oscillations are in agreement with theoretical predictions), the stable monoperiodic oscillatory states (traveling waves) just past the critical point, and the transition towards steady states for greater Rayleigh numbers. Our most striking result is obtaining, when decreasing the imposed Rayleigh number below the critical point, stable biperiodic oscillatory states (modulated traveling waves).

Free convection in binary mixtures provides a great variety of behaviors due to the competition between two scalar fields (a temperature field and a concentration field) diffusing at different rates. Before 1984, works relevant to this type of convection were mostly concerned with the linear stability theory[1] and with the study of a generalized Lorenz model with five Fourier coefficients;[2,3] a few experiments have shown the increase of the critical Rayleigh number (in comparison with 1708 for pure liquids) induced by a negative value of the separation ratio, together with the existence of oscillations[4] detected by inserting a thermocouple or a diode inside the solution. The "state of the art" up to about 1984 may be found in Ref. 5.

Recently, there has been a huge increase of theoretical[6-10] and experimental[11-16] works on this subject. Experiments have shown the rich dynamics of this type of convection: superposition of standing and traveling waves during the onset of convection;[13] existence of stable states characterized by a system of traveling waves just past the critical point;[14] possibility of modulated traveling waves in some part of the container, etc.[16]

In all cited experiments, the temperature field inside the convective cell is determined (by thermal lens effect); the Nusselt number is also recorded. Such techniques do not allow a quantitative description of the velocity field in the system of rolls. Moreover, visualization of the temperature field implies the use of a transparent conductive upper plate (e.g., a sapphire plate). Even if the thermal conductivity of sapphire is quite good (around 60 times that of water), the use of a copper plate (with a thermal conductivity of more than 600 times that of water) will be much better for comparison with theoretical results which suppose infinite conductivity of the upper and lower plates.

We present in this paper experimental results of different dynamical convective behaviors in two mixtures of water-isopropanol presenting a negative value of the separation ratio and heated from below. To obtain quantitative measurements of the velocity inside a roll, we use high-resolution laser Doppler velocimetry (LDV). The convection cell has an aspect ratio of 1:3.6:28 (all dimensions are reduced by the height $h = 4.15$ mm). The fluids are 10 wt.% isopropanol in water (mixture A) and 20 wt.% isopropanol in water (mixture B) at the mean temperature of 21 °C. The upper and lower plates are made of copper and the cavity is laterally bounded by two Plexiglass frames and glass windows. The temperature gradient is imposed by two thermoregulated water flows (± 0.01 K) in contact with the copper plates.

Linear stability theory[5] predicts, for these mixtures, an oscillatory instability of the rest state. The study of the onset has attracted a lot of attention for several years[6,7,11-16] and, therefore, we shall not give a lot of details. Figures 1 and 2 present the vertical component of the velocity at the center of the cell during the onset of convection. The initial period of the oscillations is 57 s for mixture A and 121 s for mixture B. It is very important to compare these values with theoretical predictions (in the frame of linear stability theory). Values of the separation ratio allows the theoretical determination of the critical Rayleigh number and of the period of initial oscillations. The values found in the literature,[17] which are provided by direct measurements of the concentration gradient induced by a temperature gradient, are too imprecise to be used here. So, there is a suggestion to measure the separation ratio from its effect on the critical Rayleigh number

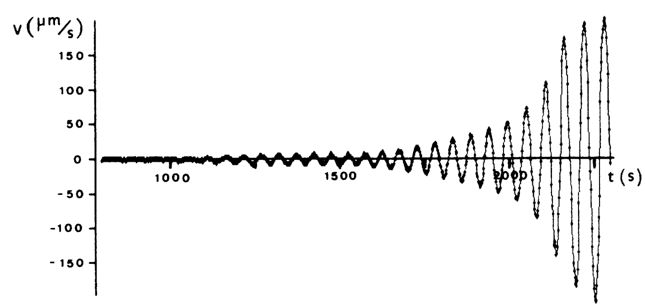

FIG. 1. Vertical component of the velocity during the onset of free convection in mixture A: 10 wt.% isopropanol in water $[D_T/D = -1.2\times10^{-2}\ K^{-1}$ (Ref. 18); $N_{Pr} = 12.6$; $N_{Sc} = 1600$; $N_{Ra}^{th} = 357\,000$ (Ref. 21)]. Zero time corresponds to the increase of the Rayleigh number from $N_{Ra} = 2962$ to $N_{Ra} = 3125$ ($N_{Ra}^{crit} = 2987$) or of ΔT from 3.06 K to 3.18 K.

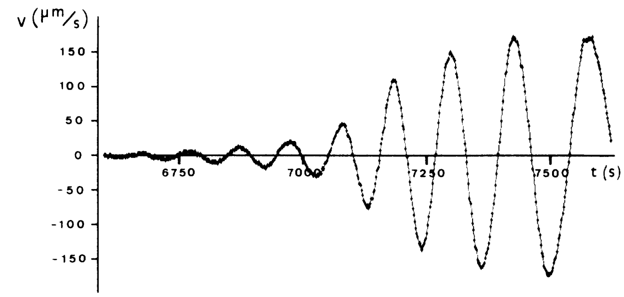

FIG. 2. Vertical component of the velocity during the onset of free convection in mixture B: 20 wt.% isopropanol in water [$D_T/D=-2.75\times10^{-3}$ K^{-1} (Ref. 18); $N_{\rm Pr}=20$; $N_{\rm Sc}=2300$; $R_{\rm Ra}^{\rm th}=348\,000$ (Ref. 21)]. Zero time corresponds to the increase of the Rayleigh number from $N_{\rm Ra}=2017$ to $N_{\rm Ra}=2295$ ($N_{\rm Ra}^{\rm crit}\simeq2071$) or of ΔT from 1.74 to 1.98 K.

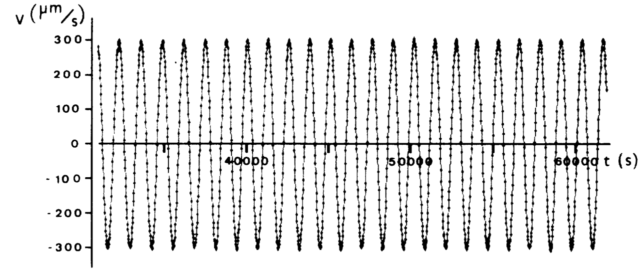

FIG. 3. Time records of the vertical component of the velocity in a mixture 10 wt.% isopropanol in water (mixture A) during the first stable oscillatory state. The Rayleigh number is 3125 ($N_{\rm Ra}^{\rm crit}=2987$). These oscillations correspond to a lateral motion of the system of convective rolls (traveling waves).

(the increase of the critical Rayleigh number) using the linear stability theory and afterwards, from the same theory, to determine the period of initial oscillations. For example, for mixture A, the experiment gives $N_{\rm Ra}^{\rm crit}=2987$, from which we deduce the Soret coefficient[18] D_T/D. Using the method described elsewhere,[19] based on a Galerkin-type numerical technique, we have found a value of $D_T/D=-1.2\times10^{-2}$ K^{-1}, which gives back the experimental critical Rayleigh number of 2987. For the same D_T/D, the linear theory gives (using always the same Galerkin-type numerical technique) the nondimensional frequency $\omega=1.84\times10^{-1}$ and coming back to dimensional values (h^2/ν has been used as scaling factor for time), we find $\omega=1.78\times10^{-2}$ Hz or $T=56$ s. Of course, the kinematic viscosity ν of the mixture at the mean temperature has been measured separately using a standard Ostwald viscosimeter (we have found $\nu=1.67$ cS at 21 °C). For the mixture B, the predicted value of the period is 120 s. Thus, the perfect agreement between experimental values of the temperature gradient at the onset of convection and the frequency at onset gives some confidence to the results and to the way we determine the Soret coefficient.

After several hours, a stable monoperiodic oscillatory state is reached. Periods of oscillations are now 1300 s for mixture A (Fig. 3) and 325 s for mixture B (not shown).

Let us remark that, in all other recent studies of the onset of free convection[11–16] in binary mixtures, the heat flux is imposed. Thus, in these studies, at the onset of convection, there is a drop in the Rayleigh number for a small increase of the heat flux. In contradistinction, we impose here the temperature gradient and thus, at the onset of convection, we move along a vertical line in a graph Nusselt $=f$(Rayleigh). Despite this difference, traveling waves are the preferred mode of convection. In LDV, the experimental proof of this behavior is the following (Fig. 4): In a system of fixed rolls (standing waves), a displacement of the measurement probe produces a time record of the vertical component of the velocity which is still an oscillation but with another amplitude. On the other hand, in a system of traveling waves, after the same displacement, we will record the same oscillation with the same amplitude but with a difference in the phase. Experimentally, we have shifted the measurement probe half a roll (at time $t1$ in Fig. 4): The same oscillation is recorded again with a difference of phase of $\sim\pi/2$. A shift of half a roll in the other direction (at time $t2$ in Fig. 4) gives the same oscillations with a difference of phase of $\sim\pi/2$ in the other direction. We have another proof that these oscillations correspond to a system of traveling waves: Between time $t3$ and time $t4$ (Fig. 4), we move the measurement probe 0.1 mm each 15 s (i.e., 8.7 mm in 1300 s, which is almost the speed of the wave: 2 rolls in a period). A horizontal line is almost obtained which means that the measured velocity is nearly constant. On the other hand, if we move the measurement probe 0.2 mm each 15 s (between time $t5$ and time $t6$ in Fig. 4) in the opposite direction, the recorded period is one-third of the initial period.

Further increases of the temperature gradient decrease the frequency of the oscillations. The branch of traveling waves terminates on a branch of steady-state solutions (Fig. 5). The time evolution towards steady state is quite long and is thus associated with the establishment of a new stable concentration gradient in a convective regime.

When the Rayleigh number decreases from the branch of steady states, traveling waves are recovered (Fig. 6). There are two parts in this process. (i) Just after the modification of the temperature gradient, the velocity de-

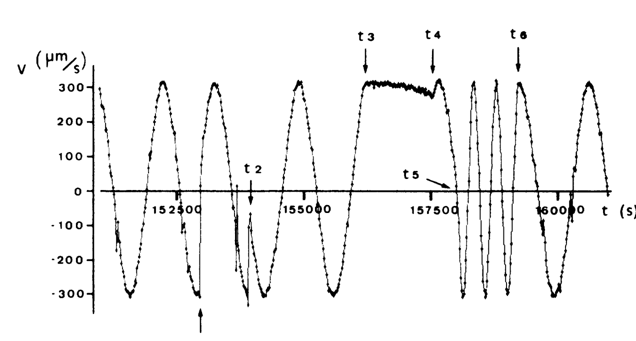

FIG. 4. LDV proof of the existence of a system of traveling waves (mixture A) at $N_{\rm Ra}=3125$: at time $t1$ and time $t2$, the measurement probe is shifted half a roll; between time $t3$ and time $t4$, the measurement probe "follows" a roll; between time $t5$ and time $t6$, the measurement probe is continuously shifted in the opposite direction.

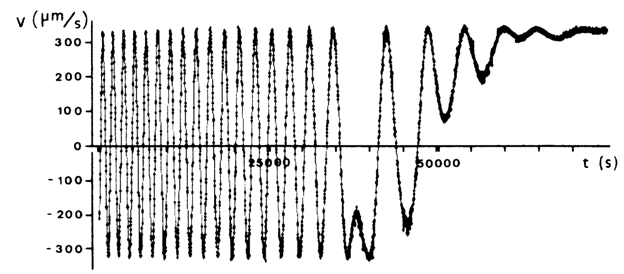

FIG. 5. Mixture *A*: vertical component of the velocity during evolution towards steady state when increasing the Rayleigh number from 3125 ($\Delta T = 3.18$ K) to 3243 ($\Delta T = 3.30$ K). The evolution towards a steady state is quite long; after a continuous increase of the period of oscillations during 10 h (corresponding to a slower and slower displacement of the system of rolls), the system of rolls oscillates around a fixed position.

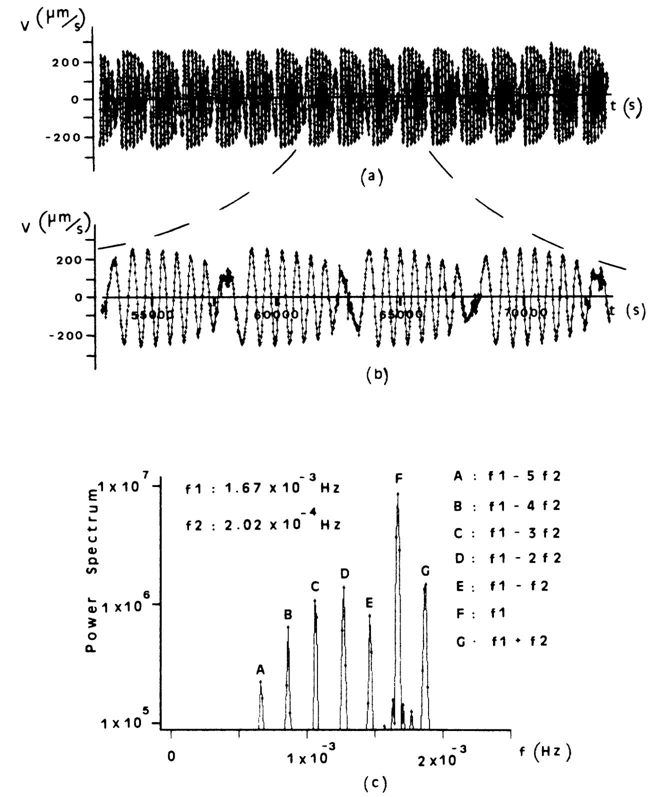

FIG. 7. Mixture *A*: time record (a) of the vertical component of the velocity at a Rayleigh number of 2565 (thus, below the critical point). Extended part of (a) is given by (b). By taking the Fourier transform (c) of the signal, the two frequencies $f1$ and $f2$ are found. The power spectrum consists of lines at frequencies given by $mf1 + nf2$ (m,n integers).

creases rather quickly (during ~5 min) but the system of rolls remain fixed; and (ii) after a long time (1 h in Fig. 6 but sometimes more), oscillations are recovered. This long time is a proof that oscillations are connected to the establishment of the new stable concentration gradient in the convective regime.

Further decreases of the temperature gradient below the critical Rayleigh number lead the system towards a new state characterized by modulated traveling waves (Fig. 7). By taking the Fourier transform of the signal [Fig. 7(c)], the two frequencies $f1$ and $f2$ are found. The power spectrum consists of lines at frequencies given by $mf1 + nf2$ (m,n integers). It seems that there is no frequency locking [see extended part of Fig. 7(a) on Fig. 7(b)]. These experiments are reproducible, and repeated at intervals of several weeks.

The exact nature of the second "oscillator" is not yet clearly established, but the following should be emphasized: In the case of traveling waves, the axes of the rolls are parallel to the shorter side of the container. Thus, the same temporal signal is obtained irrespective of the position of the optical probe along the roll axis. In case of modulated traveling waves, however, the rolls are distorted and their axes are no longer parallel to the shorter side of the container. The velocity shown in Fig. 7 was recorded at the center of the cavity. When measuring the velocities near the front or back walls, the fundamental frequency $f1$ is not the same in the case of modulated traveling waves. Let us call these two frequencies $f1$ and $f1'$. The second frequency $f2$ is almost (20% error) the difference between $f1$ and $f1'$. Thus, the picture of these modulated traveling waves could be a kind of "zipper state."[11] Another possibility is to relate the variation of $f1$ to a possible existence of a small ramp of Rayleigh number along the axes of the rolls. At large values of the Rayleigh number, the rolls are "strong" and prevent the creation of defects. They adopt a mean propagating velocity. At smaller Rayleigh numbers, when the rolls are "weaker," the creation of defects becomes possible and the propagating velocity could be different near the front and back walls.[20] This explanation is actually under investigation by imposing voluntarily a temperature gradient along the axes of the rolls.

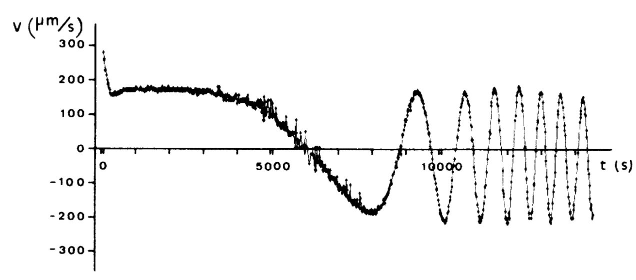

FIG. 6. Mixture *B*: records of the vertical component of the velocity during the decrease of the temperature gradient from a Rayleigh number of 3234 (steady state) to 2295 ($N_{Ra}^{crit} = 2071$). The new stable state is characterized by traveling waves.

In conclusion, we have presented a first LDV description of several convective patterns of natural convection in binary mixtures: traveling waves, transitions towards steady states, and modulated traveling waves. Detailed studies of all behaviors and of transitions between them are still in progress.

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[18]D_T/D is the ratio of the thermal diffusion coefficient D_T to the isothermal diffusion coefficient D. Other authors use the separation ratio ψ. To convert, use the relation

$$\psi = (D_T/D)N_1(1-N_1)\beta/\alpha$$

with N_1 the mass fraction of water, α the thermal expansion coefficient ($\alpha = 3.126\times10^{-4}\ \mathrm{K}^{-1}$ for mixture A and $\alpha = 5.075\times10^{-4}\ \mathrm{K}^{-1}$ for mixture B) and β the mass expansion coefficient ($\beta = 0.126$ for mixture A and $\beta = 0.152$ for mixture B). α and β are provided by interpolation of densities of mixture water-isopropanol [from *International Critical Tables of Numerical Data—Physics, Chemistry and Technology* (McGraw-Hill, New York, 1928), Vol. III, p. 120].

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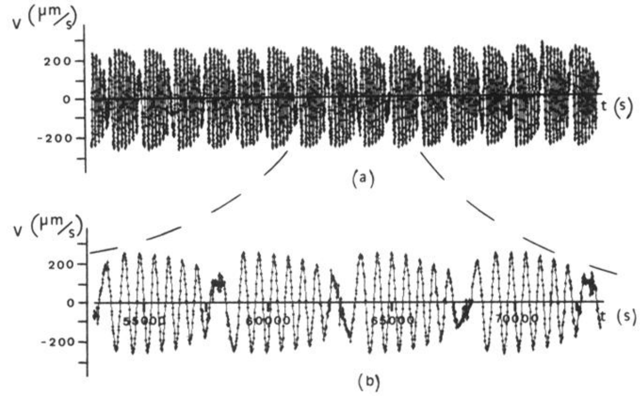
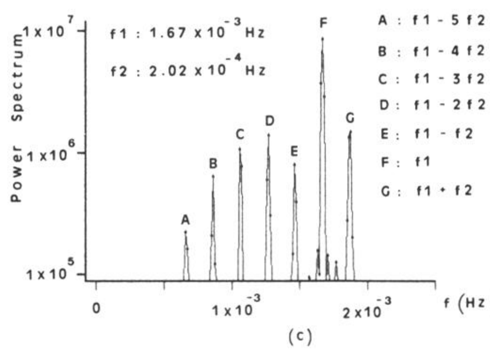
[21]N_{Pr} is the Prandtl number, N_{Sc} is the Schmidt number, and $N_{\mathrm{Ra}}^{\mathrm{th}}$ is the Rayleigh number of thermodiffusion $g\beta N_1 h^3/\nu\kappa$ with g the gravity acceleration and κ the thermal diffusivity (other parameters are already defined).





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