Large-scale convection induced by the Soret effect

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We present an experimental study of free convection in binary mixtures when convection is theoretically predicted to appear in the form of rolls of large extent. Quantitative measurements of the very slow velocities associated with this convective pattern are presented.

Rayleigh-Bénard convection in binary mixtures has attracted much attention as it provides the opportunity to study a rich variety of dynamical behaviors due to the coupling between two scalar fields (a temperature and a concentration field) diffusing at different rates (a difference of several orders of magnitude between heat and mass diffusive times is usual). The interest in this problem is such that in 1988, Physical Review and Physical Review Letters have published not less than 18 papers on the subject (Refs. 1 to 18). Attention is usually restricted to negative values of the Soret coefficient D_T/D (or of the so-called separation ratio ψ defined by $\psi=(D_T/D)N_1N_2(\beta/\alpha)$, where D_T is the thermal diffusion coefficient, D the isothermal diffusion coefficient, N_1 the mass fraction of the denser component 1, $N_2=1-N_1$, α is the thermal expansion coefficient, and β is the mass expansion coefficient). In that case, a lot of dynamical behaviors has been experimentally discovered (steady overturning convection,[19] traveling waves,[9,13,20–22] modulated traveling waves,[9,23] counterpropagating modulated waves,[24] localized traveling waves,[17,25] zipper,[20] . . .).

Little attention has been given to the positive Soret coefficient and to the double diffusive convection, except in the older references (for a review, see, e.g., Ref. 26). To our knowledge, only two recent papers[7,27] has been devoted to the case of a positive value of the separation ratio. In such a case, it is well known[28–31] that in the frame of a linear stability analysis, the critical Rayleigh number R_{ex} (the index ex indicates that the "principle of exchange of stability" is verified) goes down and asymptotically tends to zero when ψ tends to infinity. In recent papers, Knobloch and Moore[1] and independently Cross and Kim[3,4] give the "exact" solution in contradistinction with, e.g., the restricted variational formulation (of Galerkin type) used in Ref. 31. Also the critical wave number k_{ex} tends to zero and that means that convection should take place in rolls or in convective cells of large size. Using the same equations as in Refs. 1, 3, and 4, a typical plot of R_{ex} and of k_{ex} as a function of the separation ratio is given on Figs. 1 and 2 where the Lewis number L is 0.01 (the Lewis number L is defined by $L=\kappa/D$, where κ is the thermal diffusivity). This value of the Lewis number more and less corresponds to the mixture 58 wt. % water–42 wt. % isopropanol used in one of the laboratory experiments described below. For this mixture, the direct measurement of the Soret coefficient gives[32] $D_T/D=5.125\times10^{-3}$ K^{-1}. The values of the other physical parameters are $\kappa\simeq1\times10^{-3}$ $cm^2\,s^{-1}$, $D\simeq1\times10^{-5}$ $cm^2\,s^{-1}$, $\alpha=7.57\times10^{-4}$ K^{-1}, and $\beta=0.231$ (the values of α and β are well known from tables;[33] the value of κ corresponds to an interpolation between the

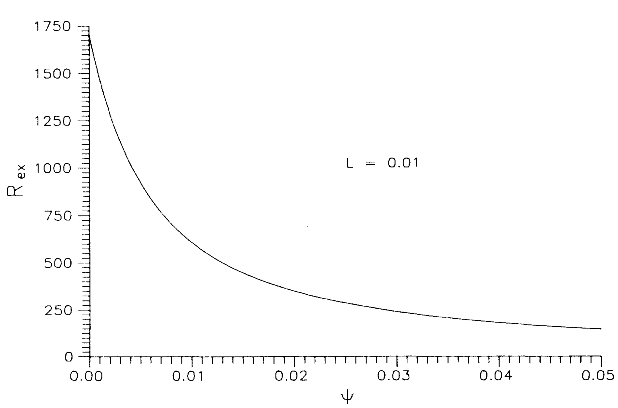

FIG. 1. Plot of the critical Rayleigh number for the exchange of stability R_{ex} as a function of the separation ratio ψ for a Lewis number of 0.01.

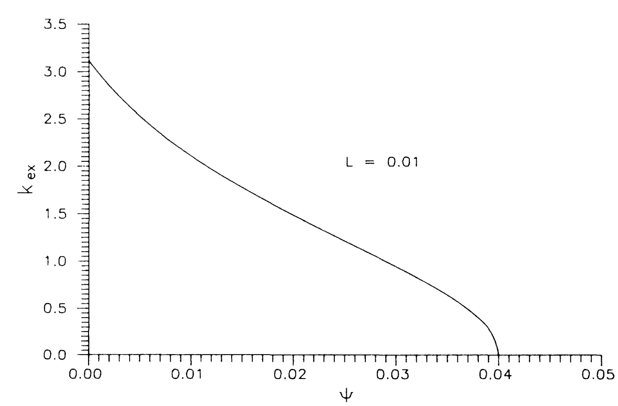

FIG. 2. Plot of the critical wave number k_{ex} as a function of the separation ratio ψ for a Lewis number of 0.01.

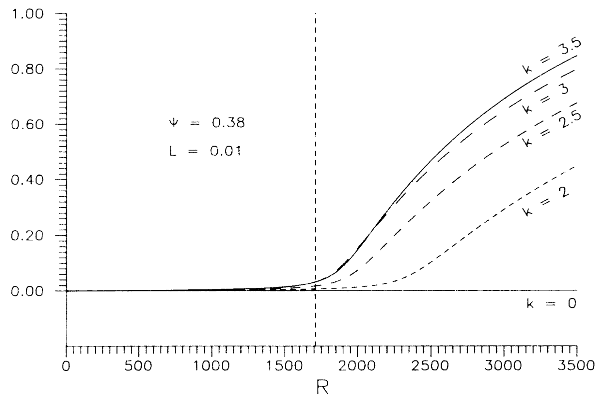

FIG. 3. Result of the study of a five-modes "Lorenz-like" model for rigid conducting and impervious boundaries at $\psi=0.38$ and $L=0.01$: plot of the Nusselt number as a function of the Rayleigh number for several values of the wave number k. Whatever the wave number may be, the Nusselt number remains small (but greater or equal to 1) as long as $R_{ex} < R < R^0$ (R^0 is the critical Rayleigh number in the case of a pure fluid layer, i.e., at $\psi=0$) (R^0 is shown by the vertical dashed line).

values of pure liquids; for D, we give an order of magnitude). From the calculated value of the separation ratio $\psi=0.38$ and results of the linear stability analysis,[1,3,4] we find $R_{ex}\simeq 20$ and $k_{ex}=0$. It is intuitively clear that in such a case, convection is unable to transport heat and that a Schmidt-Milverton plot will not be able to reveal an instability at small Rayleigh numbers. Let us, for example, mention the experiments at $D_T/D>0$ performed by Legros *et al.* on the systems CCl_4-C_6H_6 (Ref. 34) CCl_4-C_6H_5Cl (Ref. 34), CCl_4-cycloC_6H_{12} (Ref. 35) and $C_2H_2Br_4$-$C_2H_2Cl_4$ (Refs. 36 and 37) which only show a change of slope in the heating curves at much higher Rayleigh numbers, probably indicating a secondary instability corresponding to the breakdown of the unique convective cell into a usual multicellular flow. This is also clear from nonlinear calculations using a Fourier expansion or a finite difference technique. For example, Platten and Chavepeyer[38] used a five-modes representation (probably sufficient to describe steady convection not too far from the critical point) in the case of free conducting and permeable boundaries and found that the Nusselt number remains small as long as $R_{ex} < R < R^0$ (R^0 is the critical Rayleigh number in the case of a pure fluid layer, i.e., at $\psi=0$); near $R=R^0$, there is a sudden increase of the Nusselt number. However, in the case of free conducting and permeable boundaries, the critical wave number remains constant ($k_{ex}=\pi/\sqrt{2}$) and does not go to zero, a situation which finally is not comparable with real laboratory experiments. Study of the equivalent model (five modes) for rigid conducting and impervious boundaries shows that the Nusselt number N remains 1 if $k=0$ and that N strongly increases near $R=R^0$ (Fig. 3) whatever the wave number may be. The decrease of the wave number is also clear in a few numerical simulations using the finite difference technique to integrate the governing equations in the case of rigid conducting and impervious boundaries[26]: When $\psi=0$, in a rectangular container of aspect ratio $l/h=10$ (l is the length of the container and h the depth), a steady state with 10 rolls is reached but when ψ increases ($\psi>0$) steady convection is found at smaller and smaller Rayleigh numbers and at the same time, the number of rolls decreases; for example, when $\psi=0.028$, $L=0.01$, a Galerkin-type technique[31,39] gives $R_{ex}=250$. The exact linear theory[1,3,4] provides $R_{ex}=255$. The numerical experiment described in Ref. 26 was done at a Rayleigh number of 400. The convective structure is characterized by only 6 rolls in the container (instead of 10 when $\psi=0$), thus a decrease of the wave number, but not as drastic as predicted by the linear theory.

In partial conclusion, heat-flow measurements are inadequate to study convection at $\psi>0$ and it is reasonable to ask if a direct measurement of the velocity itself is possible and could throw some light on the problem of convection for $R_{ex} < R < R^0$.

As explained elsewhere,[40] our laboratory is equipped with a high-resolution laser Doppler velocimetry (LDV) system permitting the measurement of extremely small velocities: Values as small as a few $\mu m\,s^{-1}$ are not unusual. Since the optical probe in LDV has a typical diameter of 100 μm and if the duration of the Doppler burst is typically of the order of 20 sec (and there is no difficulty for a photomultiplier tube to follow a light intensity variation of small frequency), a typical velocity of 5 $\mu m\,s^{-1}$ can be measured; of course, conventional counters cannot be used; instead the Fourier transform of the signal is performed. A typical Doppler burst and its Fourier transform recorded during one of the experiments described below is shown on Fig. 4. In order to obtain the velocity in $\mu m\,s^{-1}$, the frequency of the Doppler burst [e.g., 6 Hz on Fig. 4(b)] must be multiplied by the fringe spacing (1.7 μm in our conditions). Thus high-resolution LDV seems appropriate to study the velocity field at small Rayleigh numbers.

As already said, we have performed an experiment in a mixture 58 wt. % water–42 wt. % isopropanol using the same experimental cell as described elsewhere.[9,40] Let us recall that the aspect ratios are 1:3.6:28 (all dimensions are reduced by the height $h=4.15$ mm). We tried to measure the vertical component of the velocity V_z at $z=h/2$ as a function of the imposed temperature difference ΔT. Table I gives the maximum values of V_z and V_z^2 that could be obtained by translating the measurement probe in the horizontal direction. Below $\Delta T=1.00$ K, no vertical velocity component could be detected in the central part of the container. Figure 5 gives $(V_z^{max})^2$ as a function of ΔT and the dashed lines indicate an "instability" at $\Delta T\simeq 1.1$ K, i.e., at a Rayleigh number of $\simeq 1500$ (using the values of the physical parameters previously given and the measured value of the kinematic viscosity $\nu=4$ cSt). Let us also emphasize that in the cases marked with an asterisk in Table I, we have found the presence of a standard roll pattern comparable with the one observed in the case of a pure fluid. However, since the first instability is expected in the form of a large convective cell, it is, of course, evident that the detection of this type of convection needs the measurement of the horizontal ve-

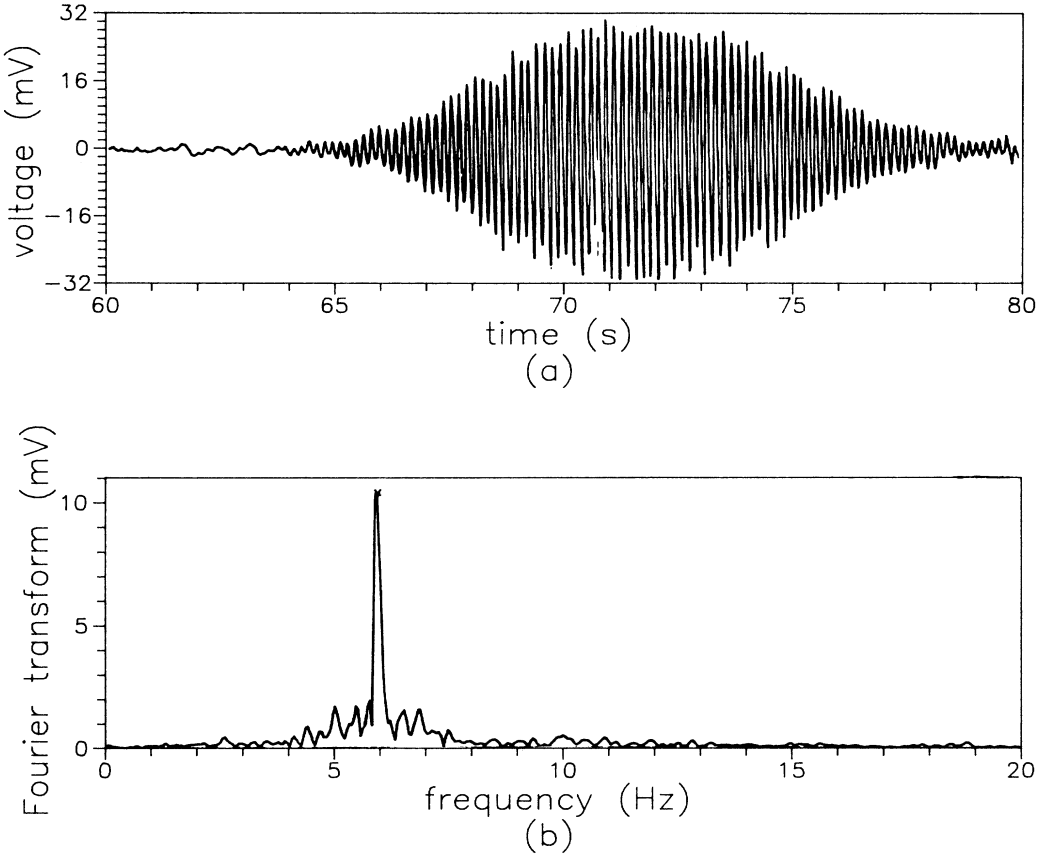

FIG. 4. Example of a Doppler burst (a) and its Fourier transform (b). In order to obtain the velocity in μm s^{-1}, multiply the frequency by the fringe space 1.7 μm.

locity component V_x. This has been done at $z=0.8h$ where it is expected that V_x is maximum. Table II and Fig. 6 show the observed values of $V_x^{\max}$ as a function of ΔT. In contradistinction with measurements of the vertical component, velocities are observed for $\Delta T=0.5$ K (or a Rayleigh number of 661). Thus convection is detected for a value of the Rayleigh number largely smaller than the critical value in the case of a pure layer (1708). The inlet of Fig. 6 clearly shows the destabilizing effect of a positive value of the separation ratio. Let us remark that the extrapolation of the curve $(V_x^{\max})^2=f(\Delta T)$ and the calculation of R_{ex} suggest the existence of convection even for $\Delta T=0.22$ and 0.34 K, not observed from Table II, probably due to the limit of detection of our LDV system.

TABLE I. Measurement of the vertical component of the velocity as a function of the imposed temperature gradient. In the cases marked with an asterisk, we have found the presence of a standard roll pattern comparable with the one observed in the case of a pure fluid.

ΔT (K)	$V_z^{\max}$ (μm s^{-1})	$(V_z^{\max})^2$ [(μm s^{-1})^2]
1.08	3.9	15.21
1.22*	124	15 376
1.34*	149	22 201
1.64*	200	40 000
1.98*	264	69 696
2.37*	340	115 600
2.80*	389	151 321

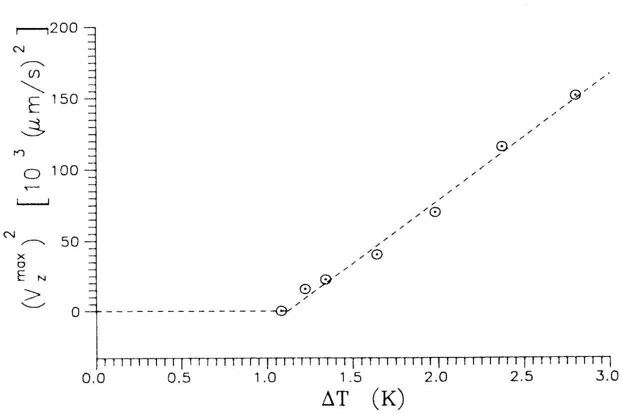

FIG. 5. Plot of $(V_z^{\max})^2$ as a function of the imposed temperature difference in the mixture water 58 wt. %–isopropanol 42 wt. %.

TABLE II. Measurement of the horizontal component of the velocity as a function of the imposed temperature gradient.

ΔT (K)	V_x^{max} (μm s^{-1})	$(V_z^{max})^2$ [(μm s^{-1})^2]
0.00	a	a
0.22	a	a
0.34	a	a
0.50	2.9	8.41
0.59	3.9	15.21
0.73	4	16
0.83	3.8	14.44
0.88	3.9	15.21
0.92	4.5	20.25
1.03	4.9	24.01
1.16	31	961
1.24	117	13 689
1.32	133	17 689
1.44	159	25 281

[a]No detected Doppler burst.

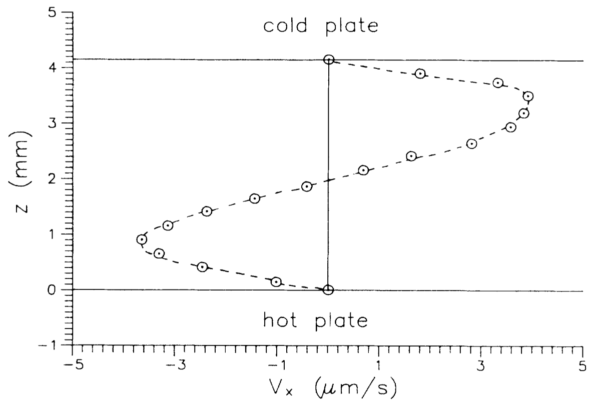

FIG. 7. Measurement of the horizontal component of the velocity as a function of the height in the mixture water 58 wt. %–isopropanol 42 wt. % at $\Delta T = 0.88$ K.

In order to study the convective structure associated with the velocities presented at small ΔT in Table II, we have measured V_x as a function of the vertical coordinate (Fig. 7) at $\Delta T = 0.88$ K. We have also verified that V_x^{max} remains constant along the two horizontal direction (except for the boundary effects). So we have detected that the convective structure consists in only one cell.

In order to prove that the velocities measured in the mixture are relative to the Soret effect and not due to some possible artifact, we have repeated the experiment in pure water in the same container for temperature differences smaller than the critical value ($\Delta T_{ex} = 1.68$ K): $\Delta T = 0.23$, 0.90, and 1.28 K. Sometimes, some small horizontal velocity is recorded, related to "some noise," since the measurements of V_x as a function of the height in pure water at $\Delta T = 0.9$ K has shown the absence of an organized structure in contradistinction with the measurements in the mixture. Another possible artifact is the existence of a small inclination of angle θ of the container giving rise to an horizontal temperature gradient that could induce a velocity profile comparable to the one represented on Fig. 7. The question is thus "what is the value of θ needed in order to induce a maximum velocity of ~ 4 μm s^{-1}?" The horizontal velocity in an inclined container is given by

$$V_x = \frac{g\alpha \Delta T h^2}{12\nu}(\sin\theta) Z(2Z-1)(Z-1) \quad (0 \le Z \le 1) . \quad (1)$$

This law has been checked in pure water (having the same ratio α/ν as the studied mixture) with a value of θ of 2°. The mean measured value of V_x^{max} is 9.9 μm s^{-1} whereas the calculated value is 9.8 μm s^{-1}. By using Eq. (1), we may find that the value of θ needed to obtain a maximum value of V_x of 3.9 μm s^{-1} is 1°. This nonhorizontality (2 mm over the length of the container) easily visible with an air level could not be missed in the experiment on the mixture. So we conclude that all the measurements reported in Table II are related to the Soret effect.

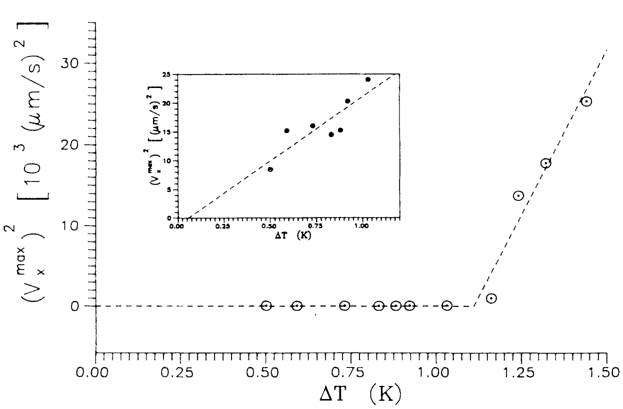

FIG. 6. Plot of $(V_x^{max})^2$ as a function of the imposed temperature difference in the mixture water 58 wt. %–isopropanol 42 wt. %. Inlet is a magnification of the first seven points ($\Delta T \le 1.03$ K).

We have also studied convection in a water-isopropanol system presenting a negative value of the separation ratio heated from above. In such a case, convection is unexpected[41] as it is predicted to appear in a hydrostatically stable system. This is the so-called "double diffusive convection." Study of the stability[28,41,42] of the rest state shows numerous similarities with the convection at a positive value of the separation ratio: Convection is predicted to appear at very small values of the negative Rayleigh number once again in the form of cells of a large extent. In the mixture water-isopropanol used (90 wt. % in water; $\psi = -0.44$, $L = 0.007\,95$), convection is predicted to appear for a Rayleigh number of -13 which means a temperature difference of the order of -0.013 K. So it seems very difficult to experimentally determine such a critical value. Moreover, study of the convection requires the possibility of detection of very small velocities. Indeed, in the frame of a nonlinear

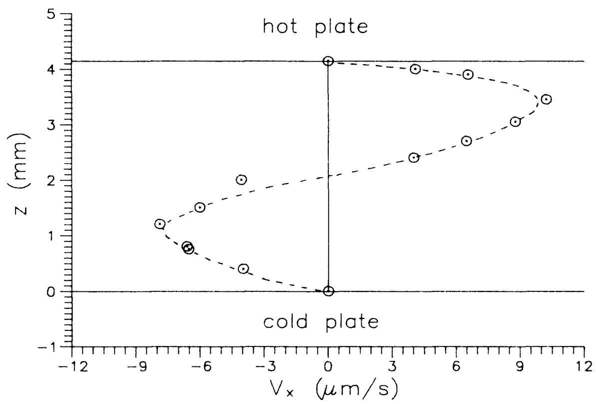

FIG. 8. Measurement of the horizontal component of the velocity as a function of height in a mixture of water 90 wt. %–isopropanol 10 wt. % heated from above at a reduced value (reduction by the length of the cell) $X=0.12$ at $\Delta T=-5.64$ K.

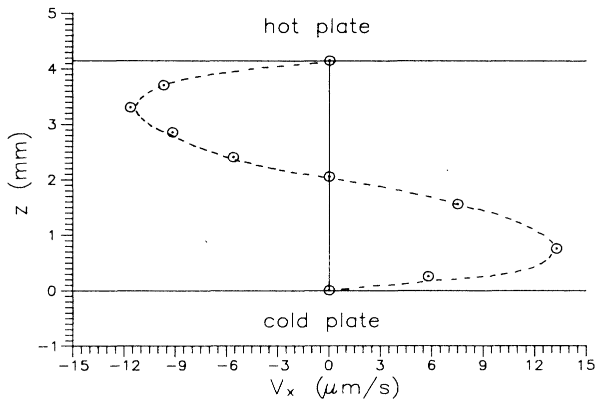

FIG. 9. Measurement of the horizontal component of the velocity as a function the height in a mixture of water 90 wt. %–isopropanol 10 wt. % heated from above at a reduced value (reduction by the length of the cell) $X=0.74$ at $\Delta T=-5.64$ K.

theory,[42] estimation for a LiI-H_2O mixture in a cell of 1 cm depth, for a temperature difference of 10 K gives a horizontal velocity of 32 $\mu m\,s^{-1}$. Such a small velocity explains why, to our knowledge, there is no direct experimental evidence of this type of convection. In order to obtain an experimental proof of convection, we have heated from above the mixture 90 wt. % water–10 wt. % isopropanol ($\Delta T=-5.87$ K). As in the experimental case described above for a positive value of the separation ratio, no vertical component of the velocity was detected and only horizontal component of the velocity was observed. At a given point, at a reduced height of 0.8, we have measured V_x, recording 129 bursts during a time interval of 1 h (one of them is presented on Fig. 4). Since we want to prove that the observed velocities are due to the Soret effect, we repeated the same experiment in pure water with no change in the experimental conditions: No burst was observed during the same time interval. Next, we want to determine the convective structure. Figures 8 and 9 present the graph $V_x=f(z)$ at two different values of x ($\Delta T=-5.64$ K): The reduced values of x (reduction by the length of the cell) are $X=0.12$ (Fig. 8) and $X=0.74$ (Fig. 9). These results suggest the existence of two large rolls.

As a conclusion, we have presented some experimental proofs of Soret driven convection in two cases where theoretical predictions suggest the appearance of convection in the form of cells of large wavelength. This origin has been established by comparison with similar experiments performed in pure water.

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