RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 48, NUMBER 2 AUGUST 1993

Measurement of reflection of traveling waves near the onset of binary-fluid convection

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(Received 22 March 1993)

We present direct experimental measurements of the reflection coefficient r and of the group velocity s for traveling-wave convection in a binary fluid. We measure the dependence of r and s on the separation ratio ψ. Theory predicts that, for small enough $|\psi|$, r should vary as $|\psi|^{1/2}$. This dependence was not found. Instead, the value of r is almost constant over the range $-0.136 < \psi < -0.0096$. We conjecture that the variations in concentration near the cell's end walls due to the traveling waves are responsible for the discrepancy.

PACS number(s): 47.27.Te, 47.20.Bp

It is widely accepted that Rayleigh-Bénard convection in a binary fluid is an appropriate system to study pattern formation and selection due to a Hopf bifurcation [1,2]. This system is described by four dimensionless parameters: the Rayleigh number R, which is proportional to the temperature difference across the fluid layer; the Prantdl number Pr, which is the ratio of the viscosity ν to the thermal diffusivity κ; the separation ratio ψ measuring the coupling between the temperature and the concentration gradients produced by the Soret effect; and the Lewis number L, which is the ratio of the concentration diffusivity D to κ. All parameters are easily accessible and are very well controlled experimentally, making convection in a binary fluid an attractive experimental system.

The traveling-wave (TW) state, which appears as a result of a primary Hopf bifurcation, exhibits a remarkably rich variety of pattern dynamics common to a number of different nonequilibrium systems [3]. It was shown both experimentally [3,4] and theoretically [5–7] that the existence of end walls in the direction of TW propagation has a profound effect on the convection onset as well as on the stability of the TW in the convectively unstable regime. For a geometrically confined system an additional parameter, namely, the reflection coefficient r, is needed to characterize the convection onset [5]. In a finite system the spatial growth of the wave amplitude is balanced by losses due to reflection at the end walls.

A direct consequence of the finite geometry is a shift of the convection onset $\epsilon_s = \frac{\Delta T_s - \Delta T_\infty}{\Delta T_\infty}$, compared with the onset for an infinite cell, occurring at ΔT_∞. According to Cross, the physical mechanism causing the reflection is the local heating of the end wall as the TW impinges upon it [5]. Thus, the value of the reflection coefficient depends on the relative values of the thermal properties of the fluid and the walls. Based on this assumption, Cross was able to calculate both the magnitude and the ψ dependence of the reflection coefficient [5,8]. Analytical calculations were performed for unphysical free slip and permeable boundary conditions [5,7]. Recently, numerical calculations were published for realistic boundary conditions [8]. Note that this approach neglects the variations in the concentration field near the wall and the corresponding boundary conditions; i.e., the boundary conditions at the end walls are taken to be the same as in a pure fluid [5,7].

These theoretical efforts provide an opportunity for a quantitative experimental verification of the theoretical predictions, particularly for the expected ψ dependence of r. Previous experiments [9,10] consistently indicated that r is independent of ψ, even down to very small values of $|\psi| \geq 0.01$. On the other hand, the qualitative dependence of r on the thermal properties of the sidewalls agrees well with the theory [10]. Another experiment [11], performed in the range of very negative values of ψ, $-0.262 \geq \psi \geq -0.558$, also indicated that r is about constant within the experimental resolution.

In this Rapid Communication we present a direct experimental determination of r as a function of ψ. We measured, in the same experiment, all the relevant parameters for the determination of the reflection coefficient from the shift in the convection onset. These are ΔT_∞, the threshold temperature difference for the onset of convection in an infinite layer; ΔT_s, the temperature difference at which patterns are first observed; s, the group velocity of the TW; and τ_0, the characteristic time scale of the convection. As a result of the experiment we find that for a wide range of ψ, $-0.136 < \psi < -0.0096$, r is almost constant and does not show the predicted $|\psi|^{1/2}$ dependence.

The experimental system has been described elsewhere [2]. The experiment was conducted in narrow rectangular cells of aspect ratios $\Gamma = l/d = 25.6$ and 34.4 and width $2d$. The cell height $d = 1.874$ mm was adjusted within ~ 1 μm, as measured interferometrically. The lateral walls of the cell were made of a low thermal conductivity plastic (polypropylene, $\lambda = 1.2$ $\frac{\mathrm{mW}}{\mathrm{cm\,K}}$). We used ethanol-water mixtures, with weight concentrations of ethanol ranging from 23% to 28%. In this range, $\mathrm{Pr} \approx 17$ and $L \approx 0.02$, are approximately constant. Small heaters were mounted inside both short lateral walls.

The shift of the convection onset caused by the reflection at the lateral walls is given by [5]

$$\epsilon_s = -\frac{s\tau_0}{\Gamma} \ln r + O(\Gamma^{-2}). \quad (1)$$

We use this expression to directly evaluate r. In order to achieve this goal, we measure in the same experiment ΔT_s and ΔT_∞, which give ϵ_s, s, and τ_0. ΔT_s and τ_0 are evaluated by measuring the temporal growth of the

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amplitude of the linear waves during transients according to the now standard procedure described in Refs. [2,12]. We found $\tau_0 = 0.1 \pm 0.005$, independent of ψ within the experimental error and in good agreement with the theory [13]. The relative uncertainty in ΔT_s for various ψ's is better than 1.0×10^{-4}.

To evaluate ΔT_∞ and s, we developed a thermal pulse technique, which, together with autocorrelation and cross-correlation analysis, allows for a reduction in the experimental error, thus dramatically improving the resolution of r and s, enabling reliable evaluation both of their magnitude and, particularly, of their ψ dependence.

In order to find ΔT_∞ we perturb the system at one of the short sidewalls by launching a heat pulse. The pulses are short, $\delta t/t_0 < 0.1$ (t_0 is the period of the TW) and of small amplitude: $Q/Q_c \approx 2.5 \times 10^{-4}$, where Q is the perturbation heat flux per period and Q_c is the critical heat flux needed to support convection. Figure 1 shows a typical time sequence of the propagation of a heat pulse. As the initial pertubation travels across the cell, it grows and spreads. In order to quantitatively describe the pulse evolution, we assume that the initial perturbation has a Gaussian form. Then, the shape of the propagating pulse remains Gaussian [14] and at any given time t can be described by

$$A(x,t) = a(t)\sigma \exp[\sigma^2 (x - x_0)^2/2] \sin(kx + \phi) \ . \qquad (2)$$

Here $a(t)$ and σ^{-1} are the amplitude and the width of the pulse at the time t, $x_0 = st$ is the pulse peak position, k is the wave number, and $\phi = vt + C$, where v is the TW phase velocity and C is a correction due to dispersion. As was found in Ref. [9], this correction is of the next order and becomes significant only for large values of x. In order to significantly reduce the effect of geometrical imperfections of the convection cell and the optical system, which, small as they are, become very important as ϵ becomes smaller, we compute the spatial autocorrelation function for the pulse shape as is given by Eq. (2). Fitting the pulse shape to Eq. (2) [9] involves five fitting parameters, and produces much larger scatter in the results and larger error bars, although the results obtained by both methods are similar. Thus, the amplitude and width of the pulse are obtained by fitting the autocorrelation of the experimental results to the following expression obtained from Eq. (2):

$$A_{\rm corr}(\Delta x) = \sqrt{\frac{\pi}{2}} \frac{a^2 \sigma}{2} e^{-\sigma^2 \Delta x^2/2} \times [\cos(k\Delta x) - e^{-k^2/2\sigma^2} \cos(k\Delta x + \phi)] \ . \qquad (3)$$

Using the autocorrelation procedure reduces the number of fitting parameters to three, which are a, σ, and k. We neglect the second term in the square brackets since in our experiment $(k/\sigma)^2 = O(100)$. A typical profile of the autocorrelation function is shown in Fig. 2(a). The temporal dependence of $a(t)$ gives the growth rate for a given value of ΔT, which in the linear regime is equal to ϵ/τ_0, where $\epsilon = \frac{\Delta T - \Delta T_\infty}{\Delta T_\infty}$. From the dependence of the growth rate on ΔT we find ΔT_∞ and τ_0. Figure 3 shows the results of this procedure for $\psi = -0.025$. On the same plot we present results for the determination of both ΔT_∞ and ΔT_s.

In order to determine the group velocity s, we computed the spatial cross-correlation function for the propagating pulse for all possible time differences Δt:

$$C_{\rm corr}(\Delta x, \Delta t, t) = \int A(x,t) A(x + \Delta x, t + \Delta t) dx = \sqrt{\frac{\pi}{2}} \frac{a^2 \sigma}{2} e^{-\sigma^2 (\Delta x - s\Delta t)^2/2} \cos(k\Delta x). \qquad (4)$$

Each result was fitted by this expression, which is similar to Eq. (3), except that the Gaussian is shifted by $\Delta x = s\Delta t$. A typical result of this procedure is shown in Fig. 2(b). The relevant parameter of the fit, Δx, is plotted in the inset of Fig. 4 as a function of the time delay in the cross correlation Δt. The slope gives the group velocity.

Figure 4 presents the ratio of the group velocity to the phase velocity, s/v, as a function of ψ obtained by this procedure. Good agreement with the theoretical predictions [13], shown as the solid curve in Fig. 4, is obtained over the whole range of ψ.

We would like to point out the experimental difficulties and limitations in measuring ΔT_∞ and the TW group velocity. The pulse amplitude should be small enough to remain linear; otherwise both s and ΔT_∞ would be significantly in error. On the other hand, it is clear that a too small pulse amplitude makes the determination of the growth rate uncertain due to a small signal-to-noise ratio. The linear pulses grow exponentially downstream with a growth rate $\epsilon/s\tau_0$. The linear dependence of the growth rate on ϵ leads to a strong dependence of the pulse amplitude on the spatial inhomogeneity of the cell. Thus, a height nonuniformity of 5×10^{-4} causes spatial variations in ϵ on the order of $\delta\epsilon = 1.5 \times 10^{-3}$, i.e.,

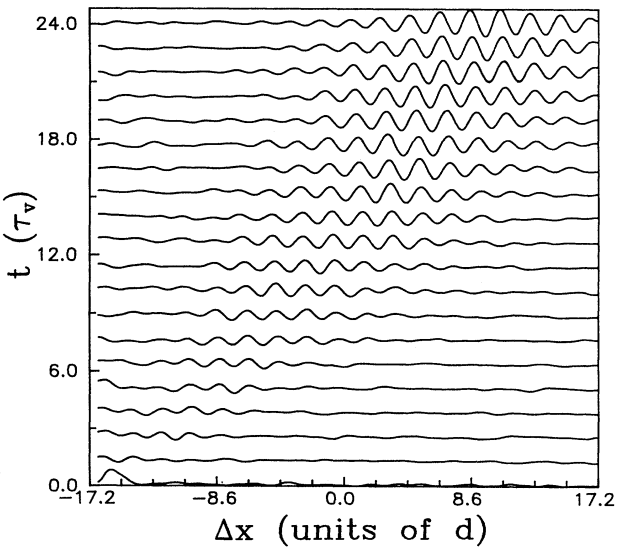
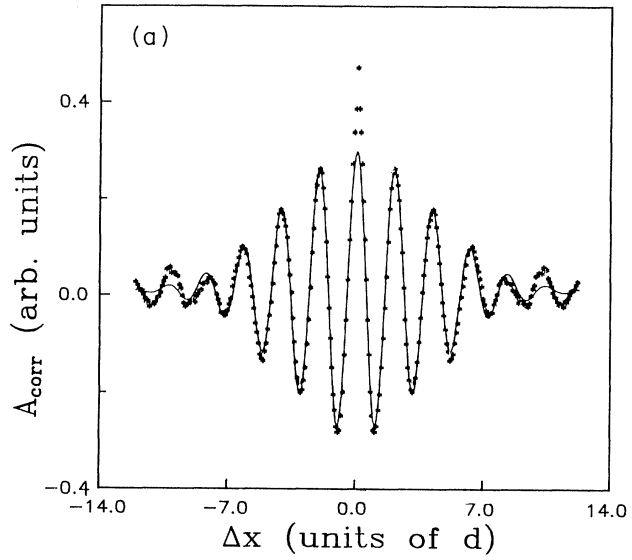


FIG. 1. Spatiotemporal evolution of a pulse. Data are taken at $\Gamma = 34.4$, $\psi = -0.069$, and $\epsilon = 0.005$. The time is measured in units of the thermal diffusion time $\tau_v = d^2/\kappa = 34$ sec.

more than 15% error in ϵ in the working range. In order to overcome both problems, we sampled fast enough to conduct the measurements close to the excitation boundary, where the pulses are still in the linear regime. We verified the linearity of the pulses by checking the deviation of the TW frequency from the neutral frequency as well as checking the pulse spectrum for the existence of higher-order modes. Furthermore, we computed the cross-correlation function for all available data and averaged over the parameter t, appearing in Eq. (4). Thus, for example, the inset in Fig. 4 was plotted by performing ~ 1000 cross-correlation computations. By limiting ourselves to measurements over small time intervals, using cross-correlation analysis instead of directly using the data and performing many averages, we reduce the error in determining s to within 2–3%.

Using all the measured parameters ΔT_∞, ΔT_s, s, and τ_0 we can now compute the value of the reflection coefficient as a function of ψ by substituting them into Eq. (1).



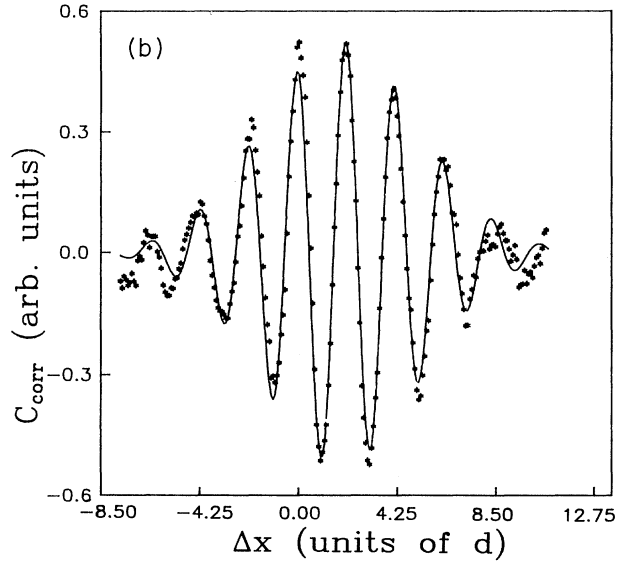


FIG. 2. (a) A typical autocorrelation profile. Data are taken at $\psi = -0.025$, $\epsilon = 0.004$. The fit procedure did not take into account the data within $\pm d$ around zero. (b) A typical cross-correlation profile, for the same set of parameters as in (a). The time difference for the data is $\Delta t = 24\tau_0$.

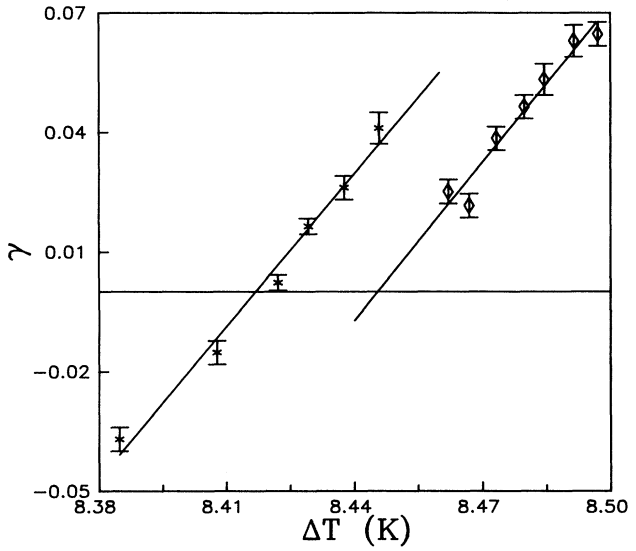


FIG. 3. Temporal growth rate γ for ΔT_s (diamonds) and for ΔT_∞ (stars) as a function of the temperature difference ΔT. $\psi = -0.025$. ΔT_∞ and ΔT_s are obtained from the intersection of the plots with zero. τ_0 is evaluated from the slopes.

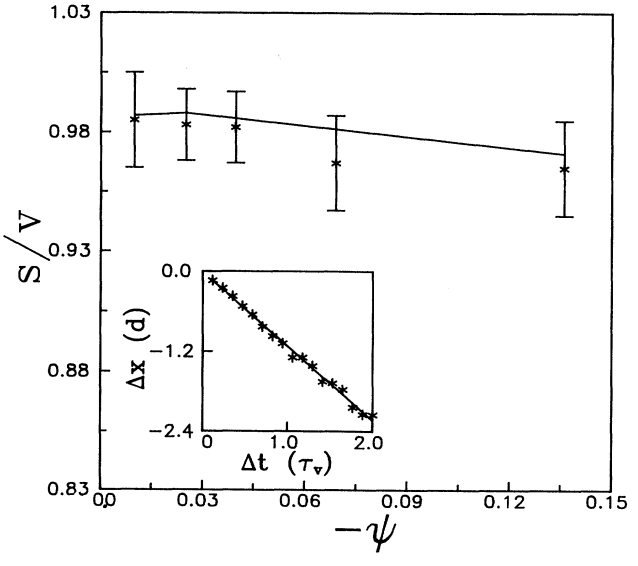


FIG. 4. Group velocity divided by the phase velocity as a function of ψ. The solid line is the theoretical prediction [13]. The inset: the shift of the Gaussian in the cross correlation, Δx as a function of the time delay Δt at $\psi = -0.025$, and $\epsilon = 0.004$.

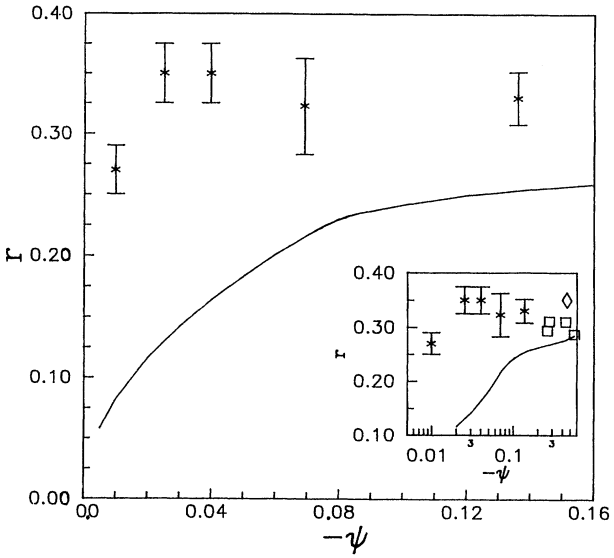


FIG. 5. The reflection coefficient as a function of ψ. The solid line is the theoretical prediction [8]. The inset: our data ($\times$) combined with previously published data [9,11]. Diamonds, data taken from Ref. [9], squares: data taken from Ref. [11].

The results are shown in Fig. 5, together with the recent theoretical calculations for realistic boundary conditions [8]. The main source of error in the determination of r, besides the above estimated error in s, is the uncertainty in the determination of ϵ_s. For fixed s and τ_0, one gets $\frac{\delta\epsilon_s}{\epsilon_s} = \frac{\delta r}{r|\ln r|}$. Then, for a relative uncertainty of 1×10^{-4}, which we find for ΔT_s and ΔT_∞, we obtain $\frac{\delta r}{r} \approx 0.03$. Thus, we estimate the overall error in r to be about 5–7%. The discrepancy between the experimental data and the theory is well outside the experimental error and is especially clear for smaller values of $|\psi|$. In the inset we also present experimental data from Refs. [9] and [11]. Those data were obtained in cells made of ULTEM, a plastic material having twice the thermal conductivity of polypropylene, leading to slightly lower reflection coefficients. It is obvious from the inset that the agreement between theory and experiment at $\psi = -0.558$ is accidental. One can safely conclude from the data presented that the reflection coefficient is almost constant down to very small values of $|\psi|$.

In conclusion, a significant discrepancy between the theoretical predictions and the experimental data has been found. The discrepancy is especially striking considering that $s(\psi)$ and τ_0, which are also computed by the linear theory, are found to agree well with the predicted values. In light of these observations, we conjecture that the problem lies in the boundary conditions for the concentration field of the TW. We suggest that the simplified linear theory, which completely neglects the variation of the concentration field near the lateral boundaries, should be modified. Further theoretical development is needed to explain the experimental data presented.

This work was partially supported by the U.S.–Israeli Binational Scientific Foundation (BSF), Grant No. 90-00412, the Israeli Science Foundation, and the German-Israel Foundation, Grant No. I-130091.

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