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Phase-separating pattern formation in the boundary layer convection of xenon near the gas–liquid critical point

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Abstract. The buoyancy-driven boundary layer convection on a heated vertical plate was investigated in critical xenon employing phase-contrast microscopy. In a boundary layer of about 6 μm thickness vapour tubes with a diameter of the same size and up to several mm in length are observed to form a pattern with a period O(10 μm). The pattern formation is restricted to the fluid temperature interval -3 mK $< T_{fl} - T_c < 1.5$ mK and to the region below the position of the meniscus. The vapour tube formation is attributed to spinodal decomposition due to a local temperature lowering of the boundary layer relative to the bulk fluid.

1. Introduction

Pattern formation is the object of current research in many areas of science [1]. Among the best-known investigations are convection experiments on fluids under normal thermodynamic conditions. Investigations of pattern formation near the gas–liquid critical point have been rare [2] although new phenomena are to be expected due to the anomalous behaviour of many thermodynamic quantities in this region.

Recently we reported on the observation of pattern formation in the boundary layer of critical argon and oxygen on a heated vertical plate [3]. The effect is restricted to bulk fluid temperatures $|\Delta T_c| \equiv |T_{fl} - T_c| < 1$ mK and the region below the position of the meniscus where the bulk fluid density is supercritical. At temperature differences O(1 mK) between the surface of the convection plate and the bulk fluid, vertical convection structures were observed to form in the boundary layer convection with a horizontal period of 40–60 μm and lengths of up to several mm. Here we report on further investigations of this effect in critical xenon which offers the advantage of working at room temperature and of employing high-resolution phase-contrast microscopy.

2. The experimental set-up

The critical xenon ($T_c = 289.7$ K and $p_c = 58.4$ MPa [4]) with a purity of 99.99 vol% was contained in a cell made of copper. Its horizontal bore of 40 mm diameter was closed with two hardened quartz windows of 8 mm thickness and 3.8 mm separation. A heating element ($12 \times 2 \times 2$ mm^3) was glued onto one window about 7 mm below and with its long side parallel to the meniscus in order to generate convection on the inner side of the window (figure 1).

The temperature of the fluid cell was stabilized by means of a water thermostat and an electronically regulated heating coil. The cell was surrounded by a thermal radiation

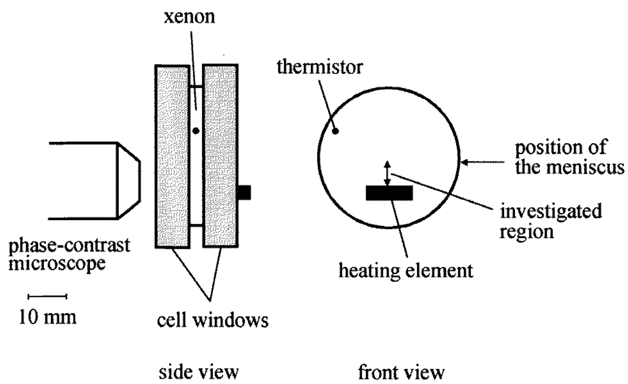

Figure 1. A schematic drawing of the fluid cell for optical investigations of buoyancy-driven boundary layer convection in critical xenon on a heated vertical window; the cell body and window flanges are not shown. The convection is generated by means of the heating element glued onto the outer side (the vacuum side) of one cell window. The pattern formation in the boundary layer is observed by employing phase-contrast microscopy.

shield also connected to the water thermostat and operated in vacuum. The temperature of the cell and the fluid was measured with thermistors. Further details on the temperature measurement and control can be found in [3]. The temperature stability of the cell was better than 50 μK over 5 h.

The cell was filled with liquid xenon in such a way that the meniscus disappeared at T_c in the middle of the cell. The mean fluid density in the cell was estimated to be equal to the critical density with an accuracy of better than 0.4%. The temperature at which the meniscus disappeared could be determined with a precision of better than ± 0.3 mK.

The optical investigations were carried out employing a phase-contrast microscope with CCD readout (20 ms exposure time/frame) and digital image processing in real time. The microscope as well as the head of the CCD camera were contained in the vacuum tank. The temperature of the objective lens and the CCD camera head was stabilized by means of the water thermostat. The lateral resolution of the microscope within the plane of focus was checked with bar gratings to be about 1 μm at $\lambda = 546$ nm. The depth of field along the microscope axis [5] was about 6 μm. The cell could be moved within the vacuum tank by remote-controlled translation stages.

The microscope illumination was placed outside the vacuum tank. Its heat radiation was eliminated by heat absorption windows. No convection due to the illumination could be observed on the cell windows. Due to the density stratification near the critical point [4], beam bending of the illuminating light was to be expected. With the aid of an additional lens which could be moved by remote control into the microscope light path, it was possible to check the illumination of the phase ring in the objective lens during the experiment. The investigations described below were restricted to those regions of fluid height where the beam-bending effect could be ignored and phase-contrast imaging was ensured.

3. Results

While the cell was cooled at a rate of -2 mK h^{-1} from $\Delta T_c = 2$ mK to $\Delta T_c = -6$ mK, CCD image sequences of the boundary layer convection were recorded on video disc for later evaluation. The electrical power of the heating element (figure 1) was adjusted to

0.5 mW resulting in a temperature difference O(1 mK) between the inner side of the cell window and the bulk fluid.

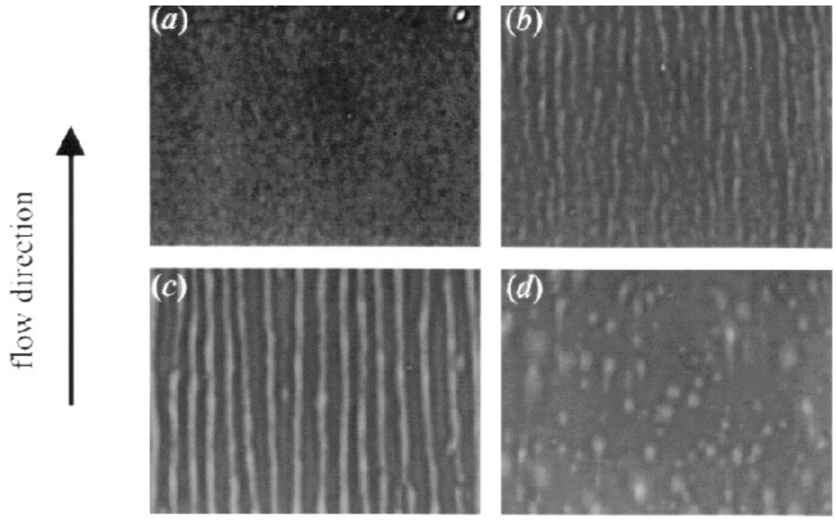

Figure 2. Microscope images of the boundary layer in critical xenon taken with the phase-contrast method at $T_{fl} = T_c - 1.2$ mK and showing the development of the phase-separating pattern at a height of (*a*) −6.4 mm, (*b*) −5.9 mm, (*c*) −4.9 mm and (*d*) −1.1 mm relative to the meniscus (cf. figure 1). The horizontal width of an image is 225 μm.

In figure 2 microscope images of the boundary layer are shown at $\Delta T_c = -1.2$ mK. At a height of −6.4 mm relative to the position of the meniscus the boundary layer shows a granular intensity pattern of low contrast (figure 2(*a*)) which moves upward at a velocity O(100 μm s^{-1}). Focusing into the bulk fluid the upward movement disappears and a speckle pattern [3] with even lower image contrast is observed. A correlation analysis yields a correlation length O(1 μm) corresponding to the lateral resolution of the microscope.

With increasing height the granular pattern in the boundary layer gradually changes to vertically elongated and regularly arranged structures (figure 2(*b*)) whose order and vertical extent increases. In figure 2(*c*) tube-like structures reaching several mm in length and showing a width of typically 6 μm have developed. Near the meniscus these structures eventually disintegrate into small bubbles (figure 2(*d*)) demonstrating that the observed structures are due to liquid–vapour phase separation.

Figure 3 shows a horizontal correlation analysis of the structures corresponding to the experimental conditions of figure 2(*c*). The resulting period of 13 μm is significantly smaller than the one observed in argon and oxygen [3]. The vertical velocity of the tube structures was established to be O(100 μm s^{-1}) by correlating small image windows of size 5 μm × 5 μm in space and time. A typical measurement of the velocity distribution is shown in figure 4.

The location of the vapour tubes relative to the surface of the window was determined by measuring the contrast [3] of the structures as well as the contrast of surface markers (sub-μm dust grains) as a function of the focus position of the microscope in the axial direction (figure 5). The positions of these objects obtained from the contrast maxima are found to be 2.5 ± 0.5 μm apart. Since the width of a single vapour tube is ∼6 μm, this finding is consistent with a tube-like shape of the structures.

We emphasize that structures similar to those in figure 2(*c*), i.e. with comparable width,

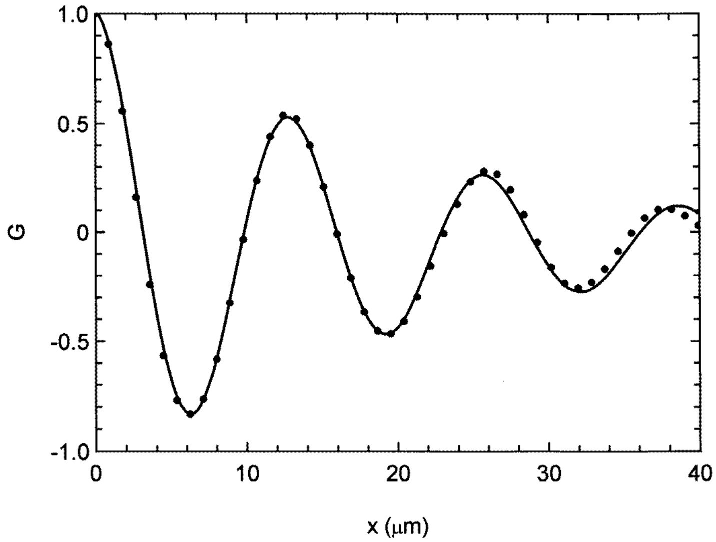

Figure 3. Horizontal correlation G of the tube-like structures shown in figure 2(*c*). The harmonic with an exponential damping fitted to the data yields a period of 13 μm and a correlation length of ∼22 μm.

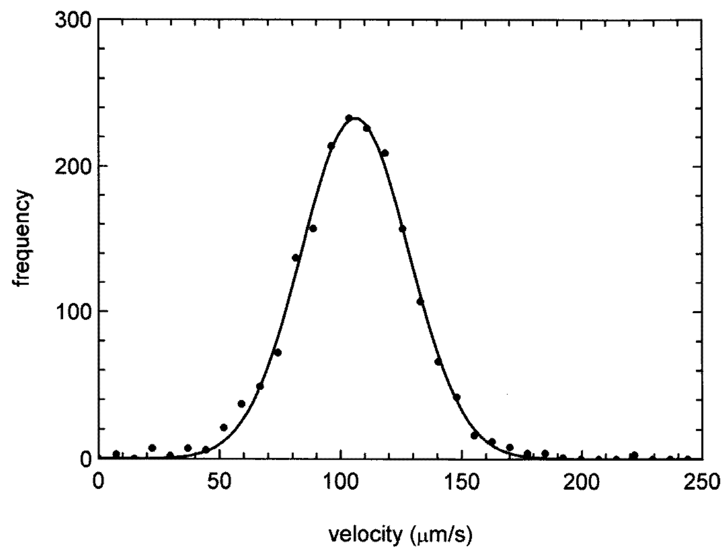

Figure 4. The velocity distribution of the tube-like structures shown in figure 2(*c*). A normal distribution has been fitted to the data.

vertical extension and horizontal period are observed in the boundary layer up to bulk fluid temperatures of $\Delta T_c = 1.5$ mK. Their image contrast decreases with increasing bulk fluid temperature and eventually disappears at temperatures $\Delta T_c > 1.5$ mK. At temperatures above T_c a disintegration of these structures in small bubbles is not observed any longer; instead with increasing height a continuous decrease of the contrast occurs until the structures

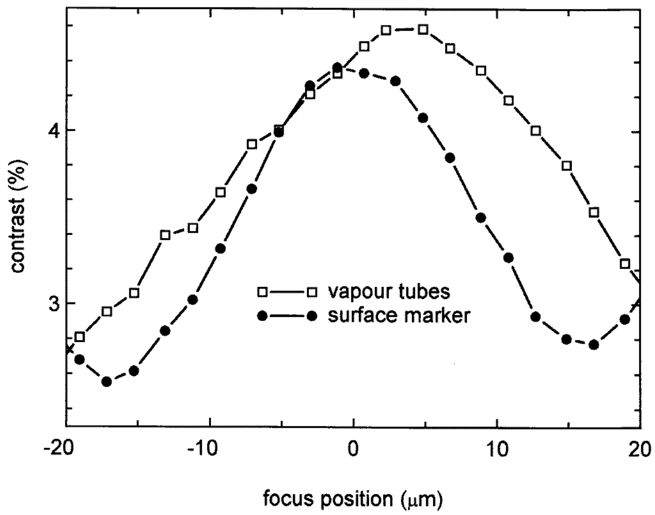

Figure 5. Contrast measurements of a surface marker (dust grain) on the fluid side of the heated window (dots) and of the vapour tubes in the boundary layer of critical xenon (squares) as a function of the focus position at $\Delta T_c = -0.5$ mK and -5 mm below the meniscus. Parabola fits to the contrast maxima result in a displacement of 2.5 μm between the centre of the vapour tubes and the surface of the window.

dissolve just below the position of the meniscus. At temperatures $\Delta T_c < -3$ mK only bubbles with a diameter O(10 μm) form in the boundary layer. In the region above the position of the meniscus no tube-like structures are observed.

4. Interpretation

The pattern formation of the kind described here has been observed, though with less optical resolution, also in critical argon and oxygen [3]. A similar effect was reported in investigations of the boundary layer in critical CO_2 [6] but was not followed up then. Since the effect only appears in a very small temperature range around T_c, it is attributed to the critical properties of fluids.

A crucial point in the interpretation is the temperature of the boundary layer. It is expected to attain its highest value on the surface of the heated window and to decrease towards the interior of the cell due to convection and thermal diffusion until it reaches the bulk fluid value on the scale of the thickness of the boundary layer. From the observed disintegration of the structures in small bubbles at $T_{fl} < T_c$ (compare figure 2(*d*)) it is concluded that also the temperature in the boundary layer is below T_c. With increasing bulk fluid temperature only the image contrast of the structures decreases while their characteristic properties remain about the same. Since this is valid for bulk fluid temperatures up to $\Delta T_c = 1.5$ mK, it is concluded that even in this small temperature interval above T_c the temperature in the boundary layer still remains at values below T_c.

This conclusion is supported by recent numerical simulations by Zappoli *et al* [7] on buoyancy-driven convection after application of a thermal step to a vertical plate in a near-critical fluid. Although these calculations were carried out for a thermally insulated container, they demonstrate that in contrast to normal fluid behaviour the temperature within the boundary layer can reach values below the temperature of the bulk fluid. This surprising effect is related to the so-called adiabatic or piston effect [8] which first was shown to be of importance for the fast temperature equilibration in critical fluids under microgravity. The calculated temperature drop in the boundary layer relative to the bulk fluid temperature is found to be O(10 μK) at $\Delta T_c = 1$ K, but is expected to increase on approaching the critical point [9].

In the present experiment the heating of the cell window creates a stationary and localized vertical temperature gradient. Volume elements of the fluid are transported via convection through this zone at a velocity O(100 μm s^{-1}), i.e. in times O(1 s) which is comparable to the time-scale for the formation of the temperature drop in the boundary layer found by Zappoli *et al* [7]. Since the tube-like structures are observed to disappear at fluid temperatures $\Delta T_c > 1.5$ mK, it is concluded that the lowering of the temperature in the boundary layer relative to the bulk fluid is O(1 mK).

The temperature drop in the boundary layer leads to spinodal decomposition of the fluid triggered by the critical density fluctuations. This assumption is supported by the observed granular pattern (figure 2(*a*)) which shows stronger density fluctuations in the boundary layer than in the bulk fluid. The development of vapour domains in the liquid phase under the influence of the convective shear flow in the boundary layer eventually leads to the formation of vapour tubes. A related effect was recently reported for the spinodal decomposition of a polymer mixture [10] where a superimposed shear flow was observed to generate string-like structures (the 'string phase').

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