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Strange interface deformation pulses in the flow of layered liquids

By B. K. CHATTERJEE, M. H. ENGINEER†, B. ROY AND PRADIP ROY

Department of Physics, Bose Institute, Calcutta 700009, India

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The interface in acoustically driven layered liquids, in an enclosed geometry, was found to be unstable to the production of spatially localized, quasi-periodic pulses of deformation. The most interesting property of these pulses is that they propagate in a direction *opposite* to that of the axial streaming velocity of both liquids. Experimental data are presented describing aspects of the phenomenon which occur only when both the interfacial tension and the density difference are small.

1. Introduction

Properties of flows of layered liquids and the inherent instabilities of such flows are of much importance in the atmosphere, hydrosphere, and in the oil and chemical industries. Stratified fluids flowing at different speeds parallel to their separating interface are known to develop instabilities of the Kelvin–Helmholtz (KH) type (Helmholtz 1868; Kelvin 1910, p. 69; Chandrasekhar 1961, p. 481). The KH instability is, at low fluid velocities, manifested in the form of a crinkling of the interface which then develops into larger spatially periodic structures co-moving with the interface. This instability has been the subject of exhaustive experimental and theoretical investigations (Thorpe 1969, 1987).

In this paper we report experimental observations of a strange and interesting kind of KH instability, occurring in a narrow range of flow parameters, in gravitationally stratified, immiscible liquid layers. To the best of our knowledge, such an instability has not been observed before.

2. Experimental setup

In one set of experiments, a closed cylindrical glass tube with inner diameter 0.0342 m, length 0.17 m was used as the experimental container. The container was held with its axis horizontal and filled with roughly equal amounts of two immiscible liquids. A gold plated X-cut quartz crystal transducer (Valpey Fisher Division, USA) was mounted axially at one end of the tube. It was driven by a tunable RF oscillator of Hartley type. The crystal, which had a diameter of 0.0254 m, was edge-mounted in an air-backed-type brass housing. An effective diameter of 0.0185 m of its grounded surface was in contact with the experimental liquids. The RF was tuned to 4.98 MHz which was one of the resonant modes of the quartz crystal. The ultrasonic intensity was varied by changing the input RF power to the transducer.

† Author for communication.

When energized the crystal generates, due to the so-called 'quartz wind', a flow of the fluids in the tube.

Since we wished to drive the fluids in the forward direction a travelling-wave ultrasonic field was essential. Accordingly, to curtail reflection of the ultrasonic beam from the far end, polythene absorbers were always used as terminators. The absorber was, usually, a flat disc of diameter 0.034 m and thickness 0.006 m, although a conically tapered piece of polythene, especially designed to gradually match the various acoustic impedances (much like the tapered terminations used in microwave waveguides), was also used in an attempt to further minimize the amplitude of the reflected ultrasonic wave.

We chose an ultrasonic beam to generate the basic laminar flow whose instabilities we wished to examine in the nonlinear domain, so that, by utilizing the well-known 'quartz wind', we could establish a high-velocity flow with minimum turbulence. High-velocity flows were, of course, essential if we were to reach the nonlinear domain.

We now describe the main features of the flow pattern when only one liquid is present; this paragraph and the following one will be useful when we come to §3, where the results will be described. The liquid, driven forward by the 'quartz wind' turns around (in differing degrees) at each cross-section along the tube before returning to the generator end via the annular region surrounding the ultrasound beam (Bhadra & Roy 1975). In this simple situation the flow pattern is axially symmetric and the streaming velocity profile is given by (Piercy & Lamb 1954; see also Eckart 1948)

$$\left.\begin{aligned} u_z &= \frac{\alpha I_0 a^2}{c\eta}\left\{-\ln\left(\frac{a}{b}\right)+\frac{1}{2}\left(1-\frac{r^2}{a^2}\right)-\left(1-\frac{r^2}{b^2}\right)\left(1-\frac{a^2}{2b^2}\right)\right\} \quad \text{for} \quad r \leqslant a, \\ \text{and} \quad u_z &= \frac{\alpha I_0 a^2}{c\eta}\left\{-\ln\left(\frac{r}{b}\right)-\left(1-\frac{r^2}{b^2}\right)\left(1-\frac{a^2}{2b^2}\right)\right\} \quad \text{for} \quad r > a, \end{aligned}\right\} \tag{1}$$

where a and b are the effective radii of the crystal and the tube respectively, α and I_0 the ultrasound amplitude attenuation coefficient and the input ultrasound intensity ($I \approx I_0$ for small α), and c and η the ultrasound velocity and the dynamic viscosity of the medium, respectively.

The profiles of the individual streaming velocities will, of course, differ when two liquids are present. This can happen for various reasons, the two main ones being differences in the ultrasonic driving pressure gradients and in the viscosities. The velocity field is continuous everywhere across the interface, but its gradient is discontinuous there. This discontinuity causes the flowing interface to become unstable; the destabilization is countered by gravity and interfacial tension.

Experiments were performed with a number of liquids. The results presented here (see §3) are for a 0.2% solution of surfactant in distilled water, and an oil mixture whose density was adjusted to be very close to that of the surfactant solution. We prepared the oil solution by mixing carbon tetrachloride (density = 1610.94 kg/m^3) and petroleum ether (density = 647.98 kg/m^3) in suitable proportions. The density of this volatile oil mixture was deliberately kept slightly larger than that of surfactant solution. Accordingly, it constituted the bottom layer of our two-liquid system. By this simple strategy any change in its density due to unequal evaporation rates was prevented. The densities of the liquids were measured to within an accuracy of 0.1%. The velocities of sound in the surfactant solution and in the oil mixture were observed to be 1528 and 987 m/s respectively. The attenuation length and the velocity of sound in water, carbon tetrachloride and petroleum ether, at 4.98 MHz, are 1.84, 0.0757 and 0.269 m and 1494, 928 and 985 m/s respectively.

One of the important requirements for the observation (see §3 for a full description) of the strange interfacial waves, referred to in the title of our paper is to reduce the interfacial tension between the water and the oil solution to as small a value as possible. The interfacial tension was lowered to a value of about 0.002 N/m by using a 0.2% solution of Triton X-100 (Fluke Chemie) in distilled water. A negligible amount of blue, water-soluble (but oil-insoluble) dye was also added to the water, for clear identification of the liquids and the interface.

Transferring two liquids whose densities are so close and where, additionally, the interfacial tension has also been kept small, from their respective containers into the experimental glass tube is a tedious process. Not only does the inner surface of the tube have to be cleaned (by repeated washing with dilute HCl, water and ethanol), but also the bottom liquid has to be pipetted below the water with great caution. Additionally, it is necessary to allow a long settling time. The smallest difference in density (between the two fluids) which allowed formation of a stable horizontal interface (rather than one of the fluids rolling up into a ball) was about 5.0 kg/m^3. In a typical run, the densities of the surfactant solution and the oil mixture were found to be 1012.0 and 1021.0 kg/m^3 and their viscosities were measured (using an Ostwald viscometer at 26 °C) to be 0.000882 and 0.000440 N s/m^2 respectively. The interfacial tension value of 0.002 N/m was measured by the capillary rise method of one liquid on top of the other. Measurement of such small interfacial tension values is quite difficult, as is well known.

Other liquid systems like silicone oil (2 cS octamethyl cyclotetra siloxane from Metroark Pvt. Ltd, India) and a water–methanol mixture (methanol is insoluble in silicone fluid) were also employed to observe the strange waves. The viscosities of the silicone fluid and the water–methanol mixture were found to be 0.001883 and 0.001293 N s/m^2 at 26 °C. The densities were 974.0 and 995.0 kg/m^3 respectively.

The amplitudes of the strange waves were measured using a travelling microscope; their velocities were determined by noting the time of transit of the pulse crest (see §3) through a fixed distance, and their periodicity was recorded using a chart recorder.

3. Observations and results

We now summarize the main features of the experimental observations, for sufficiently low values of both the interfacial tension and the density difference only. At very low driving amplitudes the steady flow is locally laminar everywhere, the interface remaining absolutely flat despite the steady motion. This steady state persists till the strength of the driving reaches a critical value. Above this value the nature of the steady flow changes abruptly and a static crinkling of the smooth interface appears. The wavelength of the crinkles is around a few mm. At first the crinkling amplitude increases with increasing driving force but, then, a slowly undulating roll, whose half-wavelength is around the length of the tube, appears (the tube length was 0.17 m, but the behaviour of the roll near the ends could not readily be seen because of two brass flanges fitted there to hold the transducer and the terminator). Finally, above a second critical driving amplitude, a further transition to a new steady state occurs: large-amplitude, spatially localized pulses of interface deformation form and propagate in a direction *opposite* to the streaming velocities of

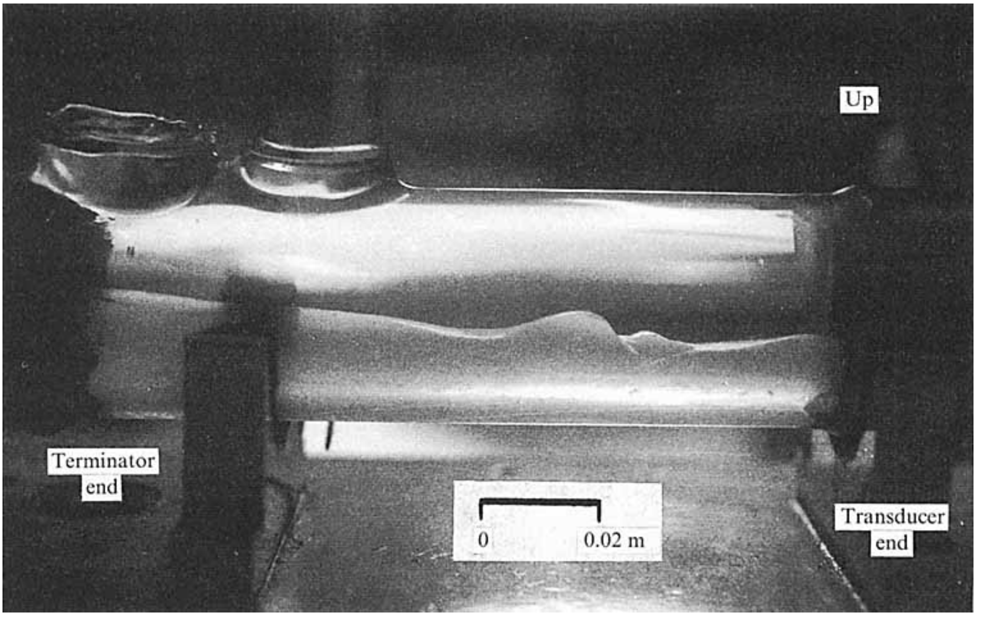

FIGURE 1. Photograph showing (transverse) profile of a CID pulse generated in an experimental tube (length 0.17 m, diameter 0.0342 m). Top liquid, 0.2% water–surfactant mixture (density 1012.0 kg/m^3); bottom liquid (density 1021.0 kg/m^3), a mixture of CCl_4 and petroleum ether. An ultrasonic source (frequency = 4.98 MHz) mounted at right, and a reflectionless polythene terminator at the left end of the tube are shown. Bulk flow is from right to left; the CID pulse travels in the opposite direction.

Amplitude	0.0032 ± 0.0009 m	($360 < I < 800$ W/m^2)
Frequency	0.26 ± 0.005 Hz	($I < 520$ W/m^2)
	0.38 Hz	($I = 660$ W/m^2)
Velocity	0.01237 m/s	($I > 450$ W/m^2)

TABLE 1. Characteristic values for the CID pulses in between the layers of 0.2 % surfactant solution in water and CCl_4–petroleum ether oil mixtures for various acoustic intensities (I). The density and viscosity of the surfactant solution are 1012.0 kg/m^3 and 0.000882 N s/m^2, and of the oil mixture are 1021.0 kg/m^3 and 0.00044 N s/m^2 respectively. The interfacial tension was 0.002 N/m.

the two liquids. These counter-propagating interface deformation (CID) pulses (an example is shown in figure 1) are generated at periodic intervals; the frequency of their generation is given in table 1.

The main features of each individual pulse are now briefly summarized. Viewed from upstream (which is the direction in which the pulse travels) each pulse consists of a small hump, followed by a small trough and, finally, a large hump. The humps always point from the less to the more viscous fluid; in the present case, since the oil mixture which has the smaller viscosity, that forms the bottom layer of the fluid pair, we have used the word hump. If (by adding a tiny amount of petroleum ether) we put the oil mixture on the top of the water the hump would have a negative amplitude, i.e. we would have to call it a trough. This curious fact – for which we have no explanation – has actually been observed.

We estimate that when the CID pulses first appear, i.e. at threshold, the effective input ultrasonic intensity (I_0) is approximately 310 W/m^2. It is worth emphasizing

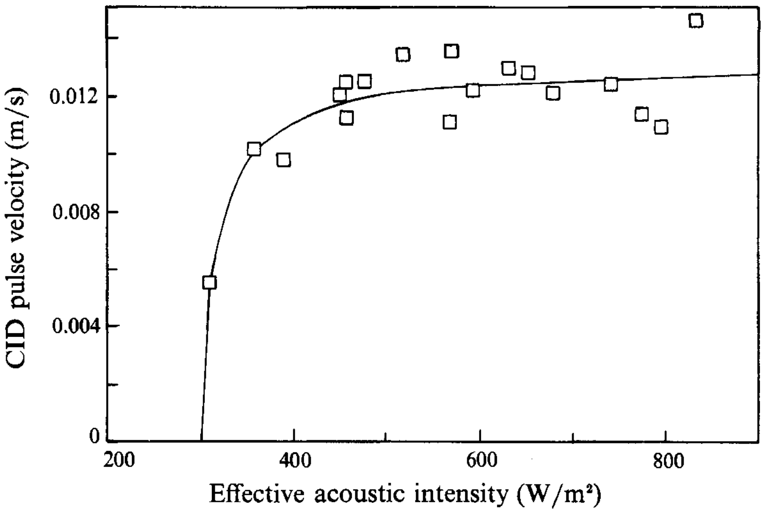

FIGURE 2. The variation of the observed group velocity (U) of the CID pulses as a function of the acoustic intensity (I). The solid line $U = 0.0131((I-303)/(I-269))^{0.534}$ m/s indicates the least-squares fit to the observed data.

that this effective intensity is an average, over near-field fluctuations, of the ultrasonic intensity present in the tube. We arrived at this number by a separate experiment where the tube was filled entirely with water, all else remaining unchanged, and the variation of the streaming velocity with the DC voltage applied to the RF generator was determined by timing the rectilinear motions of fine suspended particles in the liquid. From these measured values of streaming velocity in water the ultrasonic intensity was calculated using equation (1). Typically a streaming speed value of 0.00988 ± 0.0061 m/s yields $I_0 = 979.68$ W/m^2; at this speed the flow was observed to be laminar.

The pulses could be observed up to effective intensities of about 830 W/m^2; beyond that the flow became so violent that droplets of one liquid were thrown into the other and the pulses disappeared completely. The frequency and amplitude of these pulses were observed to have little dependence, if at all, on the effective ultrasonic intensity (see table 1). The speed of the pulses is, however, intensity dependent; observations indicate a levelling off of the speed beyond an effective intensity of around 450 W/m^2 (see figure 2) (hereafter all ultrasonic intensities are *effective*).

In general CID pulses propagate within an interval that extends from about 0.01 m to about 0.12 m (as measured from the terminator end of the experimental tube). As can be seen from figure 3, at relatively low acoustic intensity values the moving CID pulses appear to vanish within a few cm of their starting positions. However, a short while later, they renew themselves a short distance upstream of the apparent vanishing point and travel further upstream for quite a distance before, finally, being destroyed. Remarkably, the time interval between their first disappearance and the subsequent reappearance was observed to be quite close to the time it would have taken the original pulse to traverse the distance involved; this suggests that we are observing one and the same disturbance. Observations at somewhat higher intensities (see figure 3) seem to confirm this suggestion.

At even higher intensities each CID pulse gets sharper, often sending finger-like projections against the streaming flow, i.e. the instantaneous wave profile is typically

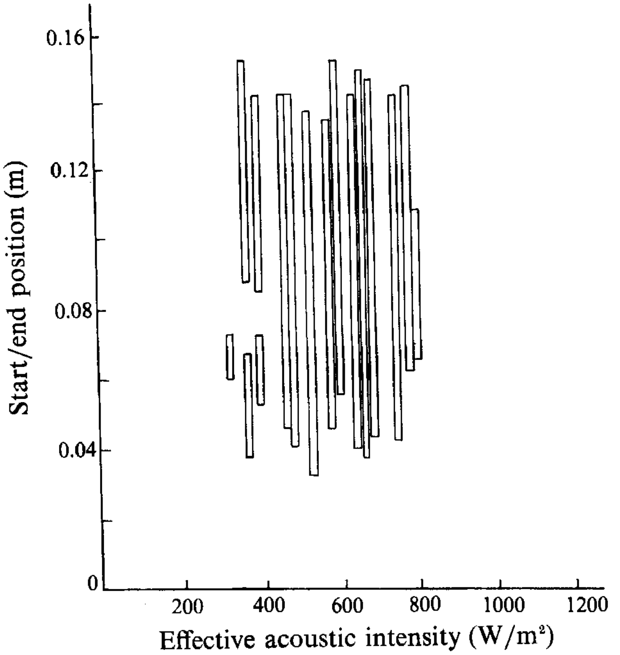

FIGURE 3. Region, along the tube length, within which the CID pulses were observed to propagate, versus acoustic intensity. The origin of the ordinate axis refers to the terminator end of the tube.

triple-valued. Droplets split off at random from these finger-like projections and are carried away by the fluid current. At even higher acoustic intensities the flow becomes extremely turbulent, forming a froth of droplets of one liquid in the other.

The data presented so far refer to the 0.17 m long tube. Though CID pulses were observed for differences in liquid densities smaller than 9 kg/m^3, none were observed for differences greater than 20 kg/m^3, at an interfacial tension of 0.002 N/m. At interfacial tensions higher than 0.002 N/m the heights of the pulses become vanishingly small.

Besides using the 0.17 m long cylindrical container, experiments were also performed in two other cylindrical containers, of lengths 0.346 and 0.637 m (the tube diameter was not changed). In the 0.346 m tube, CID pulses were observed to originate from two distinct places, one near the far end of the tube, and the other around its middle. In the case of the 0.637 m tube, a single stationary hump of interfacial deformation was first observed to form; upon increasing the driving intensity the hump, while remaining stationary, deformed even further, developing into a finger about a centimetre long (of the more-dense fluid enveloped within the less-dense one). This finger pointed towards the driving crystal. The finger itself was barely stable; it writhed about incessantly, and occasionally droplets torn from its tip would be convected away by the forward motion of the upper fluid (water).

Beautiful CID pulses were observed in two rectangular tank geometries (of inner dimensions 0.45 × 0.106 × 0.127 m and 0.445 × 0.127 × 0.127 m). In one experiment the liquids used were a CCl_4–petroleum ether mixture and water, and in the second a methanol–water solution and silicone fluid were used. Of course, in both cases a 0.2 % water-soluble surfactant solution was added to reduce the interfacial tension. The acoustic driving power was provided by a similar X-cut quartz crystal housed in a watertight housing immersed in the liquids.

The quantitative results for the liquid system consisting of the surfactant solution and CCl_4–petroleum ether solution have been presented. For this particular liquid system the average amplitude, velocity and frequency of the CID pulses in the given range of acoustic power are shown in table 1, while the variation of the velocity of the CID pulses with acoustic intensity can be seen in figure 2.

We turn, finally, to the results of one more small experiment, undertaken in order to throw light on the mechanism of generation (not propagation) of the CID pulses. In this case, the earlier experiment using a 0.17 m tube was repeated, but this time a thin-walled inner glass tube of shorter length (0.081 m), open at both ends, was placed coaxially within the larger-diameter tube. The diameter of the inner tube was just equal to the diameter of the exposed face of the quartz crystal, and one end of it was placed as close as possible to the crystal. In this Poiseuille type of situation (so far as the flow inside the inner tube is concerned) it was observed that CID pulses propagate *within* the inner tube only, although the interface was present in the annular space as well. Furthermore, in this geometry the generation of the pulses takes place beyond the far end of the inner tube, at the point where both fluids turn to begin their return journey, through the annulus.

4. Conclusions

The main conclusion of the present work is that large pulses of interfacial deformation can propagate in stratified fluid flows. While the theory behind the phenomenon remains obscure, its universal nature seems reasonably clear from the facts that the pulsed excitations are (*a*) geometry independent and (*b*) found to occur for various pairs of immiscible fluids. It is also a remarkable fact that the shapes and motion of the pulses generated are, qualitatively identical – observed from the generator end a small hump, followed by a shallow trough and then a large hump, moves against the main direction of the bulk flow. Finally, the humps always extend from the less- into the more-viscous fluid. A theoretical explanation of the phenomenon would be a most welcome indicator of the path that future experiments in this exciting new field needs to take.

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