On dispersion relation for Faraday waves in a near-critical fluid under weightlessness

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Abstract. One reports experiments and numerical simulations of Faraday waves at the liquid - vapor interface of fluids (typically CO₂ and H₂) near its critical point when submitted to vibration in a weightlessness environment. It is known that in such a system striation is formed under the action of vibrations. The boundary between the liquid and gas phases is located perpendicular to the direction of vibrations, and a streaky structure in the form of periodic stripes (striations) is formed in the cavity. It is shown that under vibrations of sufficient intensity an instability develops on the striation interface perpendicular to the direction of vibrations, leading to Faraday waves. A theoretical and numerical study of this instability is carried out, the critical parameters of the appearance of Faraday waves are determined taking into account the interaction of waves on adjacent striations. Dispersion relations for Faraday waves are obtained numerically. It is shown that in the vicinity of the critical point there is a significant deviation of the dispersion relation from the classical theoretical dependence.

1. Introduction

In the space environment gravity effects are not present and a number of phenomena, of marginal effect on earth, can become predominant. The use of fluids in the vicinity of their liquid-vapor critical point is particularly appealing to carry out such investigations. In addition to its use by the space industry near such conditions ("supercritical state"), the thermodynamic properties of fluids when expressed under reduced temperature or density distance from the critical point can be conveyed under universal, scaled functions [1]. Using such scaling properties, studying one fluid permits to study all due to the possibility to vary the fluid parameters simply by changing temperature.

Under vibration, the shape and stability of the interface between two fluids depend on the relative directions of vibration and interface. When the vibration direction is parallel to the fluid interface, quasi-stationary wave patterns (frozen wave instability [2–4]) can occur, caused by a shear-driven mechanism similar to the Kelvin-Helmholtz (K-H) instability. The interface deformation is immobile in average in the
reference frame of oscillating boundaries. When vibration is directed perpendicularly to the interface, its parametric excitation, the Faraday wave instability [5–7], follows. Typically, the fluid (here CO₂ or H₂) is submitted to harmonic vibrations and the relative distance to the critical temperature

$$\varepsilon = \frac{T - T_c}{T_c}$$

is varied ($T$ is temperature, $T_c$ is critical temperature). The liquid vapor interface is seen to successively exhibit the following behavior as $\sigma$ and $\Delta \rho$ diminishes with $\varepsilon$.

(i) $\varepsilon < 3.3 \times 10^{-3}$: The vapor bubble oscillates, with a number of modes that increases when $\varepsilon$ decreases;

(ii) $10^{-3} < \varepsilon < 3.3 \times 10^{-3}$: The bubble shows a flat interface perpendicular to vibration [6];

(iii) $3 \times 10^{-4} < \varepsilon < 10^{-3}$: Faraday waves (square pattern) develop on the liquid-vapor interface, whose wavelength decreases when $\varepsilon$ decreases [6];

(iv) $7 \times 10^{-5} < \varepsilon < 2 \times 10^{-4}$: The Faraday waves square pattern becomes a two-dimensional (2D) roll pattern due to the small wavelengths which increase dissipation [5];

(v) $\varepsilon < 7 \times 10^{-5}$: The liquid vapor interface orders in parallel bands perpendicular to vibrations in a way reminiscent of the earth-bound frozen wave instability where the interface deformation is immobile in the reference frame of the oscillating boundaries.

In the band regime (v), Faraday waves can also be observed on the band interfaces (which are perpendicular to the vibration direction). The Faraday instability corresponds to the Faraday roll pattern on the single liquid-vapor interface as observed in (iv). There is, however, a noticeable difference as the band thickness is much smaller than the sample size. Such band patterns have also been noted under weightlessness with miscible liquid mixtures just after they were put into contact, and the same kind of Faraday waves were also observed. In [7] theoretical and numerical investigations of the Faraday waves were carried out and the critical parameter for the onset of instability were derived taking into account the interaction between the bands.

The purpose of this paper is to analyze Faraday-on-bands dynamics from direct two-dimensional numerical simulation and experimental data collected with CO₂ under weightlessness as provided by the sounding rocket Maxus 7 [8] and with H₂ under magnetic levitation [6].

2. Theoretical background

The Faraday wave instability under a gravity field can be modeled as two layers of immiscible and incompressible viscous fluids subjected to vibration perpendicular to the interface. The stability problem derived from the Navier-Stokes equations was treated by Kumar and Tuckerman [9] using a Floquet analysis. They carried out numerical calculations based on the linearized Navier-Stokes equations and analytical calculations by introducing the viscosity in a phenomenological way into the Mathieu equation for the low viscosity case. The dispersion relation in the low viscosity case was given by:

$$\left(\frac{\omega}{2}\right)^2 = \left(\frac{\rho_1 - \rho_2}{\rho_1 + \rho_2}\right) g k + \frac{\sigma k^3}{\rho_1 + \rho_2} - 4k^4 \left(\frac{\eta_1 + \eta_2}{\rho_1 + \rho_2}\right)^2$$

where $\omega$ is the vibration angular frequency $\eta_1$ and $\eta_2$ are the dynamic viscosities of fluids “1” and “2” respectively and $\sigma$ is liquid vapor surface tension. The latter goes to zero with $\varepsilon$ as

$$\sigma = \sigma_0 \varepsilon^{2\nu}$$

Here $\nu = 0.64$ is a critical exponent and $\sigma_0$ is a fluid dependent amplitude.
For the inviscid case ($\eta_1 = \eta_2 = 0$) it coincides with the dispersion relation for gravity-capillary waves. The threshold amplitude for non-viscous case is zero. For low viscosity fluids (when the dynamic boundary layer $\delta = \sqrt{2v/\omega}$ near the interface can be considered small), the rigorous linear stability analysis performed by Lyubimov et al. gives for the stability curve $a_c(\omega)$, with $a = a_c$ the vibration amplitude at the instability threshold:

$$a_c = \frac{\rho_1 \rho_2}{(\rho_2 - \rho_1)(\rho_2 \sqrt{\nu_1} + \rho_1 \sqrt{\nu_2})} \sqrt{\frac{2\nu_2}{\omega}}$$

Here $\nu_{1,2} = \eta_{1,2}/\rho_{1,2}$ is the kinematic viscosity of fluids 1, 2. The dispersion relation for zero-gravity conditions in low viscosity case is given by Eq. (2) where one makes $g = 0$ can be written as

$$\left(\frac{\omega}{2}\right)^2 = \frac{\sigma k^3}{\rho_1 + \rho_2} - 4k^4 \left(\frac{\eta_1 + \eta_2}{\rho_1 + \rho_2}\right)^2$$

3. Experiments
The details on the experimental setup can be found in the work [8]. The experimental module is of TEM-FER type. The thermostat is an Al cylinder in which is inserted the CuCoBe block. It is thermally regulated by a Peltier element within 0.3 mK accuracy. Temperature can be adjusted within 1mK steps. As the module has to work under vacuum during the flight, the thermostat remains pressurized at 1 bar pressure. The cell body is made of a block of copper-cobalt-beryllium alloy in which are drilled one cylinder with radius $R = 5.0$ mm and thickness $e = 2.189$ mm, closed at each end by two parallel sapphire windows. Using CuCoBe alloy ensures good thermal conduction between the fluid and the thermostat. The sample is illuminated by parallel light issued from a LED and observed in transmission by two cameras. Coherent illumination is used to enhance the refractive-index gradients and then the density gradients. The cell is filled at critical density with a precision of 0.1%, by checking the temperature variation of the meniscus position on ground.

4. Numerical simulations
Direct numerical simulations (DNS) were performed using the Volume of Fluid (VoF) method describing the dynamics of the interface of immiscible fluids. The method is based on the introduction of a volume fraction function for each phase. The function is constant inside a selected phase and exhibits sharply changes at the interface between phases. A multiphase system can thus be treated as a single medium with sharply changing parameters at the interfaces. The physical properties of this phase are calculated in any point by the values of the volume fractions. The Navier-Stokes equations describing the system can be solved over the entire computational domain. The VoF method is implemented in ANSYS Fluent used for modeling the pattern formation.

| Table 1. Fluid parameters |
|---------------------------|
| Fluid | $T_c$ (K) | $p_c$ (MPa) | $\rho_c$ (kg.m$^{-3}$) | $B$ | $\sigma_0$ (10$^{-2}$ N.m$^{-1}$) | $\eta$ (10$^{-3}$ Pa.s$^{-1}$) |
|------|--------|--------|----------------|------|----------------|----------------|
| C0$_2$ | 304.14 | 7.375 | 467.8 | 1.60 | 6.72 | 4.21 |
| n-H$_2$ | 33.19 | 1.315 | 30.11 | 1.61 | 0.542 | 0.27 |
The influence of vibrations is taken into account by introducing a periodically varying volumetric force. Temporal discretization is performed by the first-order scheme. Note that the time step has to be much smaller than the period of vibrations while the frequency of vibrations is quite high. The computational error of the first-order scheme is therefore small. The spatial discretization is performed using the third-order MUSCL scheme (Monotone Upstream-Centered Schemes for Conservation Laws). Fluid parameters used in calculations are shown in Table 1.

![Figure 1. Faraday waves for $f = 5.5$ Hz, $a = 2.5$ mm](image)

Under vibration of sufficient intensity on the interface’s transverse to the direction of vibrations, the development of instability is observed, leading to the appearance of Faraday waves. The example of such waves developing on band pattern is shown in Figure 1. The extensive theoretical and experimental analysis of this instability is performed in [7].

5. Results and discussion
The viscosities of liquid and vapor phases in near critical fluids are low. Besides, when approaching the critical point, the viscosities remain nearly constant while the surface tension decreases drastically according to Equation (3). As a result, the contribution of the negative second term in Equation (5) becomes more and more important in comparison with the first term. The frequency of Faraday waves thus tends to zero when $T$ tends to $T_c$. In the close vicinity of $T_c$, $\omega$ should even become negative. When using the parameters of the CO$_2$ experiment (Table 1) the frequency remains positive only at small enough values of $k$, a behavior which, however, is not detected in the experiments. This means that close to $T_c$ the viscosity cannot be treated as a perturbation and the low viscosity assumption of Eq. (5) is no more valid.

The comparison of the experimental and numerical results for CO$_2$ with the dispersion relations for inviscid fluid and low viscous fluids is shown in Fig. 2. As one can see, experimental and numerical data correspond to much higher values of $\omega$. Thus, we can conclude that for the case of low surface tension interfaces, the range of the validity of the dispersion relation Equation (5) is very narrow. In order to verify this conclusion, we performed numerical simulations for parameters corresponding to CO$_2$ but with viscosity 10 times lower. In Fig. 2 are reported the results of these calculation. Data are in a very good agreement with the dispersion relation Equation (5). It is worthy to note that while the dispersion relation Equation 5 and DNS at low viscosity give $\omega$ values smaller than the values given by the dispersion relation for the inviscid case, experiments and DNS at larger viscosity exhibit much larger values. The influence of viscosity on the dispersion relation is thus quite important and a specific approach has to be considered, which nevertheless goes beyond the scope of the present work.
In previous experiments by Gandikota et al. [7] Faraday waves were studied on a single interface in a two-phase near-critical H$_2$ under weightlessness. It was found that the experimental results for the square patterns were in good agreement with the dispersion relation for the zero-gravity case Equation (5). Marked deviations from the dispersion relation were, however, observed for temperatures very close to the critical point, where the transition from square patterns to line patterns occurs, similar to what was observed under 1-g conditions. In addition, a saturation of the wavelength to a finite value was detected at small $\varepsilon$. Deviations to Equation (5) and saturation of the wavelength were attributed to the increasing viscous dissipation due to the decrease of the pattern wavelength (see below). It has been observed that a pattern with parallel lines can be also obtained with ordinary liquids in the capillary-wave regime if the dissipation is large enough [10]. In addition, a wavelength saturation can be understood as a viscous cutoff length for which the surface wave is too strongly damped to be parametrically amplified [5].

Thus, it is possible to expect that Faraday waves developing on band interfaces very near to $T_c$, which are also line patterns, show substantial viscous dissipation. The viscous dissipation expressed per unit volume between two adjacent interface modulation (wavelength $\lambda$) can be written to be proportional to $\eta u^2/f\lambda^2$, with $u=a\omega$ the velocity of the fingers equal for both phases close to the critical point. The viscous dissipation term is thus on order of $\eta(a\omega)^2/f\lambda^2$ and increases as $a\omega$ is increased or $\lambda$ is decreased, $\eta$ being almost independent of the temperature near the critical point. The vibrational energy term per unit volume is proportional to $\Delta\rho(a\omega)^2$ and decreases as $T_c$ is approached. Following Gandikota et al. [6], a non-dimensional number $V$ can thus be defined as the ratio of the viscous dissipation energy to the vibrational energy to describe the relative influence of dissipation. It was seen that the threshold value of $V$ for the transition square-line was around 0.1, corresponding for H$_2$ to $\varepsilon=2\times10^{-4}$ [6] and for CO$_2$ to $\varepsilon=7\times10^{-5}$ [5], values below which the band pattern indeed appears [6].

6. Conclusion
Faraday instabilities in a pattern of narrow band interfaces perpendicular to vibrations were observed in CO$_2$ under weightlessness conditions and studied numerically and theoretically. For the dispersion relation, the classical theory could not fit both the experimental and simulation data - although the latter, however, show agreement with each other. The reason is the presence of the increasing dissipation very
near the critical point due to the pronounced pattern wavelength decrease, that theory which assumes weak
dissipation can not describe. Further theoretical investigations are planned to clarify these issues.

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