

Stratification effects in fluids near their liquid-vapor critical point

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Near-critical fluids are known to exhibit anomalous behavior in their thermophysical properties, such as diverging compressibility and vanishing surface tension on approaching the critical point. These fluids undergo high-density stratification very close to the critical point. The Navier-Stokes equations coupled to a phase field model are solved with a van der Waals equation of state. The pressure is expanded around the critical point and shows a cubic variation with the reduced density. Density-stratified base states are computed numerically. The supercritical case is treated through the linear stability analysis. The spectrum, the eigenmodes/eigenfunctions, and sound waves are then computed and plotted for temperatures close to the critical point (CP). Very close to CP, the speed of sound significantly changes as a function of the vertical direction, and the frequency bands strongly differ from the situation of an ideal gas. The same process of linearization has been performed for the subcritical state. Here, the Rayleigh-Taylor instability (RTI) is considered. Finally, numerical simulations confirm the propagation of acoustic waves in the supercritical case. In the subcritical case, spinodal decomposition is found for a randomly distributed initial density close to the critical one. Time series in the Rayleigh-Taylor unstable domain show interesting features. The time evolution of the total kinetic energy exhibits three distinct phases: high-frequency sound waves appear at early times, then RTI manifests, and finally, at longer times, surface waves appear with a much smaller frequency.

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I. INTRODUCTION

Near-critical fluids are a class of fluids that exist beyond and below the liquid-vapor critical point. At the supercritical state, they may be considered as an intermediate state between a liquid and a gas. They exhibit some properties of liquids, like high density and solubility, and some properties of gases, such as low viscosity and high compressibility [1]. On approaching the critical point, the various thermophysical properties show a singular behavior such as diverging isothermal compressibility, heat conductivity, specific heat at constant pressure and constant volume, and a vanishing thermal diffusivity [2,3]. These properties have been found to behave according to the

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universal power laws with a different critical exponent for each property. They can be modeled on the basis of a common parameter $\varepsilon = (T_i - T_c)/T_c$, where T_c refers to the critical temperature and T_i is the initial temperature of the fluid. The most unusual behavior (and new at that time) of near-critical fluids was observed by Nitsche and Straub [4] during the measurement of the heat capacity at constant volume in their sounding rocket experiment. They observed that the thermalization of their cell was extremely fast despite a vanishing thermal diffusivity. Three worldwide independent teams [5–7] have explored this unusual phenomenon and explained that the critical acceleration of heating is due to the piston effect, attributed to the high compressibility and thermal expansion near the critical point. A new timescale, called the piston-effect timescale, has been defined, which vanishes at the critical point, and the tiny remaining thermal nonhomogeneity is achieved by the very slow heat diffusion (the thermal diffusivity vanishes near the critical point).

Internal waves are gravity-driven oscillations that occur within the interior of a fluid, typically at the interface between layers of different densities. These waves are ubiquitous in geophysical and astrophysical contexts, such as in atmospheres, oceans, lakes, stellar interiors, and even the air in buildings naturally stratified by heat or accidentally stratified by fires [8]. They play a significant role in mixing and energy transport processes [9,10]. In a more controlled laboratory setting, internal waves can also be generated in near-critical fluids where the fluid’s compressibility and thermal expansion coefficient become anomalously large. For xenon in 1 g, for example, the density varies by as much as 10% in 1 cm [11]. In this context, the study of internal waves in fluids near their critical point provides a unique window into the interplay between thermodynamics and fluid dynamics under extreme conditions.

Near the critical point, the thermodynamic and transport properties of fluids exhibit strong fluctuations and divergences, which substantially modify wave dynamics [12]. For instance, the density stratification in near-critical fluids can be driven not only by temperature gradients but also by small pressure variations due to the enhanced compressibility. As a result, the internal wave behavior in such fluids differs markedly from classical Boussinesq approximations typically used in stably stratified fluids. This makes the study of internal waves in near-critical fluids a fertile ground for understanding non-Boussinesq effects coupled with critical phenomena in hydrodynamics. Moreover, it can be ascertained that there is an imminent need to strengthen the competencies in super/subcritical fluids phenomenon for scientific advancements and the socioeconomic-environmental impacts. Their presence in key areas is very active, such as global energy needs [supercritical CO₂ (scCO₂)]-based power plants and refrigeration systems), tackling climate change (geothermal storage of CO₂, hydrogen), greener chemical processes (using scCO₂), waste treatment and recycling. These applications necessitate addressing fundamental questions related to the behavior near the critical point and developing advanced simulation tools to better understand the underlying physical processes.

The key challenge is to capture phase transitions, besides the usual monophasic/diphase behavior, in a single unified model within the sphere of continuum fluid mechanics. The phase-field method is a computational approach used to model and simulate the evolution of interfaces and phase transitions in complex systems. Instead of explicitly tracking sharp boundaries between different phases, it introduces a continuous field variable (called the phase field) that smoothly varies across interfaces, distinguishing one phase from another. This method is particularly well-suited for problems involving moving boundaries, such as solidification, multiphase fluid flows, and microstructure evolution, because it naturally handles changes in topology like merging or splitting of interfaces. The governing equations are typically derived from thermodynamic principles, where the system’s free energy is minimized, leading to partial differential equations that describe the dynamics of the phase field. As a result, the phase-field method provides a flexible and robust framework for capturing complex interfacial phenomena without the need for explicit interface tracking. The salient feature of this method lies in defining the interface as a thermodynamic region, contributing to the free energy through interface surface energy (the interface region being obtained by minimization of the free energy [13,14]). This ensures that if

the current state lies in the unstable two-phase region, the fluids separate into two phases. This unique feature renders the model an excellent choice for the problem at hand. Further, as the interface region is defined to have a finite thickness, the current method is coherent with physical characteristics near the critical point as the interface thickness diverges on approaching the critical point [15,16].

Though the literature is abundant with studies using this method, primarily focused on incompressible flows, the goal of capturing these phase transitions in a compressible environment is yet to be achieved. In recent experiments at the National Institute for Standards and Technology (NIST), Berg *et al.* [11] studied internal gravity waves in near-critical xenon contained in a small cell with an immersed apparatus configured as a viscometer. Using an inviscid incompressible fluid model, they calculated the frequencies of the internal-wave motion and compared these with their experimental results. Later, Anderson and McFadden [17] proposed a model based on a thermodynamic formulation that included contributions from a gradient-energy term that led to localized but smooth transitions at phase boundaries. The gradient energy term leads to a capillary tensor first suggested by Korteweg [18]. This tensor appears in the momentum equation, making local contributions in the transition layer that represents the diffuse liquid-vapor interface. Carlès and El Khouri [19] performed a theoretical analysis of internal gravity waves in near-critical fluids. They have shown that incompressible equations are applicable even very close to the critical point, and that the piston effect and internal gravity waves never couple, unlike what happens in the case of forced oscillations. They concluded the relevance of using near-critical fluids to simulate internal gravity waves in large geophysical systems.

Rayleigh-Taylor instability (RTI) occurs at the interface between two fluids when subjected to a gravitational field directed from the denser fluid toward the less dense fluid. The incompressible RTI has been deeply studied in the literature with Chandrasekhar's pioneering work [20]. There is much less work in compressible RTI. Kull [21] has treated fluid systems describing inhomogeneous, viscous, compressible, and isobaric flows. He addressed the stability analysis in layered materials applied to viscous boundary layers, compressible atmospheres, and stationary ablation fronts in laser-driven plasma experiments. Peng *et al.* [22] have studied the collapse of compressed bubbles in cryogenic liquids through RTI. They have shown that the bubble is most unstable when it collapses moderately, whereas it is stabilized as the collapse intensity is strengthened. In [23], Lai *et al.* investigated the effects of compressibility on RTI by inspecting the interplay between thermodynamic and hydrodynamic nonequilibrium phenomena via a discrete Boltzmann model (DBM). They have shown that the compressibility delays the initial stage of RTI and accelerates the later stage. To the best of our knowledge, the only study near the critical point of the Rayleigh-Taylor-like instability (RTL) has been treated through the density stratification due to the diverging compressibility [24,25]. This instability in a one-phase fluid seems to be similar to that which occurs between two miscible liquids, where the species diffusion is replaced by the heat diffusion. They found that this RTL configuration becomes stable when the heat diffusion length on the timescale of the faster unstable mode becomes larger than the bottom hot layer thickness.

In this work, we present the Navier-Stokes equations coupled to a phase field model. To describe the dependence of the pressure on density and temperature qualitatively, a van der Waals equation of state is applied. We perform a linear stability analysis and determine the eigenmodes and the corresponding spectrum. The speed of sound, the frequency band, and the eigenfunctions are determined as a function of the proximity to the critical point. The analysis is performed above and below the critical point. In the second part, the Rayleigh-Taylor instability very close and below the critical point is considered. Results from linear stability analysis are compared with the case of incompressible fluids and also with direct numerical simulations.

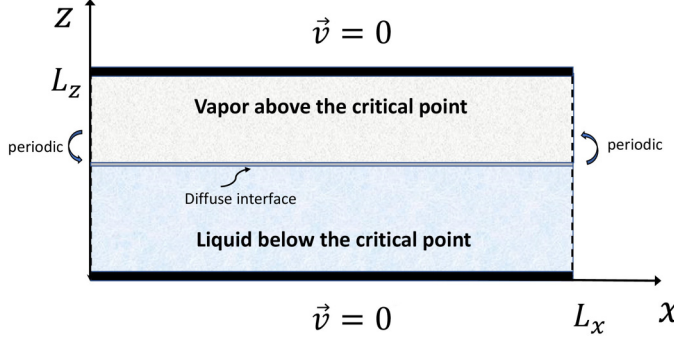


FIG. 1. The two-dimensional fluid layer.

II. THE THEORETICAL MODEL

A. The Navier-Stokes-Korteweg equations

The compressible Navier-Stokes-Korteweg equations (NSKE) in two spatial dimensions (x, z) can be written as [26,27]

$$\rho \frac{d\vec{v}}{dt} = \eta (\nabla^2 \vec{v} + \nabla(\nabla \cdot \vec{v})) + \left(\zeta - \frac{2}{3} \eta \right) \nabla(\nabla \cdot \vec{v}) - \nabla P - \rho g \hat{e}_z + K \rho \nabla \nabla^2 \rho, \quad (1a)$$

$$\frac{d\rho}{dt} = -\rho \nabla \cdot \vec{v}, \quad (1b)$$

where ρ is the density, $\vec{v} = (u, w)$ is the two-dimensional velocity, $P(\rho, T)$ is the thermodynamic pressure, T is the temperature, η , ζ are the shear and volume or bulk viscosities, and g is the gravitational acceleration. The last term on the right-hand side is the Korteweg stress with K representing the gradient energy coefficient [17,18,27] that is related to the surface tension, see Appendix A 3, Eq. (A14). For isothermal systems, T is a control parameter, and the density ρ can be considered as the phase field.

Equations (1) have to be completed by appropriate boundary conditions (see Fig. 1). At $z = 0, L_z$, we assume

$$\vec{v} = 0, \quad \partial_z \rho = 0. \quad (2)$$

At $x = 0, L_x$, we take periodic continuation in the horizontal direction of all variables.

B. van der Waals equation of state

The relation between pressure, density, and temperature is expressed by an equation of state (EOS). The van der Waals EOS shows the features of a phase transition and the existence of a critical point. It reads

$$P(\tilde{\rho}, \tilde{T}) = P_c \left(\frac{8\tilde{T}\tilde{\rho}}{3 - \tilde{\rho}} - 3\tilde{\rho}^2 \right), \quad (3)$$

with the nondimensional variables defined as

$$\tilde{\rho} = \rho/\rho_c, \quad \tilde{T} = T/T_c,$$

and

$$P_c = \frac{3}{8} R^* T_c \rho_c.$$

Here, $R^* = R/M$ is the specific gas constant with the universal gas constant R and the molar mass M . The critical pressure, temperature, and density are denoted by P_c , T_c , ρ_c , respectively.

C. Expansion close to the critical point

We expand the van der Waals pressure around the critical point $\rho = \rho_c$, $T = T_c$. Since at the critical point, $dP/d\rho = d^2P/d\rho^2 = 0$, the expression can be written as

$$P(\rho, T) = P_c + \left. \frac{\partial P}{\partial T} \right|_{\rho_c, T_c} T_c \varepsilon + \left. \frac{\partial^2 P}{\partial T \partial \rho} \right|_{\rho_c, T_c} T_c \rho_c \varepsilon \hat{\rho} + \frac{1}{6} \left. \frac{\partial^3 P}{\partial \rho^3} \right|_{\rho_c, T_c} \rho_c^3 \hat{\rho}^3 + O(\hat{\rho}^4) + O(\varepsilon \hat{\rho}^2), \quad (4)$$

where higher derivatives in T than the first one vanish. Here, we have introduced the reduced temperature

$$\varepsilon = \frac{T - T_c}{T_c} = \tilde{T} - 1 \ll 1,$$

and the reduced density

$$\hat{\rho} = \frac{\rho - \rho_c}{\rho_c} = \tilde{\rho} - 1 \ll 1.$$

Note that both quantities are dimensionless. Using Eq. (3), Eq. (4) takes the form

$$P(\hat{\rho}, \varepsilon) = P_c \left[1 + 4\varepsilon + 6\varepsilon \hat{\rho} + \frac{3}{2} \hat{\rho}^3 \right] + O(\hat{\rho}^4) = P_c \hat{P}(\hat{\rho}, \varepsilon) + O(\hat{\rho}^4), \quad (5)$$

with $\hat{P}$ being also dimensionless. It is shown in Appendix A 2 that P can be derived from the bulk free energy density

$$f(\hat{\rho}) = P_c \left[\tilde{f}_0 + \tilde{f}_1(\varepsilon) \hat{\rho} + 3\varepsilon \hat{\rho}^2 + \frac{3}{8} \hat{\rho}^4 \right] + O(\hat{\rho}^5), \quad \tilde{f}_1 = 1 + 4\varepsilon,$$

according to

$$P = \frac{df}{d\hat{\rho}}.$$

For $\varepsilon < 0$, $f(\rho)$ has two minima,

$$\hat{\rho}_b = \pm 2\sqrt{-\varepsilon}, \quad (6)$$

with

$$\hat{P}(\hat{\rho}_b) = \hat{P}(-\hat{\rho}_b) = 1 + 4\varepsilon = \hat{P}_e,$$

where $\hat{P}_e$ is the equilibrium pressure. The values $\hat{\rho}_b$ in Eq. (6) correspond to the two equilibrium densities defining the two phases, gas and liquid (binodals), see Fig. 2. The spinodals follow from $d\hat{P}/d\hat{\rho} = 0$ as

$$\hat{\rho}_s = \pm 2\sqrt{-\varepsilon/3}, \quad (7)$$

and mark the totally unstable region in the $P - \rho$ diagram.

For $\varepsilon > 0$, only one equilibrium point at $\hat{\rho} = 0$ exists, denoting the supercritical phase.

D. Dimensionless NSKE close to the critical point

In the following, we use the cubic pressure expansion equation (5) in Eq. (1). We scale all lengths with ℓ , velocities with U_0 , time with ℓ/U_0 , densities with ρ_c , and pressure with P_c , and obtain the dimensionless set (for the isothermal case only) below the critical

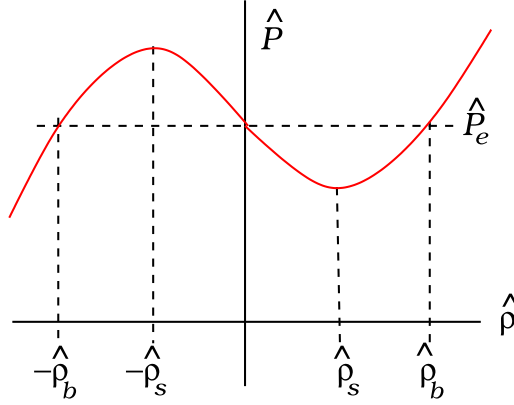


FIG. 2. Sketch of the pressure $\hat{P}$ according to Eq. (5). For $\hat{P} = \hat{P}_e$, the liquid ($\hat{\rho} = \hat{\rho}_b$) and the gaseous ($\hat{\rho} = -\hat{\rho}_b$) phase are in equilibrium.

point ($\varepsilon < 0$)

$$\frac{d\vec{v}}{dt} = \frac{1}{Re} \left(\nabla^2 \vec{v} + \frac{1}{3} \nabla (\nabla \cdot \vec{v}) \right) - E_u \nabla \left(6\varepsilon \hat{\rho} + \frac{3}{2} \hat{\rho}^3 \right) - G \hat{e}_z + \frac{1}{We} \nabla \nabla^2 \hat{\rho}, \quad (8a)$$

$$\frac{d\hat{\rho}}{dt} = -(1 + \hat{\rho}) \nabla \cdot \vec{v}, \quad (8b)$$

where we have assumed $|\varepsilon| \ll 1$. For the sake of simplicity (Stokes' hypothesis), we set the bulk viscosity $\zeta = 0$. The dimensionless groups read

$$Re = \frac{U_0 \ell \rho_c}{\eta}, \quad We = \frac{U_0^2 \ell^2}{\rho_c K}, \quad G = \frac{g \ell}{U_0^2}, \quad E_u = \frac{P_c}{\rho_c U_0^2}.$$

They correspond to the Reynolds number, Weber number, Galileo number, and Euler number, respectively. With the exception of G , all these numbers are non-negative by definition. Negative G denotes the case where gravity comes from above, i.e., the system is turned upside down, a situation we shall consider below in the section on the Rayleigh-Taylor instability.

E. Parameter sets

In order to consider a quasireal case in the frame of the van der Waals EOS (see details at the end of this section), we chose CO_2 as the working fluid with the parameter sets depicted in Table I.

Note that all numbers, except for the surface tension (last row), are dimensionless in the scaling of Sec. II D. Here, Δz denotes the step size for the finite difference mesh, the surface tension $\tilde{\sigma}$, and the interface width ξ will be defined later. For all runs, we take $P_c = 7 \times 10^6 \text{ N/m}^2$ and

TABLE I. Parameter sets used throughout the work.

Set no.	ε	We	Re	ξ	Δz	L_z	$\tilde{\sigma}$ [N/m]
1	-10^{-2}	0.16	200	0.012	0.003	1.2	0.13
2	-10^{-3}	0.0014	100	0.39	0.1	40	0.044
3	-10^{-4}	7.8	100	0.017	0.004	1.6	1.9×10^{-5}
4	10^{-3}		100		0.1	40	
5	10^{-5}		100		0.1	40	

$\rho_c = 470 \text{ kg/m}^3$, for the scaling length $\ell = 10^{-3} \text{ m}$ and $U_0 = \sqrt{g\ell}$ so that $|G| = 1$. This yields $E_u = 1.5 \times 10^6$.

To achieve numerical convergence, a certain damping is needed, resulting in smaller Reynolds numbers than for the realistic case. On the other hand, our damping rate is still much lower than the natural frequencies and growth rates that occur in the system. As a consequence, the artificially small Reynolds numbers will not change our results significantly.

The surface tension of sets 1 and 2 is intentionally taken higher to obtain a thicker interface. This avoids a crucial problem occurring for phase field methods. The interface width ξ (and therefore σ) has to be taken much larger than for the real case, where it has a size of some nanometers, at least not very close to the critical point. To resolve the interface numerically, at least three or four mesh points are needed. Realistic values for σ and ξ would thus result in a (dimensional) step size of less than a nanometer and would restrict the domain for the computations to some hundreds of nanometers. The only case where the interface width and the surface tension come close to the real one is that of set 3 in Table I [3], very close to the critical point.

III. LINEAR STABILITY OF THE NSKE, SUPERCRITICAL CASE

We begin with the supercritical case $\varepsilon > 0$, where only one phase exists, and capillary stresses do not occur. Then, $1/W_e = 0$, and the last term of Eq. (8a) vanishes.

A. Density stratified base state

The base state has vanishing velocities $\vec{v} = 0$ and $\hat{\rho} = \hat{\rho}_0(z)$. A solution of Eq. (8a) is provided by the real root of

$$E_u \left(6 \varepsilon \hat{\rho}_0 + \frac{3}{2} \hat{\rho}_0^3 \right) + G(z - L_z/2) = 0, \quad (9)$$

where the integration constant is fixed so that $\hat{\rho}(z = L_z/2) = 0$, leading to $\rho = \rho_c$ in the middle plane of the layer. Equation (9) is solved using a Newton-Raphson iteration [28]. Close to the critical point, the z -dependence of $\hat{\rho}_0$ shows a significant difference from the linear behavior, see Fig. 3. All solutions of Eq. (9) possess the point symmetry $\hat{\rho}(z) = -\hat{\rho}(-z)$.

B. Linearized set

We wish to perform a linear stability analysis around the base state $\hat{\rho}_0(z)$, computed from Eq. (9). Inserting $\hat{\rho} = \hat{\rho}_0(z) + \hat{\rho}_1$ into the system (8) and taking only the linear terms in $\hat{\rho}_1$, u , w results in a linear system with z -dependent coefficients. If periodic boundary conditions in x are assumed or the horizontal extension is large compared to the depth, a plane wave decomposition according to

$$\hat{\rho}_1(x, z, t) = \bar{\rho}(z) e^{ikx + \lambda t},$$

$$u(x, z, t) = i \bar{u}(z) e^{ikx + \lambda t},$$

$$w(x, z, t) = \bar{w}(z) e^{ikx + \lambda t},$$

yields the linear ODE eigenvalue problem

$$\lambda \bar{u} = \frac{1}{Re} \left(\bar{u}'' - \frac{4}{3} k^2 \bar{u} + \frac{1}{3} k \bar{w}' \right) - E_u \left(6 \varepsilon + \frac{9}{2} \hat{\rho}_0^2 \right) k \bar{\rho}, \quad (10a)$$

$$\lambda \bar{w} = \frac{1}{Re} \left(\bar{w}'' - \frac{4}{3} k^2 \bar{w} - \frac{1}{3} k \bar{u}' \right) - E_u \left(6 \varepsilon \bar{\rho}' + \frac{9}{2} d_z(\hat{\rho}_0^2 \bar{\rho}) \right), \quad (10b)$$

$$\lambda \bar{\rho} = -\hat{\rho}_0' \bar{w} - (\bar{w}' - k \bar{u})(1 + \hat{\rho}_0), \quad (10c)$$

where primes denote derivatives with respect to z . The boundary conditions at $z = 0, L_z$ read

$$\bar{u} = \bar{w} = 0, \quad \bar{\rho}' = 0.$$

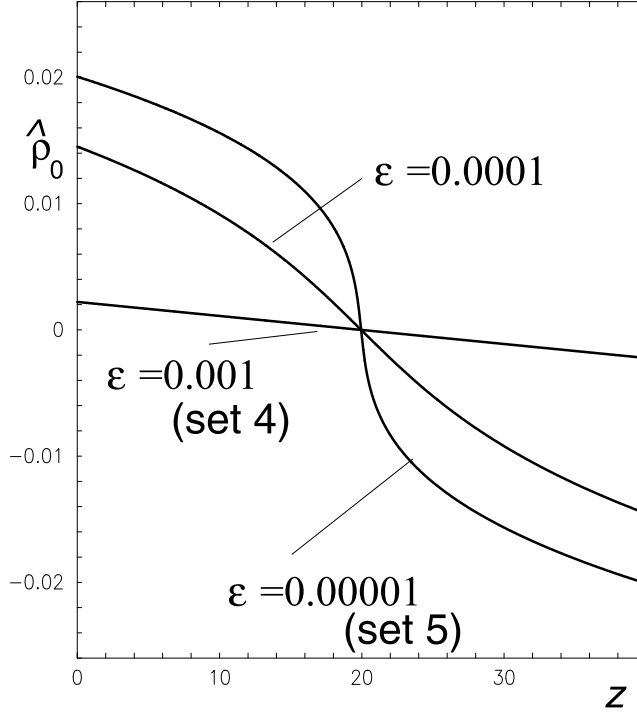


FIG. 3. Numerical solutions of Eq. (9) for $G = 1$, $E_u = 1.5 \times 10^6$, and different values of ε .

The last condition $\bar{\rho}' = 0$ means that $\hat{\rho}$ always has the same slope as the base profile $\hat{\rho}_0$. This is justified from the hydrostatic solution of the Navier-Stokes equations saying that the slope of $\hat{\rho}$ at the boundaries is fixed by G , see also Figs. 3 and 8. Discretizing the variables $\bar{u}$, $\bar{v}$, $\bar{\rho}$ and applying finite differences for the derivatives occurring in Eq. (10), an algebraic system of n_e equations is found, where $n_e = 3n_g - 4$ with n_g being the number of grid points on the z axis; for details, see the Appendix B. The system is solved by a standard linear algebra routine (LAPACK dgeev), and the spectrum

$$\lambda_j(k) = \gamma_j(k) + i\omega_j(k), \quad \gamma_j, \omega_j \in \mathcal{R}, \quad j = 1..n_e,$$

together with the eigenmodes can be computed. The real part γ_j is the growth rate of mode j , where ω_j denotes its eigenfrequency. Since the stratification $\hat{\rho}_0$ is stable, $\gamma_j < 0$ for all j .

C. Eigenmodes and spectrum, sound waves

For the approximate case of constant $\hat{\rho}_0$, and if viscous terms are neglected in Eq. (10), a wave equation for the density perturbation $\bar{\rho}$ of the form

$$\lambda^2 \bar{\rho} - v_s^2 (\bar{\rho}'' - k^2 \bar{\rho}) = 0, \quad (11)$$

is derived where v_s denotes the isothermal speed of sound that follows from Eq. (5) as

$$v_s^2(z) = \frac{P_c}{\rho_c U_0^2} \left(\frac{\partial \hat{P}}{\partial \hat{\rho}} \right)_\epsilon = E_u \left(6\varepsilon + \frac{9}{2} \hat{\rho}_0^2(z) \right). \quad (12)$$

For larger ε , $\hat{\rho}_0$ is very small (see Fig. 3) and the situation resembles that of an ideal gas where v_s depends only on temperature. Then, v_s becomes (almost) constant, $v_s^2 \approx 6E_u \varepsilon$, Fig. 4. Taking

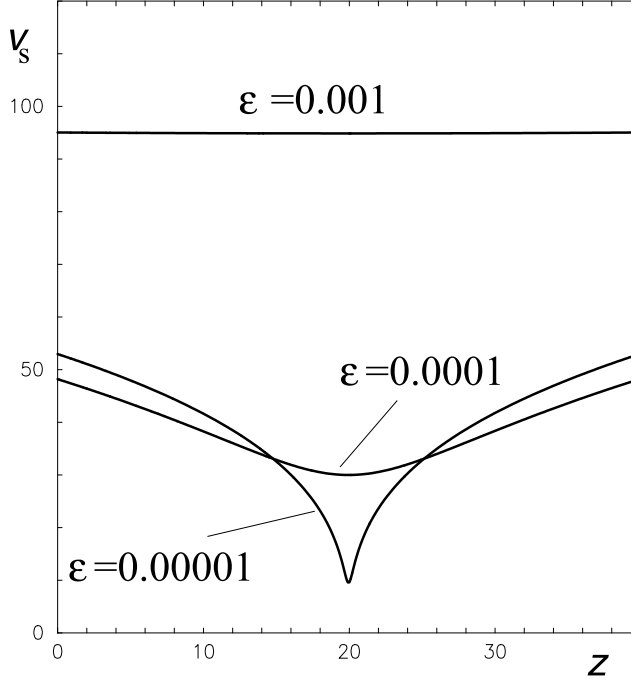


FIG. 4. Speed of sound according to Eq. (12) for $G = 1$, $E_u = 1.5 \times 10^6$, and different ε .

$\bar{\rho} \sim \cos(k_j z)$ as an exact solution of Eq. (11) yields $\lambda_j = \pm i\omega_j$ with

$$\omega_j(k) = v_s \sqrt{k_j^2 + k^2}, \quad k_j = \frac{j\pi}{L_z}, \quad j = 0, 1, \dots, n_e - 1, \quad (13)$$

shown in the left frame of Fig. 5. The frequencies (the imaginary parts of the eigenvalues λ) from the linear analysis equation (10) (black dots) agree very well with Eq. (13) (red lines). The eigenmodes have the form of classical trigonometric functions with the reduced density and velocity components shown in Fig. 6 for a selected value of $k = 0.2$.

There are drastic changes for smaller $\varepsilon = 10^{-5}$, corresponding to a temperature of only about 3 mK above the critical point for CO_2 . Then, the speed of sound depends significantly on the vertical coordinate and has a minimum in the middle plane, see Fig. 4. The eigenmodes differ from trigonometric functions, and the symmetric modes $j = 2, 4$ show pronounced peaks in the middle of the layer where the speed of sound varies at most; for the value of $k = 0.2$, see Fig. 7.

IV. LINEAR STABILITY OF THE NSKE, SUBCRITICAL CASE

A. Density stratified base state

Now we turn to the case $\varepsilon < 0$ where the two phases coexist. A stationary profile that separates the two phases is computed as a solution of Eq. (8a) with $\vec{v} = 0$:

$$\frac{1}{W_e} \hat{\rho}_0'' - E_u \left(6\varepsilon \hat{\rho}_0 + \frac{3}{2} \hat{\rho}_0^3 \right) = G(z - z_0), \quad (14)$$

where z_0 denotes the position of the interface located between the two binodals. For $G = 0$, Eq. (14) has the exact solution

$$\hat{\rho}_0(z) = 2\sqrt{|\varepsilon|} \tanh\left(\frac{z - z_0}{\xi}\right), \quad (15)$$

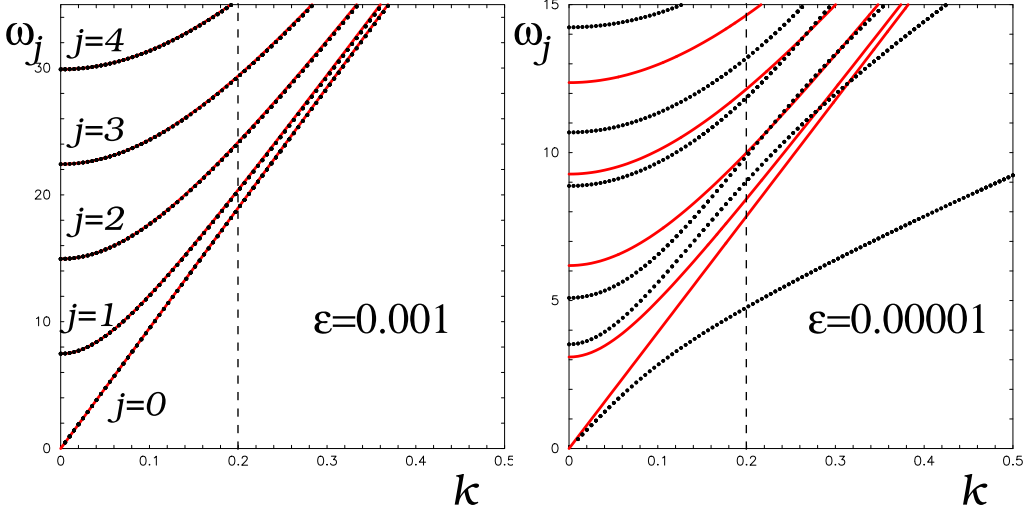


FIG. 5. Lowest frequency bands found from the linear problem equation (10) for $\varepsilon = 10^{-3}$, set 4 (left) for $\varepsilon = 10^{-5}$, set 5 (right). The red thin lines show ω_j according to Eq. (13), with averaged sound speed $\langle v_s \rangle_z$.

where

$$\xi = (3E_u W_e |\varepsilon|)^{-1/2}, \quad (16)$$

denotes the dimensionless width of the diffusive interface. The surface tension can be computed from the excess free energy as (for details, see Appendix A3)

$$\tilde{\sigma} = 16 |\varepsilon|^{3/2} \rho_c U_0^2 \ell \left(\frac{E_u}{3W_e} \right)^{1/2} = \rho_c U_0^2 \ell \sigma. \quad (17)$$

Note that $\tilde{\sigma}$ has the dimension N/m, whereas σ is dimensionless. Since we have assumed that K from Eq. (1) and therefore W_e do not depend on ε , we see that σ vanishes with $|\varepsilon|^{3/2}$ at the critical point, whereas ξ diverges with $|\varepsilon|^{-1/2}$.

One can eliminate W_e between Eqs. (16) and (17) and finds the interface width as a function of σ and ε as

$$\xi = \frac{1}{16E_u} \sigma |\varepsilon|^{-2}. \quad (18)$$

Since $\sigma \sim |\varepsilon|^s$ with $s \approx 1.2$ from the Ising model for near critical fluids [3], $\xi \sim |\varepsilon|^{-0.8}$ instead of $|\varepsilon|^{-1/2}$, but for both cases, the Ising model and van der Waals, the interface width diverges at the critical point as expected.

If $G \neq 0$, Eq. (14) can be solved only numerically. After discretization with n_g mesh points, Eq. (14) turns into an algebraic nonlinear system of n_g equations that is solved effectively by applying a multivariate Newton-Raphson scheme [28]. Results are shown in Fig. 8 for different values of G . Again, all solutions have the symmetry $\hat{\rho}(z) = -\hat{\rho}(-z)$.

B. Linearized set and eigenmodes

In the same way as for set (10), a linear ODE eigenvalue problem is derived that now reads

$$\begin{aligned} \lambda \bar{u} = & \frac{1}{R_e} \left(\bar{u}'' - \frac{4}{3} k^2 \bar{u} + \frac{1}{3} k \bar{w}' \right) - E_u \left(6\varepsilon + \frac{9}{2} \hat{\rho}_0^2 \right) k \bar{\rho} \\ & + \frac{1}{W_e} (k \bar{\rho}'' - k^3 \bar{\rho}), \end{aligned} \quad (19a)$$

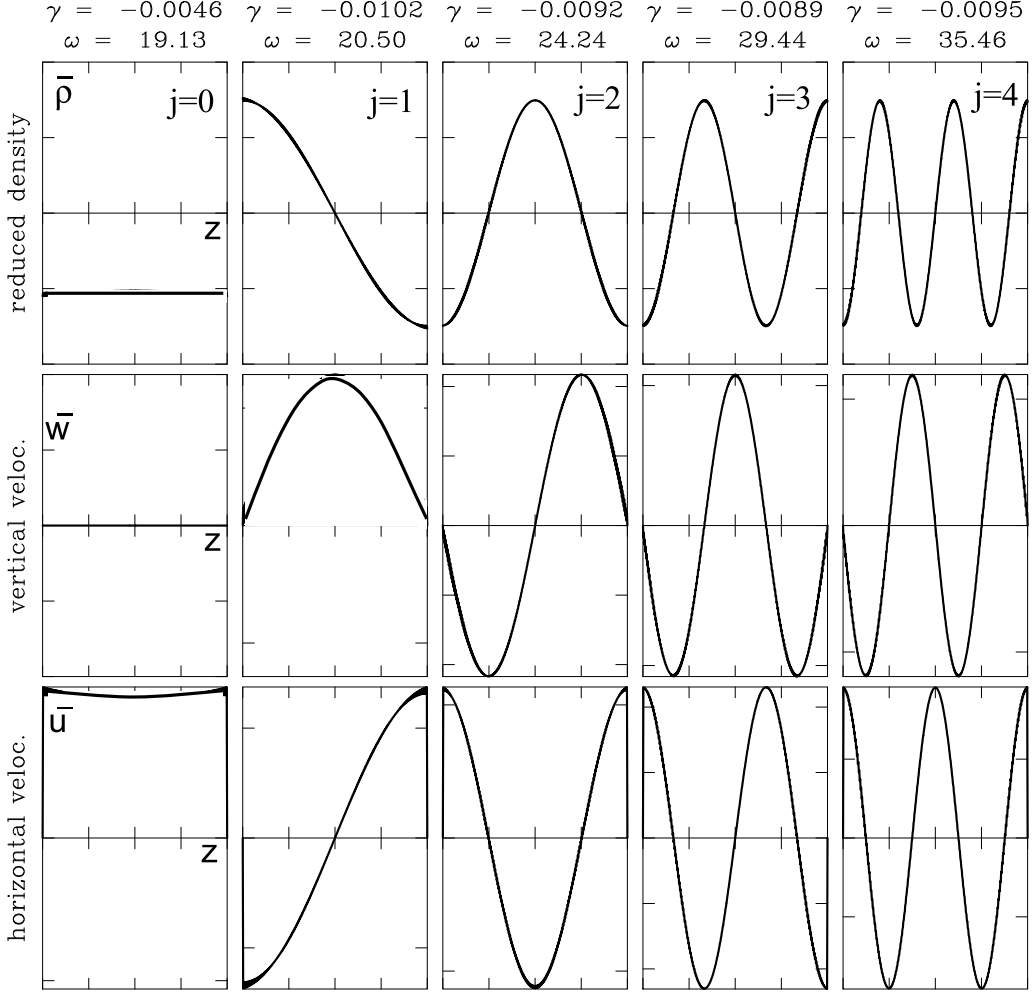


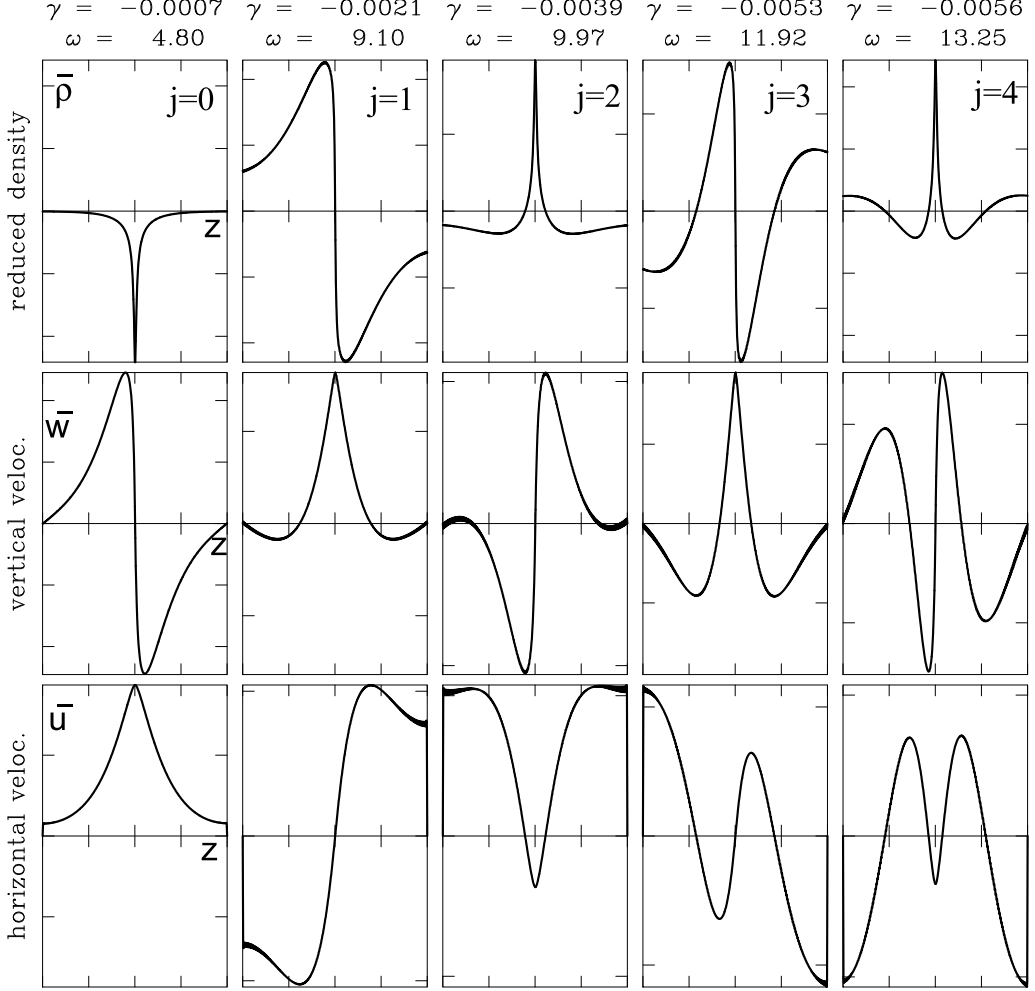
FIG. 6. First eigenfunctions over z for set 4, $\varepsilon = 10^{-3}$, belonging to the bands of Fig. 5, left frame, at $k = 0.2$.

$$\begin{aligned} \lambda \bar{w} = & \frac{1}{R_e} \left(\bar{w}'' - \frac{4}{3} k^2 \bar{w} - \frac{1}{3} k \bar{u}' \right) - E_u \left(6 \varepsilon \bar{\rho}' + \frac{9}{2} d_z (\hat{\rho}_0^2 \bar{\rho}) \right) \\ & + \frac{1}{W_e} (\bar{\rho}''' - k^2 \bar{\rho}'), \end{aligned} \quad (19b)$$

$$\lambda \bar{\rho} = -\bar{w} \hat{\rho}'_0 - (\bar{w}' - k \bar{u})(1 + \hat{\rho}_0). \quad (19c)$$

The first two eigenmodes sorted with respect to their frequencies are shown in Fig. 9(a). If $G > 0$, the stratification $\hat{\rho}_0(z)$ is stable and all eigenvalues have a negative real part $\gamma < 0$.

Although the situation is completely different for the supercritical one-phase system and the subcritical two-phase system with interface, some common features on both sides of the critical point concerning the eigenmodes and the spectrum can be seen if $|\varepsilon|$ approaches zero. For larger $\varepsilon = 10^{-3}$, the mode $j = 0, k = 0.2$ (left column in Fig. 6) is almost independent on z , corresponding to a pressure wave moving in a lateral direction with wavelength $2\pi/k$. This is not the case for smaller $\varepsilon = 10^{-5}$ where density and vertical velocity profiles resemble the profiles of the corresponding


 FIG. 7. First eigenfunctions over z for set 5, $\varepsilon = 10^{-5}$, $k = 0.2$.

mode for subcritical $\varepsilon = -10^{-4}$, compare the left columns of Figs. 7 and 9. The rather strong dependence of ρ in the supercritical case seems to mimic a real interface even for $\varepsilon > 0$ and without any Korteweg stresses. Also, the supercritical modes $j = 1, 2$ and their harmonics $j = 3, 4$ are approaching each other with respect to their frequencies for ε approaching zero, Fig. 7. For the subcritical case, this behavior is much more strongly pronounced, and their frequencies become almost equal. Note that in the central layer $z = L_z/2$, one mode always has zero vertical velocity (interface in rest), the other has maximal vertical velocity (the interface is moving upward and downward).

One main difference between supercritical and subcritical fronts is, however, their location and orientation. In the supercritical case, the front is, on average, oriented normally to the external (gravity) force, and its steepness increases with increasing force. Under microgravity conditions there would exist no “front” at all. On the other hand, in the subcritical case, fronts, or better domain walls, can emerge in a self-organized manner at each point and in each direction inside the fluid layer. This is known as spinodal decomposition and will be studied in Sec. VB 1.

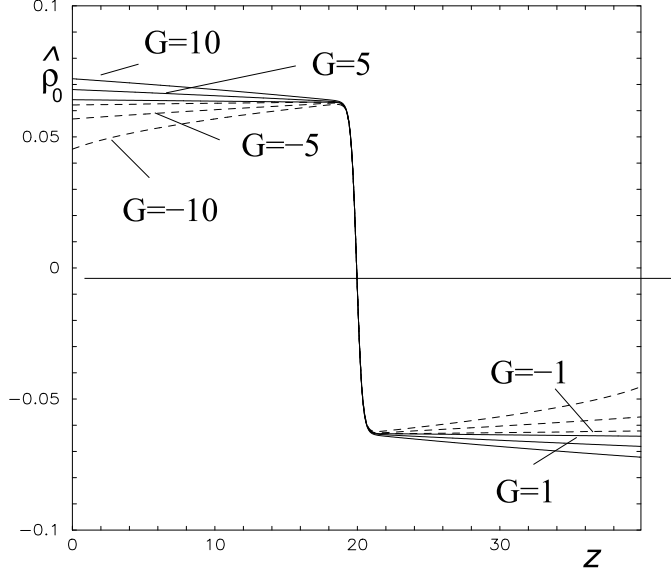


FIG. 8. Numerical solutions of Eq. (14) for set 2, $\varepsilon = -0.001$ and various G .

C. Rayleigh-Taylor instability

In the case $\varepsilon > 0$, all eigenvalues have negative real parts, and an RTI cannot occur from the linearized system. Contrary to the subcritical case, where an unstable density profile is still a stationary solution of the equations for the base state profile equation (14), this is not the case for $\varepsilon > 0$. Here, only a stable stratification can be a solution of Eq. (9).

If the layer is turned upside down with the heavier fluid (liquid) on top, we solve Eq. (14) with $G = -1$. The linear analysis then yields one mode with a positive growth rate γ , signaling the occurrence of the buoyancy-driven RTI, Fig. 10. It is interesting to compare the growth rates with those of two incompressible liquid layers separated by a sharp interface. There exists no exact analytical expression for viscous fluids in finite layers, although certain approximations are available; for a detailed discussion, see [29]. For the sake of simplicity, we resort here to the inviscid case derived already in [20]:

$$\gamma_{\text{in}} = \sqrt{\left(A_t G - \frac{1}{2} \sigma k^2\right) k \tanh\left(\frac{kL_z}{2}\right)}, \quad (20)$$

shown as red lines in Fig. 10. Here, A_t stands for the Atwood number, and for our case, it is given as

$$A_t = \frac{\rho_1 - \rho_2}{\rho_1 + \rho_2} = -\hat{\rho}_b = -2\sqrt{-\varepsilon},$$

with ρ_1, ρ_2 being the bulk densities of the lower (gas) and the upper (liquid) layer, respectively.

Previous works on RTI in two compressible layers give a controversial discussion of the dependence of the growth rate on compressibility [30,31]. The main tendency, however, is a decreasing growth rate if the compressibility increases [32]. On the contrary, we computed here much larger growth rates than those obtained for two incompressible layers separated by a sharp interface equation (20), see Fig. 10.

A possible explanation could be the different mechanism for RTI in a two-phase near-critical system with phase transition. From Fig. 9(b), we see that the unstable mode $j = 0$ has zero vertical

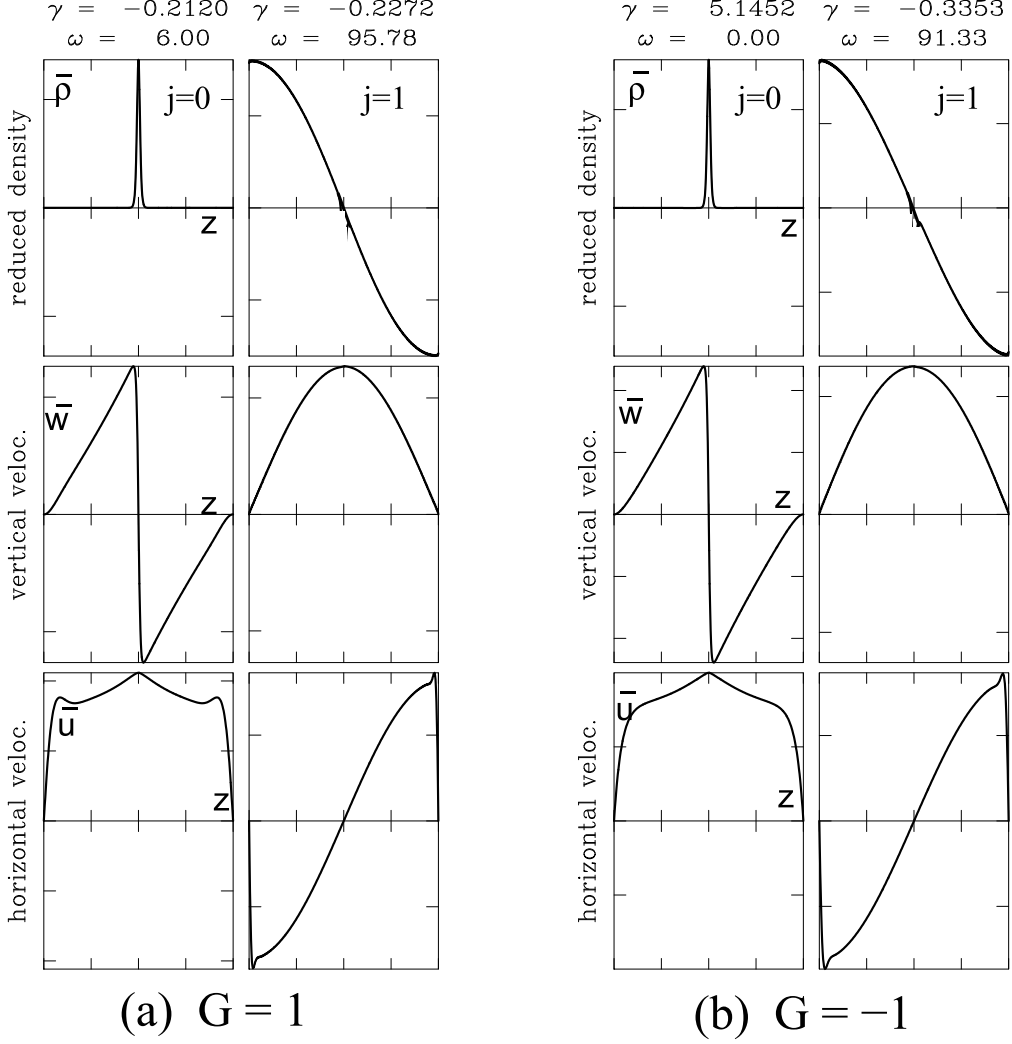


FIG. 9. First two eigenfunctions over z for set 3, $\varepsilon = -10^{-4}$, $k = 1$, (a) for $G = 1$, (b) for $G = -1$ where the heavier (liquid) layer is above the lighter one, and Rayleigh-Taylor instability occurs.

velocity $\bar{w}$ at the diffuse interface. This is in sharp contrast to the incompressible case, where $\bar{w}$ is large at the interface, which leads to a deformation and finally to RTI in this case.

In our case, $\bar{w}$ is positive below the interface and negative above; see Fig. 9(b). This leads to a compression and increases the density at the interface, which can be seen from the top frame of Fig. 9(b). As a result, the heavier fluid component invades the lighter one, and RTI sets in. This may happen on a totally different timescale than the standard RTI in two different fluids where no phase transition is involved.

Our preliminary computations show that the maximal growth rate is of $O(1/\varepsilon)$, in good agreement with Fig. 10.

On the other hand, the cutoff wave number k_0 with $\gamma(k > k_0) < 0$ and the wave number of the fastest growing modes fairly coincide. The cutoff wave number is the wave number that limits the band of positive growth rates on the right-hand side.

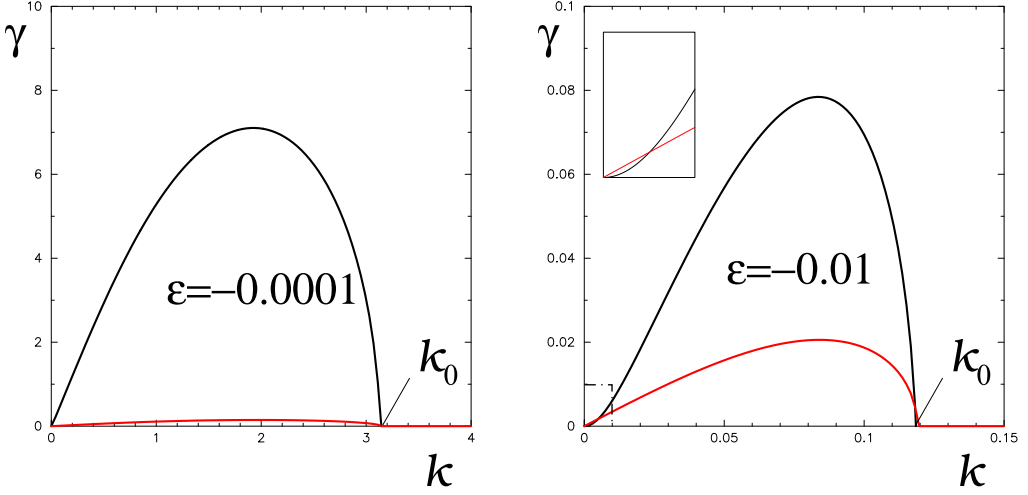


FIG. 10. Largest growth rate γ , left: for set 3, $\varepsilon = -10^{-4}$, $G = -1$, right: for set 1, $\varepsilon = -10^{-2}$, $G = -1$. Red curves mark the growth rates for inviscid, incompressible fluids according to Eq. (20). The inset shows the enlarged region close to the origin (dashed).

For the inviscid case, $\gamma \sim k$ if $k \rightarrow 0$, whereas for the viscous case $\gamma \sim R_e k^2$ [29,33], which can be distinguished clearly from the inset of the right frame of Fig. 10.

Figure 9(b) shows the first two modes for the RTI unstable situation with $G = -1$. If we compare these modes with the stable case of $G = 1$ [Fig. 9(a)], we see that the shape of the modes is almost identical. The mode $j = 0$ that becomes RT unstable for $G < 0$ corresponds to a damped internal wave along the surface for the stable case $G > 0$.

V. NUMERICAL RESULTS OF THE FULLY NONLINEAR SYSTEM

In this section, we perform direct numerical simulations of the nonlinear set (8). We apply a finite difference scheme for the spatial derivatives on a staggered grid, having $m \times n$ mesh points and side lengths $L_x = m \Delta x$, $L_z = n \Delta z$ where we chose $\Delta x = \Delta z$, Fig. 11. Here, Δz depends on the parameters and is chosen according to Table I. For the time iteration, a simple Euler forward method with a fixed time step is used [34]. For the explicit method, the time step is restricted to $O(\Delta z^2)$; we fixed it with $\Delta t = \Delta z^2/20$, leading to convergent results.

A. Supercritical states

We begin in the supercritical regime close to the critical point, parameter set 5 from Table I. As an initial condition, we take the following random distribution:

$$\hat{\rho}_{ij} = A \eta_{ij}, \quad u_{ij} = w_{ij} = 0, \quad (21)$$

with $A = 0.02$, corresponding to the maximum value of $\hat{\rho}_0$ shown in Fig. 3, and η_{ij} as equally distributed random numbers in $[-1, 1]$. Early stages in Fig. 12 show the formation of sound waves mainly in vertical directions with $k = 0$; see the movie Super1 in the Supplemental Material (SM) [35]. At later times (Fig. 13), waves having some similarity to surface waves occur, where the vertical density stratification approaches the one found for the stationary profile shown in Fig. 3, but now modulated in the x -direction and oscillating about the middle plane of the layer, see the movie Super2 in the SM [35]. Figure 14 shows density profiles along three cuts of the last stage in Fig. 13. These waves are oscillating with a frequency that corresponds to the lowest band of Fig. 5 at $k = 2\pi/L_x \approx 0.05$, Fig. 15. In this way, a “surface wave” emerges, but without a real surface and

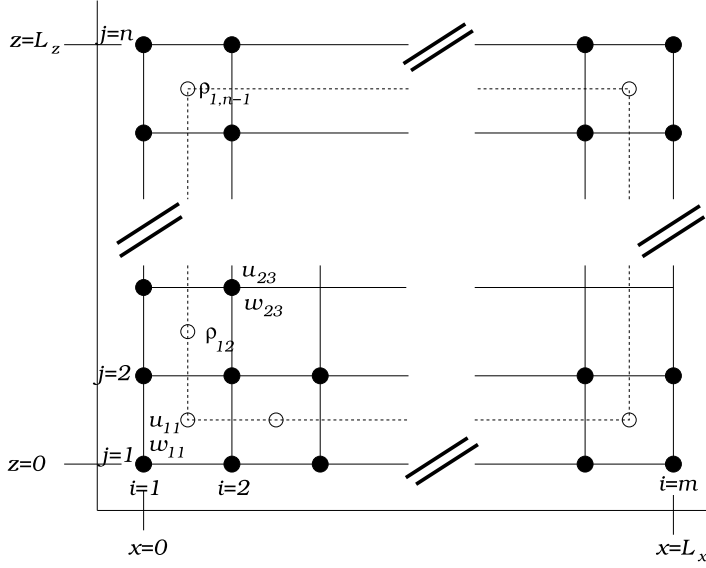


FIG. 11. Staggered grid for the numerical simulations. The vector nodes u_{ij} , w_{ij} lie at $x = (i - 1)\Delta x$, $z = (j - 1)\Delta z$; the scalar nodes ρ_{ij} , P_{ij} lie at $x = (i - 1/2)\Delta x$, $z = (j - 1/2)\Delta z$.

without surface tension in the supercritical domain. This is in excellent agreement with our results of the linearized problem, where such a “surface mode” was already detected in the eigenmodes plotted in the left column of Fig. 7.

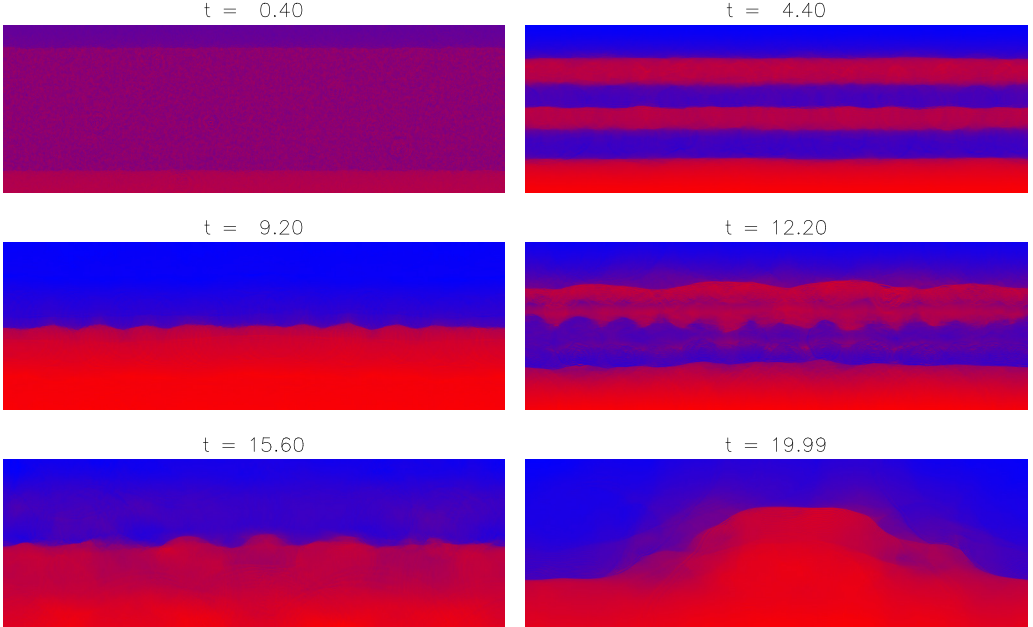


FIG. 12. Early stages of a randomly distributed initial density field $\hat{\rho}$ for $\varepsilon = 10^{-5}$, $G = 1$, set 5, 1200×400 -grid. Here, and in the following, the blue regions correspond to $\hat{\rho} < 0$; the red regions to $\hat{\rho} > 0$.

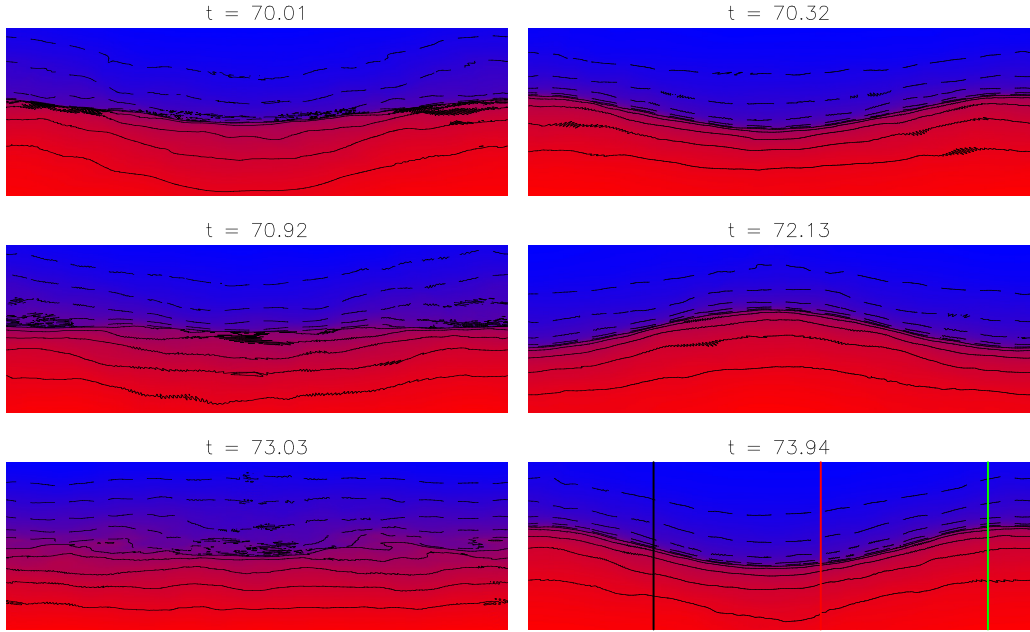


FIG. 13. Later stages of a randomly distributed initial density field for $\varepsilon = 10^{-5}$, set 5. Waves that strongly resemble surface waves can be seen where the density stratification is high. The vertical lines of the last frame correspond to the cross sections shown in Fig. 14.

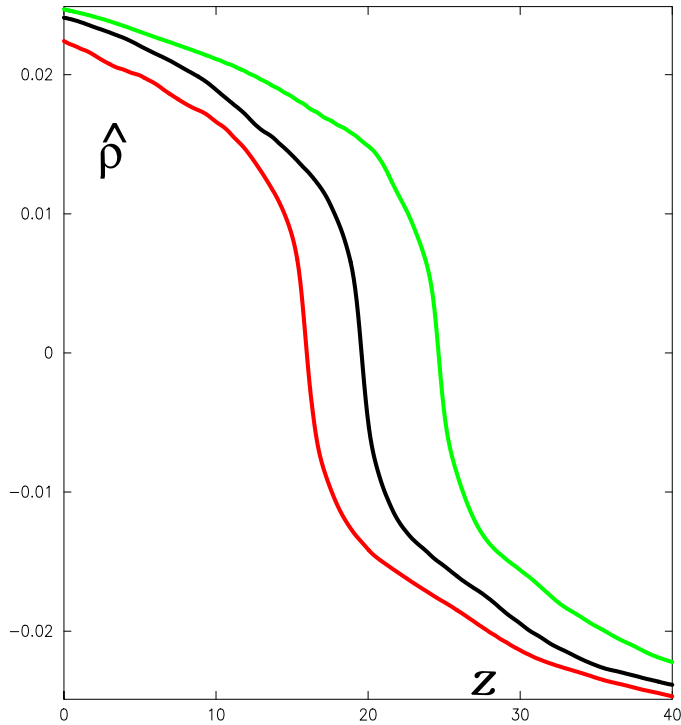


FIG. 14. Three cross sections in the z -direction for the density profiles shown in Fig. 13 at $t = 73.94$.

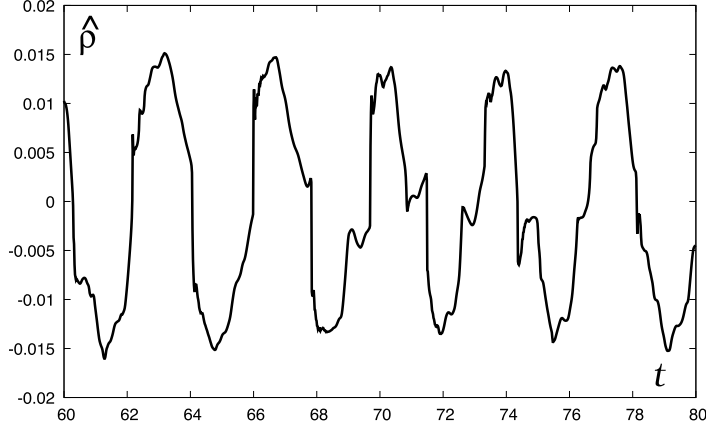


FIG. 15. Density $\hat{\rho}(L_x/2, L_z/2)$ in the middle of the layer over time for the series shown in Fig. 13. The frequency is $\omega \approx 1.7$, corresponding well to the value of the lowest band of Fig. 5, right frame for $k = 2\pi/L_x \approx 0.05$.

B. Subcritical states

1. Spinodal decomposition

Starting with the same initial condition [Eq. (21)], but now for $\varepsilon < 0$, spinodal decomposition is found in the very early phases $t < 0.004$, followed by a coarsening process that minimizes the length of the domain walls between the two binodal regions $\hat{\rho} = \pm \hat{\rho}_b$; see Fig. 16 and the movie Spinodal in the SM [35]. After a longer evolution, a state minimizing the free energy [see Appendix (A5)] is asymptotically reached, and the velocity goes to zero due to viscous damping.

2. Rayleigh-Taylor instability

The series shown in Fig. 17 was started with an initial condition found from solving Eq. (14) for set 3 and $G = 1$. Then, after $t = 0.1$, G was set to $G = -1$, and the instability was triggered, adding small fluctuations to $\hat{\rho}$. After $t \approx 0.5$, the manifestation of the Rayleigh-Taylor instability can be clearly seen, and the additional potential energy

$$\frac{\Delta E_p}{L_x} = G \left(\int_{L_z} dz (1 - \hat{\rho}_0) z - \int_{L_z} dz (1 + \hat{\rho}_0) z \right) = -2 G \int_{L_z} dz \hat{\rho}_0 \approx \sqrt{-\varepsilon} G L_z^2, \quad (22)$$

with $\hat{\rho}_0$ from Eq. (15) is turned into kinetic energy and finally into friction when a stable state with the heavier liquid on top is reached (note that gravity is now going into the positive z -direction). The total potential energy is plotted in Fig. 18(a), where the energy drop is clearly visible. The total kinetic energy

$$E_k(t) = \frac{1}{2} \int_{L_x} dx \int_{L_z} dz (u^2 + w^2), \quad (23)$$

over time is plotted on Fig. 18(b). The fast oscillations before RTI are due to sound waves and correspond to the frequency of the mode $j = 1, k = 0$ (not shown), which can be estimated with $v_s = 42$ from Eq. (12) as $\omega = v_s(\pi/L_z) \approx 83$. The much slower oscillations following RTI are surface waves and belong to the mode shown in the leftmost column in Fig. 9(a). Note that $E_k(t)$ oscillates with the double frequency of the modes $\rho, \vec{v}$ due to the quadratic dependence in Eq. (23). The movie RTI showing the total pattern formation can be found in the SM [35].

We have thus identified, for the first time, three regimes in RTI of the subcritical state: (i) the first regime corresponding to fast oscillations just before RTI; (ii) the RTI regime at intermediate

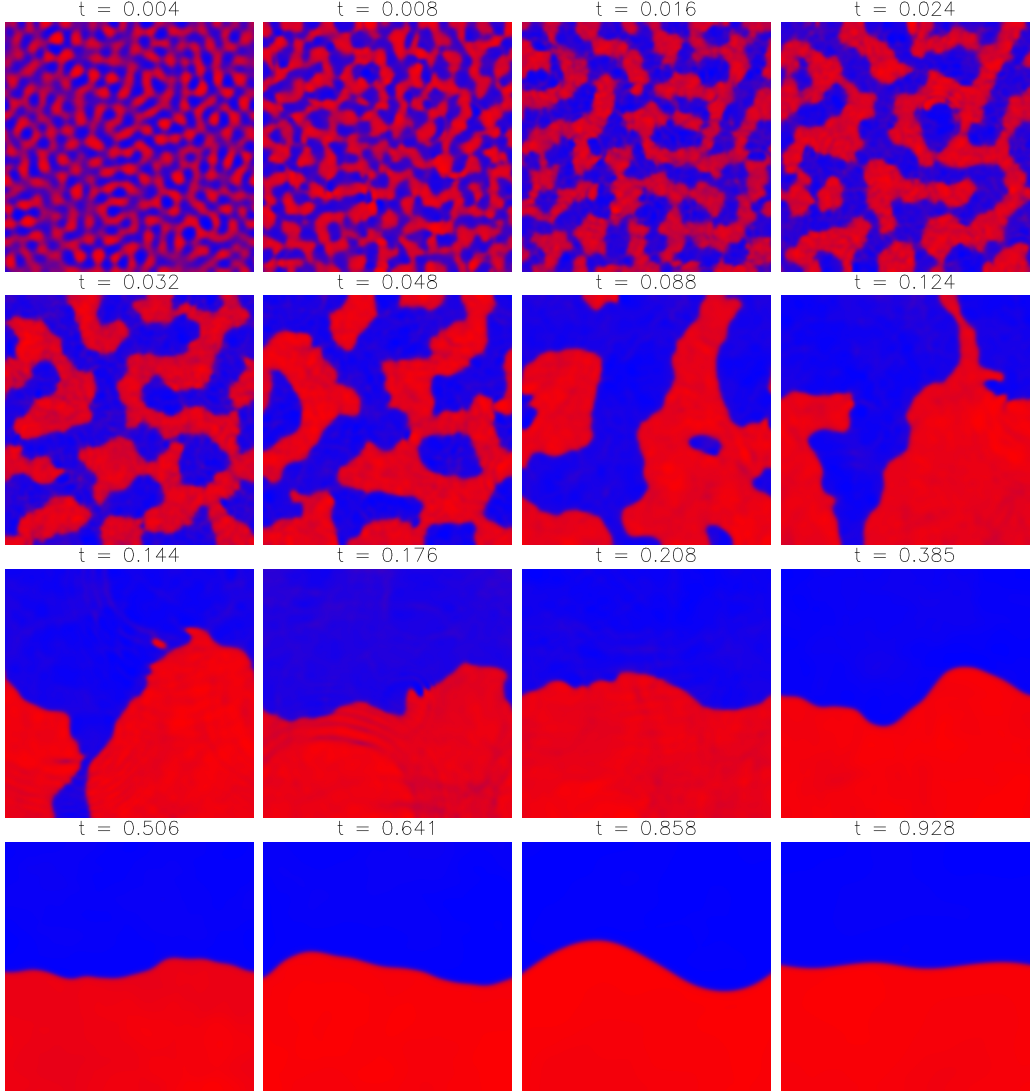


FIG. 16. Spinodal decomposition. An initial random state decomposes into the two stable phases $\pm\rho_b$. Parameters for $\varepsilon = -10^{-4}$, set 3, 400×400 -grid.

frequencies; and (iii) the slower oscillation regime (low frequency), corresponding to surface waves.

VI. CONCLUSIONS

The compressible isothermal Navier-Stokes-Korteweg equations together with a van der Waals equation of state have been considered in the present work. The expansion of the pressure close to the critical point results in a quartic polynomial in the density with well-defined liquid and gaseous equilibrium phases. Two cases are considered: (i) supercritical state and (ii) subcritical state. In the supercritical case, the linearized set leads to an eigenvalue problem with eigenmodes having negative growth rates. The linearized set determines the isothermal speed of sound as a function of the vertical axis and vanishes at the critical point. The eigenfunctions (density and

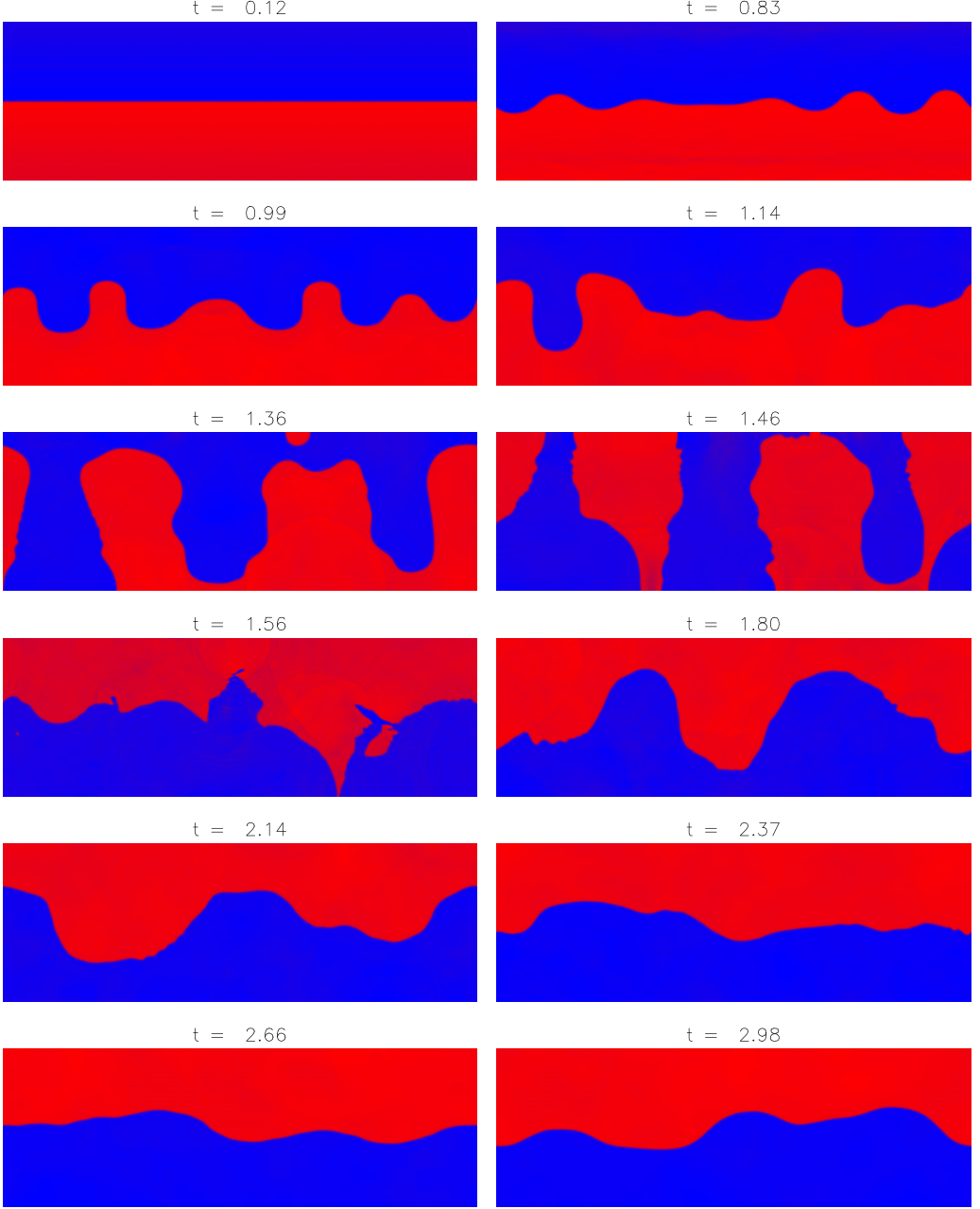


FIG. 17. The Rayleigh-Taylor instability occurs if gravity is changed from $-\hat{e}_z$ to $\hat{e}_z$. Parameters for $\varepsilon = -10^{-4}$, set 3, 1200×400 -grid.

velocity) are plotted as a function of the vertical coordinate for different values of the distance to the critical point and wave number. Very close to the critical point, the supercritical case mimics a real interface from the sharp variations of the density profiles in the midcavity where the density is at its critical value. In the subcritical case, the same approach of the linearized set is conducted with the inclusion of surface tension through the Weber number for the stratified base state density

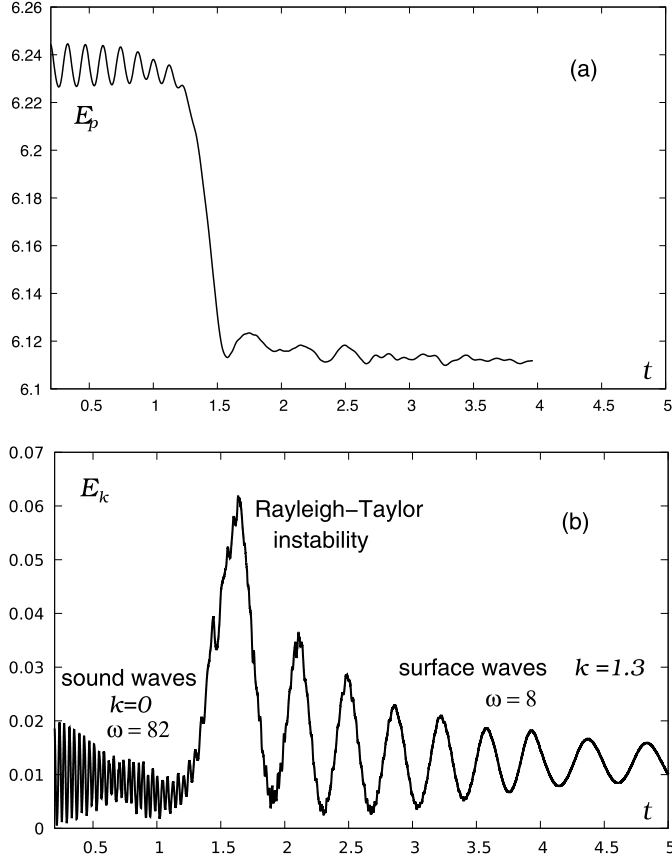


FIG. 18. (a): Total potential energy to the series shown in Fig. 17. When RTI emerges, the energy drop according to Eq. (22) is clearly observable. (b): Total kinetic energy to the series shown in Fig. 17. Until $t \approx 1$, sound waves occur with a frequency that corresponds to the lowest nontrivial $k = 0$ mode. The slow oscillations following RTI are surface waves and belong to the mode shown in the leftmost column in Fig. 9(a). The damping rate $|\gamma| \approx 0.28$ found from the linear analysis agrees fairly well with the exponential decay time seen after the RTI.

profile. The eigenfunctions show some common features on both sides close to the critical point. The Rayleigh-Taylor instability for the subcritical state shows a very large growth rate close to the critical point when compared to inviscid incompressible fluids with approximately the same cutoff wave number. We have also performed numerical simulations at the supercritical state with an initial random distribution of the density field. The stratified density field is recovered and matches well with the linear solutions. Even if the conditions are at the supercritical state, a kind of surface wave emerges, again in close agreement with the linear results. Finally, in the subcritical state, the classical spinodal decomposition is shown for a random initial distribution. For the case of the Rayleigh-Taylor instability, we found three regimes for the time evolution of the total kinetic energy: (i) at early times, the propagation of sound waves with high frequencies in the vertical direction, (ii) the Rayleigh-Taylor instability, and (iii) the low-frequency surface waves with finite wavelength.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: FREE ENERGY AND SURFACE TENSION

1. Stationary base state of NSKE

For a stationary solution,

$$\vec{v} = 0, \quad \partial_t \vec{v} = 0, \quad \partial_r \rho = 0,$$

and $\rho = \rho(z)$. The set (1) reduces to

$$K \rho''' - \frac{1}{\rho} P' - g = 0,$$

where primes denote derivatives with respect to z . This can be integrated to

$$K \rho'' - h(\rho) - g z = 0, \tag{A1}$$

with

$$h(\rho) = \int d\rho \frac{1}{\rho} \frac{dP}{d\rho} = \frac{P}{\rho} + \int d\rho \frac{P}{\rho^2}. \tag{A2}$$

Now we use the thermodynamic relation for the pressure

$$P = -\frac{dF}{dv} = \rho^2 \frac{d}{d\rho} \left(\frac{f}{\rho} \right), \tag{A3}$$

where v is the molar volume, $F = M f/\rho$ is the free energy, f is the free energy density, and M is the molar mass. Then $h = df/d\rho$, and Eq. (A1) writes as

$$K \rho'' - \frac{df}{d\rho} - g z = 0. \tag{A4}$$

2. Free energy density for the van der Waals equation

Equation (A4) is the Euler-Lagrange equation to the variational problem

$$\delta F = 0,$$

with the free energy functional

$$F[\rho] = \int dz \left[\frac{1}{2} K (d_z \rho)^2 + f(\rho) + g \rho z \right], \tag{A5}$$

which is the sum of a surface term, a bulk term, and the potential energy.

For the bulk free energy density of the van der Waals EOS (3), one finds from Eq. (A3)

$$f(\tilde{\rho}) = \frac{8}{3} P_c \left[\tilde{\rho} \tilde{T} \ln \frac{\tilde{\rho}}{1 - \tilde{\rho}/3} - \frac{9}{8} \tilde{\rho}^2 + f_0(\tilde{T}) + f_1(\tilde{T}) \tilde{\rho} \right], \tag{A6}$$

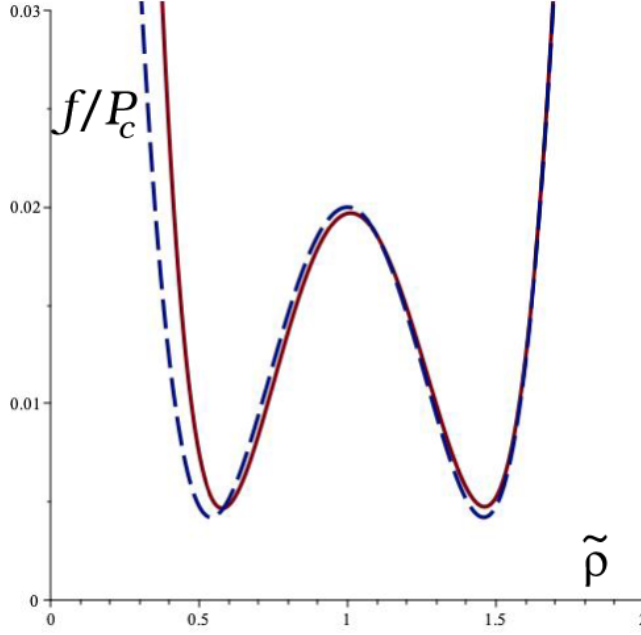


FIG. 19. Free energy density f/P_c over $\tilde{\rho}$ for a van der Waals gas with $\tilde{T} = 0.95$. Solid, red: from Eq. (A6), dashed, blue: from the quartic expansion (A8).

with $\tilde{\rho} = \rho/\rho_c$ and $\tilde{T} = T/T_c$. The functions f_i are determined by a caloric EOS and are not of interest in the isothermal case considered here. Figure 19 shows the free energy density f for $\tilde{T} = 0.95$ and $f_1 = 0.441$ where the two equilibria, the binodals, lie almost on the same level (red curve).

To find the stationary front solutions, we have to solve Eq. (A4) with f from Eq. (A6):

$$\rho_c K \tilde{\rho}'' - \frac{8}{3} \frac{P_c}{\rho_c} \left[\tilde{T} \ln \frac{\tilde{\rho}}{1 - \tilde{\rho}/3} + \frac{\tilde{T}}{1 - \tilde{\rho}/3} - \frac{9}{4} \tilde{\rho} - f_1 \right] - g z = 0. \quad (\text{A7})$$

Contrary to Eq. (14), an exact solution of Eq. (A7) is not known.

Expanding Eq. (A6) with respect to $\hat{\rho} = \tilde{\rho} - 1$ yields

$$f(\hat{\rho}) = P_c \left[\tilde{f}_0 + \tilde{f}_1 \hat{\rho} + 3 \varepsilon \hat{\rho}^2 + \frac{3}{8} (1 + \varepsilon) \hat{\rho}^4 \right] + O(\hat{\rho}^5), \quad (\text{A8})$$

which is also shown in Fig. 19 (blue dashed curve). With the approximation $\varepsilon \ll 1$, Eq. (A8) turns into

$$f(\hat{\rho}) = P_c \left[\tilde{f}_0 + \tilde{f}_1 \hat{\rho} + 3 \varepsilon \hat{\rho}^2 + \frac{3}{8} \hat{\rho}^4 \right]. \quad (\text{A9})$$

Inserting Eq. (A9) into Eq. (A4) yields

$$\rho_c K \hat{\rho}'' - \frac{P_c}{\rho_c} \left[6 \varepsilon \hat{\rho} + \frac{3}{2} \hat{\rho}^3 - \tilde{f}_1 \right] - g z = 0. \quad (\text{A10})$$

After scaling, Eq. (A10) turns into (14). The constant $\tilde{f}_1$ can be fixed to set the position of the interface, i.e., taking $P_c \tilde{f}_1 = -g \rho_c z_0$ at $z = z_0$.

3. Surface tension

To find an expression for the surface tension σ , one considers the difference of the free energy of a uniform state with $\hat{\rho}_b = \pm 2\sqrt{-\varepsilon}$ to that of the interface state $\rho_0(z)$ as a solution of Eq. (A10) (for

the sake of simplicity, we put $g = f_1 = 0$):

$$\sigma = F[\hat{\rho}_0] - F[\hat{\rho}_b] = \frac{\rho_c^2 K}{2} \int dz (d_z \hat{\rho}_0)^2 + \int dz (f(\hat{\rho}_0) - f(\hat{\rho}_b)). \quad (\text{A11})$$

From Eq. (A4), a first integral reads (energy theorem)

$$\frac{\rho_c^2 K}{2} (d_z \hat{\rho}_0)^2 = f(\hat{\rho}_0) - f(\hat{\rho}_b), \quad (\text{A12})$$

and thus

$$\sigma = \rho_c^2 K \int_{-\infty}^{\infty} dz (d_z \hat{\rho}_0)^2 = \rho_c^2 K \int_{-\hat{\rho}_b}^{\hat{\rho}_b} d\hat{\rho}_0 d_z(\hat{\rho}_0).$$

Finally, inserting $d_z \hat{\rho}_0$ from Eq. (A12) yields the important formula

$$\sigma = \rho_c \sqrt{2K} \int_{-\hat{\rho}_b}^{\hat{\rho}_b} d\hat{\rho}_0 \sqrt{f(\hat{\rho}_0) - f(\hat{\rho}_b)}. \quad (\text{A13})$$

For the polynomial expansion (A8), this evaluates as

$$\sigma = 16\rho_c \sqrt{\frac{K P_c}{3}} |\varepsilon|^{3/2}, \quad (\text{A14})$$

which, after scaling, is the same as Eq. (17).

APPENDIX B: NUMERICAL SCHEME

We discretize the variables $\bar{\rho}$, $\bar{u}$, $\bar{w}$ on a mesh with n_g equidistant nodes, according to

$$\bar{\rho}_j = \bar{\rho}(z_j), \quad \bar{u}_j = \bar{u}(z_j), \quad \bar{w}_j = \bar{w}(z_j),$$

with $z_j = (j - 1)\Delta z$ and $j = 1 \dots n_g$.

Introducing the n_e dimensional vector

$$\vec{Q} = (\bar{\rho}_1 \dots \bar{\rho}_{n_g}, \bar{u}_2 \dots \bar{u}_{n_g-1}, \bar{w}_2 \dots \bar{w}_{n_g-1}),$$

with $n_e = 3n_g - 4$, and approximating the derivatives with finite differences of $O(\Delta z^2)$, the system (19) takes the form of an algebraic eigenvalue problem

$$\lambda \vec{Q} = \underline{M} \vec{Q}.$$

The nonzero matrix elements of $\underline{M}$ for the first n_g equations for $\bar{\rho}_j$ read

$$\begin{aligned} M_{1,2n_g-1} &= -\frac{1 + (\hat{\rho}_0)_1}{2\Delta z} \\ M_{n_g,3n_g-4} &= \frac{1 + (\hat{\rho}_0)_{n_g}}{2\Delta z} \\ M_{j,j+2n_g-3} &= -(\hat{\rho}'_0)_j, \quad 1 < j < n_g \\ M_{j,j+n_g-1} &= k(1 + (\hat{\rho}_0)_j), \quad 1 < j < n_g \\ M_{j,j+2n_g-2} &= -\frac{1 + (\hat{\rho}_0)_j}{2\Delta z}, \quad 1 < j < n_g - 1 \\ M_{j,j+2n_g-4} &= \frac{1 + (\hat{\rho}_0)_j}{2\Delta z}, \quad 2 < j < n_g. \end{aligned}$$

For the next $n_g - 2$ equations for $\bar{u}_j$,

$$\begin{aligned}
 M_{j,j} &= -\frac{1}{Re} \left(\frac{2}{\Delta z^2} + \frac{4}{3} k^2 \right), \quad n_g < j < 2n_g - 1 \\
 M_{j,j-n_g+1} &= -k \left(\frac{2\beta}{We} + E_u \alpha_{j-n_g+1} \right), \quad n_g < j < 2n_g - 1 \\
 M_{j,j-n_g} &= \frac{k}{We \Delta z^2}, \quad n_g < j < 2n_g - 1 \\
 M_{j,j-n_g-2} &= \frac{k}{We \Delta z^2}, \quad n_g < j < 2n_g - 1 \\
 M_{j,j+1} &= \frac{1}{Re \Delta z^2}, \quad n_g < j < 2n_g - 2 \\
 M_{j,j-1} &= \frac{1}{Re \Delta z^2}, \quad n_g + 1 < j < 2n_g - 1 \\
 M_{j,j+n_g-1} &= \frac{k}{6Re \Delta z}, \quad n_g < j < 2n_g - 2 \\
 M_{j,j+n_g-3} &= -\frac{k}{6Re \Delta z}, \quad n_g + 1 < j < 2n_g - 1.
 \end{aligned}$$

And for the last $n_g - 2$ equations for $\bar{w}_j$,

$$\begin{aligned}
 M_{3n_g-4,n_g} &= \frac{1}{2We \Delta z^3} + \frac{\beta}{We \Delta z} - \frac{E_u}{2\Delta z} \alpha_{n_g} \\
 M_{2n_g-1,1} &= -\frac{1}{2We \Delta z^3} - \frac{\beta}{We \Delta z} + \frac{E_u}{2\Delta z} \alpha_1 \\
 M_{j,j} &= -\frac{1}{Re} \left(\frac{2}{\Delta z^2} + \frac{4}{3} k^2 \right), \quad 2n_g - 2 < j < 3n_g - 3 \\
 M_{j,j+1} &= \frac{1}{Re \Delta z^2}, \quad 2n_g - 2 < j < 3n_g - 4 \\
 M_{j,j-1} &= \frac{1}{Re \Delta z^2}, \quad 2n_g - 1 < j < 3n_g - 3 \\
 M_{j,j-n_g+3} &= -\frac{k}{6Re \Delta z}, \quad 2n_g - 2 < j < 3n_g - 4 \\
 M_{j,j-2n_g+5} &= \frac{1}{2We \Delta z^3}, \quad 2n_g - 2 < j < 3n_g - 4 \\
 M_{j,j-n_g+1} &= \frac{k}{6Re \Delta z}, \quad 2n_g - 1 < j < 3n_g - 3 \\
 M_{j,j-2n_g+1} &= -\frac{1}{2We \Delta z^3}, \quad 2n_g - 1 < j < 3n_g - 3 \\
 M_{j,j-2n_g+4} &= \frac{\beta}{We \Delta z} - \frac{E_u}{2\Delta z} \alpha_{j-2n_g+4}, \quad 2n_g - 2 < j < 3n_g - 3 \\
 M_{j,j-2n_g+2} &= -\frac{\beta}{We \Delta z} + \frac{E_u}{2\Delta z} \alpha_{j-2n_g+2}, \quad 2n_g - 2 < j < 3n_g - 3,
 \end{aligned}$$

where α_j and β are defined as

$$\alpha_j = \frac{9}{2} (\hat{\rho}_0)_j^2 + 6\varepsilon, \quad \beta = \frac{1}{\Delta z^2} + \frac{k^2}{2}.$$

The matrix elements for the supercritical system (10) are formally obtained by $W_e \rightarrow \infty$, i.e., setting all terms that contain W_e to zero.

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