On the role of thermal fluctuations in Rayleigh-Taylor mixing

Kiran Narayanan, Ravi Samtaney*

Physical Science and Engineering Division, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia 23955-6900

Abstract

We study the significance of thermal fluctuations in fluid mixing that is induced by the Rayleigh-Taylor instability (RTI) using numerical solutions of the fluctuating compressible Navier-Stokes equations. Our results indicate that thermal fluctuations can trigger the onset of RTI at an initially unperturbed fluid-fluid interface and lead to mixing of multi-mode character with growth rates of $\alpha_s = 0.035$ and $\alpha_b = 0.023$ for the spikes and bubbles, respectively. In addition, we find that whether or not thermal fluctuations quantitatively affect the mixing behavior, depends on the magnitude of the dimensionless Boltzmann number of the system, and not solely on its size. When the Boltzmann number is much smaller than unity, the quantitative effect of thermal fluctuations on the mixing behavior is negligible and the behavior is the average of the outcome from several stochastic instances, with the ensemble of stochastic instances providing bounds on behavior. When the Boltzmann number is of order unity, we find that thermal fluctuations can significantly affect the mixing behavior; the ensemble-averaged solution shows a departure from the deterministic solution at late times. We conclude that for such systems, it is important to account for thermal fluctuations in order to correctly capture their physical behavior.

Keywords: Rayleigh-Taylor instability, fluctuating hydrodynamics, compressible Navier-Stokes simulations

*Corresponding author

Email addresses: kiran.narayanan@kaust.edu.sa (Kiran Narayanan), ravi.samtaney@kaust.edu.sa (Ravi Samtaney)

1. Introduction

Atomic motion at finite temperature gives rise to thermal fluctuations in a fluid that can affect flow phenomenae such as hydrodynamic instabilities [14] and fluid dynamics in nanochannels [7]. Presently, we study the significance of thermal fluctuations on the growth of the mixing layer in Rayleigh-Taylor instability (RTI) via numerical simulations of the two-fluid, fluctuating compressible Navier-Stokes (FCNS) equations. RTI is the driving mechanism for applications ranging from fluidized beds to inertial confinement fusion. It occurs when a heavy fluid of density ρ_h rests on top of a light fluid of density ρ_l , with acceleration due to gravity g acting normal to the unperturbed interface in the direction opposite to the density gradient. Here, we consider argon and helium as the heavy and light gas, respectively, for which the Atwood number defined as $A_t = (\rho_h - \rho_l) / (\rho_h + \rho_l)$ is 0.818.

Traditional continuum descriptions of macroscopic hydrodynamic systems employ physical models such as the compressible Navier-Stokes equations (CNS), neglect the effect of thermal fluctuations. When numerically solving such equations so as to study the mixing behavior for RTI, one has to induce the instability by perturbing the interface with a spectrum [22, 21]. In contrast, when using a physical model of the system such as [Direct Simulation Monte Carlo method that naturally accounts for thermal fluctuations \[11\]](#), one starts the numerical simulation with a flat interface and subsequently, perturbations at the interface that are induced by the fluctuations, trigger the instability. [The ability of thermal fluctuations to trigger the onset of RTI was demonstrated in \[10\] using DSMC simulations of a flat interface separating Helium and Argon gas.](#) Here, the authors also showed that the DSMC method that naturally captures fluctuations produced results that were in quantitative agreement with theoretical and empirical models in the linear, non-linear and self-similar regimes. This approach of using a physical model that accounts for thermal fluctuations when performing numerical simulations of RTI, has also been emphasized by

the authors in [14], owing to their observation of the significant qualitative and quantitative effect thermal fluctuations can have on RTI-driven mixing behavior. The authors reported that MD simulations predicted growth rate values were in the range $0.05 - 0.06$ that were closer in agreement with experimental values, in comparison to CNS simulations in [8] that used short wavelength modes and yielded values in the range $0.02 - 0.035$, that were half the experimental value. Although [particle methods such as DSMC and MD](#) can successfully capture thermal fluctuations, the authors [in both \[14\] and \[10\]](#) note that the computational cost is high and limits the size of the system that can be simulated to the order of a few micrometers. [FCNS on the other hand is a meso-level coarse-grained physical model \[9\] and offers the ability to include thermal fluctuations while keeping computational cost reasonable so as to be able to simulate system sizes in the order of millimeters.](#)

Here, we demonstrate that numerical simulations of RTI using FCNS as the physical model, help capture the qualitative behavior of thermal fluctuations triggering the multimode instability. In [3], it has been shown using MD simulations up to early times for a system of Atwood number 0.86 that is commensurate with ours, that the deterministic predictions of the growth rate for RTI, are ensemble-averaged outcomes of underlying microscopic processes that occur across multiple scales, even for system lengths as small as one micron. The author in [15] motivates the need for such an investigation for RTI at late times. Such an emergence of macroscopic behavior from the average of several stochastic instances was also established in the context of the Richtmyer-Meshkov instability (RMI) in our previously published work that employed FCNS simulations [17]. Presently, we show that for systems with Boltzmann number much less than unity ($Bo \ll 1$), deterministic RTI-driven mixing behavior appears from ensemble-averaged mixing behavior of the FCNS simulations whereas for systems with $Bo \approx 1$, the fluctuations significantly affect mixing behavior such that the ensemble-averaged behavior departs from the deterministic one. [Here,](#)

the Boltzmann number is given by The Boltzmann number is given by

$$Bo = \sqrt{\frac{2}{3}} \cdot \frac{u_{th}}{u_o} \cdot \frac{1}{\sqrt{n_0}}, \quad (1)$$

where $u_{th} = \sqrt{3k_bT/m_0}$ is the thermal velocity, m_0 is the molecular mass, and n_0 is the number of molecules contained in volume L_0^3 . Bo determines the relative magnitude of thermal fluctuations.

2. Simulation Setup

Here, we use numerical simulations of FCNS. The governing equations with the exception of the gravity source term, as well as the details of the numerical method are presented in [17]. In RTI, small amplitude perturbations on the interface grow into mushroom-like structures. As reviewed in the literature [1, 18], experimental, theoretical and simulation-based studies have tried to explain the complex mechanism of fluid mixing, which is described as occurring in three stages: linear, non-linear and turbulent. In the linear stage, small perturbations grow exponentially as

$$a = a_0 \cosh(\phi t), \quad (2)$$

where $\phi = \sqrt{A_t k g}$ [19], where k is the wave number and the Atwood number $A_t = (\rho_h - \rho_l) / (\rho_h + \rho_l)$. Once the perturbation amplitude exceeds a tenth of the perturbation wavelength λ , it is expected that the growth obeys a non-linear power law, and flow features interact and merge to form larger coherent structures. Here, the effects of viscosity and diffusivity become significant; they inhibit the growth of small amplitude perturbations, and allow for the a particular most unstable wavelength to emerge, which for initially unperturbed interfaces is given by [5]

$$\lambda_m = 4\pi \left(\frac{\nu_m^2}{Ag} \right)^{1/3}, \quad (3)$$

where ν_m is, in our case, the kinematic viscosity He/Ar mixture. For the He/Ar RTI system in this study, we have that $\lambda_m = 39.9\mu m$. The value of the most unstable wavelength for helium gas is $\lambda_{m,1} = 109\mu m$ and argon gas is

$\lambda_{m,2} = 35\mu m$, at STP. These values were calculated using their individual kinematic viscosities. Once the amplitude gets large enough such that $ka \sim 1$, the flow becomes complicated and non-linear stability analysis or empirical analysis is used. One such empirical model for the growth of the bubble and spike amplitude is the Mikaelian model [16]. In the third stage, the coherent structures break up resulting in a self-similar turbulent mixing layer. In this regime, experimental [20] and simulation data [8] suggest that

$$a_{b,s} = \alpha_{b,s} A g t^2, \quad (4)$$

where α_b and α_s are the growth rate of the bubble and spike amplitude, respectively. The rate of growth of the mixing zone is $\alpha = \alpha_b + \alpha_s$, which has been estimated to be between 0.01 and 0.07 [8]. Typically, the amplitude and time scales are non-dimensionalized as $k(a - a_0)$ and $1/\sqrt{A_t g k}$, respectively. Further, non-dimensionalization has been applied to RTI mixing in [20], where the authors propose a perturbation Reynolds number Re_λ to describe the growth process, with

$$Re_\lambda = \frac{\lambda}{\nu_m} \sqrt{\frac{A_t}{1 + A_t}} g \lambda. \quad (5)$$

For the cases simulated in this work, Re_λ is listed in Table 1.

We consider a rectangular computational domain $[0, L_x] \times [-L_y/2, L_y/2]$ with X- and Y-direction lengths L_x and L_y , respectively. S.I. units are used throughout. The mesh spacing in both the X and Y directions is h and the number of cells are n_x and n_y , respectively. The light fluid (helium) is on the right side of the interface located at x_{int} . In all, three physical systems with decreasing order of magnitude of length scale labeled here as L1, L2 and L3 were considered, with initial setup parameters listed in Table 1. Two groups of simulation totaling 4 cases we considered. In group A, a single mode perturbation with amplitude $a = a_0 \cos(2\pi\lambda_p)$ was applied with wavelength $\lambda_p = L_y$, initial perturbation amplitude $a_0 = 0.01\lambda_p$, and wave number $k = 2\pi/\lambda_p$, such that $ka_0 = 0.06$. Group B simulations were started with an unperturbed interface. In group A, three systems with decreasing order of magnitude of length scale were considered. [The system width determines the length scale and here, for](#)

the systems in group A we choose system widths that were used in previously reported work that used DSMC simulations [10], so as to facilitate verification and comparisons against results therein. The simulation cases are listed in Table 2. For each of these cases, we conduct FCNS as well as CNS (no thermal fluctuations) simulations. In all simulations, the viscous scales are fully resolved. For FCNS simulations, the ensemble averaged metrics were obtained from n_{samp} number of simulation instances with n_{samp} estimated so as to ensure that the statistical error is small, specifically $\mathcal{O}(10^{-3}\lambda_p)$ for the amplitude at 95% confidence. The boundary conditions in all simulations are periodicity in Y and open in X. We ensure an initial hydrostatic equilibrium of the interface by imposing a pressure gradient $\partial P/\partial x = \rho g$. We conduct all our simulations with gravity values of $g = 10^7 \text{ ms}^{-2}$ for two reasons. First, it allowed us to verify our solver against DSMC results in [10] for the same g value. Second, because it enabled us to keep domain sizes large enough so as to achieve large simulation times to capture self-similar mixing behavior for the largest system L1 considered in this study, while at the same time ensuring that the pressure is not negative at the left edge of the domain due to the initial pressure gradient imposed. The amplitudes of spikes and bubbles denoted a_s and a_b , respectively, and the mean amplitude of the mixing layer denoted $a = (a_b + a_s)/2$ are calculated using thresholds on the span-wise spatial average of the mass fraction as described in [17]. The state of mixing Θ within the mixing layer is quantified as the ratio of molecular mixing length to the mixing amplitude a as described in [6].

3. Results

3.1. Verification

Here, we first verify our numerical method, which is described in [17], wherein it was shown to produce the correct equilibrium fluctuation spectrum for the He-Ar mixture. The FCNS numerical solution for Case AL1 to the RTI solution for an identical system in [10] that was obtained using the DSMC method for a gravity value of $g = 10^7 \text{ ms}^{-2}$. Our system size, heavy and light fluid species, as well as gravity values matched exactly to the DSMC study. We picked the

	System L1	System L2	System L3
L_x	4×10^{-3}	4×10^{-4}	4×10^{-5}
L_y	1×10^{-3}	1×10^{-4}	1×10^{-5}
n_x	4096	2048	1024
n_y	1024	512	256
h	9.76×10^{-7}	1.9×10^{-7}	3.9×10^{-8}
dt	8.0×10^{-9}	3.5×10^{-11}	8×10^{-13}
Re_λ	4147	131	4.2
n_{samp}	15	100	200

Table 1: Parameters of the different physical systems at the start of the simulation. The systems are categorized according to the physical length of domain in Y-direction (All units in S.I.).

Case	$g \text{ (ms}^{-1}\text{)}$	Interface type	Group
AL1	1×10^7	Single mode	L1
AL2	1×10^7	Single mode	L2
AL3	1×10^7	Single mode	L3
BL1	1×10^7	Flat	L1

Table 2: Simulation parameters at $t = 0$ of the five simulation cases. The cases are denoted based on their simulation group (A,B) prefixed by the system length designation (L1,L2,L3).

DSMC case for verification because of the ability of the method to naturally account for thermal fluctuations. The dimensions of the physical domain in Case AL1 are such that macroscopic mixing behavior is expected. Figure 1 shows the evolution of non-dimensional amplitude with non-dimensional time obtained from the FCNS simulations, linear model in Equation 2, Mikalean’s empirical formulae [16], and the DSMC results in [10]. All FCNS result shown are ensemble averaged results from $n_{samp} = 15$ number of instances. Error bars from the sampling for stochastic results are not shown because the error in amplitude is less than 10^{-8} m at 95% confidence. The DSMC results were only available up to a non-dimensional time value of 4. We see that the FCNS results compare well with linear theory that is valid at early times, and the Mikalean model at slightly later times. We see that for very early times, there is a slight discrepancy between the FCNS and DSMC results, and that there is agreement at times just before the non-dimensional time of 4. The discrepancy at early times is due to a a stronger role of diffusion in the DSMC simulation compared

Figure 1: Comparison of non-dimensional amplitude of mixing layer obtained from FCNS simulation (Case AL1) against DSMC simulation results in [10] and theoretical models in the literature.

to the FCNS, which is attributed to the differences in initial conditions and to the difference in the value of the perturbation Reynolds number Re_λ in the two cases, which are 2367 and $Re_\lambda = 4417$, respectively. The lower the value of Re_λ , the stronger the effect of diffusion [20]. The time evolution of our calculated values of the growth rates of the spike and bubble in Equation 4 are plotted in Figure 2. We see that in the asymptotic regime, $\alpha_s = 0.13$ and $\alpha_b = 0.06$, which compare well with the values of $\alpha_s = 0.125 \pm 0.01$, $\alpha_b = 0.06$, and $\alpha_s = 0.1$, $\alpha_b = 0.05$ reported for the three dimensional and two dimensional molecular dynamics simulations for Atwood numbers 0.867 and 0.81, respectively, in [13]. This agreement with MD results further verifies our numerical method.

3.2. Multi-mode RTI in flat interface

Here, we show that thermal fluctuations trigger multi-mode RTI. From Figure 3, which shows the log-log plot of the spike and bubble amplitude growth with time, we see that the flow enters the self-similar regime at late times that is characterized by the t^2 slope of the curve. We observe that the amplitude

Figure 2: Growth rate from FCNS solution of Case AL1.

evolution for the deterministic CNS solution suggests a predominantly diffusion-driven growth as expected, and that in the FCNS case, the fluctuations trigger self-similar growth at times after non-dimensional time of ≈ 7 . The value of the growth rates of the spike and bubble in Equation 4 for this Case are plotted in Figure 4. We find that the asymptotic value of growth rates for the FCNS case are $\alpha_s = 0.035$ and $\alpha_b = 0.023$, which are in the range of expected values 0.01-0.07 [8]. Figure 5 shows the comparison of mixing parameter Θ from FCNS and CNS solution. The mixing parameter quantifies the amount of mixing that is happening within the mixing zone [6]. We see that the fluctuations enhance mixing, whereas the mixing parameter remains a constant for the CNS result. Figure 6 shows the density field plots for different physical times for case BL1, for the stochastic (FCNS) numerical solutions. While the CNS solution is that of classical diffusion at the interface (not shown), the FCNS solution leads to the development of spikes and bubbles due to multi-mode perturbations that are triggered by the thermal fluctuations. The spectral content of these perturbations are such that the wavelength is close to the most unstable wavelength of $109 \mu m$ of the lighter gas helium. This result confirms that including thermal

Figure 3: Log-log plot of amplitude evolution for Case BL1.

fluctuations in the physical model of the system does lead to the capture of qualitative feature of the flow instability being triggered by the fluctuations. This result is further evidence of the claim that thermal fluctuations may trigger the onset of the first stage of mixing, i.e., govern the mechanisms that initiate the instability leading to the linear growth of the interface. It is in agreement with the MD simulations of RTI in [2] that demonstrated that at least for equally sized fluid particles, a long start-up is observed wherein there are spontaneous fluctuation-driven sites of mixing. Further, we see that at late times, the smaller features combine to form large features, which is the typical negative cascading observed in two dimensional RTI simulations [8].

3.3. Effect of fluctuations on mixing behavior

Here, we study the significance of thermal fluctuations in RTI-induced mixing. The amplitude and growth rate from FCNS simulations of RTI are compared to CNS simulations (no fluctuations present) of RTI, for the cases AL1, AL2 and AL3, which are of progressively smaller length scales that differ by an order of magnitude. Simulation of progressively smaller systems allows us to

Figure 4: Plot of growth rate α for FCNS and CNS (no thermal fluctuations) numerical solution for Case BL1 (flat interface).

Figure 5: Plot of mixing parameter Θ for FCNS and CNS (no thermal fluctuations) numerical solution for Case BL1 (flat interface).

Figure 6: Density evolution for FCNS numerical solution of Case BL1.

investigate systems with an at least theoretically increasing significance of thermal fluctuations, and thereby enables the investigation of when and if thermal fluctuations significantly affect mixing behavior. Figure 7 compares the bubble and spike amplitude from ensemble-averaged FCNS result to the deterministic behavior obtained from the CNS solution; the upper and lower bounds are also obtained from the extrema at each time step from among the several individual stochastic instances. We see that as the system size is reduced, the amplitude evolution is such that there is a progressively stronger role of diffusion at early times, which is consistent with the fact that the cases A1L1, A1L2, and A1L3 have decreasing order of magnitude of Re_λ as listed in 1. In addition, we see that for cases AL1 and AL2, deterministic behavior appears as the ensemble-averaged behavior, whereas for case AL3, departure from deterministic behavior is observed. As previously done for RMI in [17], we explain this departure from deterministic behavior in terms of the Boltzmann number. Table 3 lists the dimensionless numbers for the three fluctuating RTI systems simulated, that are obtained via non-dimensionalization of the governing equations. Here the

Reynolds number $Re = \rho_0 u_0 L_0 / \eta$, Prandtl number $Pr = \eta C_p / \kappa$, Mach number $M = u_0 / c_0$, the Schmidt number $Sc = \eta / \rho_0 D$, and the Froude number $Fr = u_0 / \sqrt{(g_0 L_0)}$ where ρ_0 , u_0 , L_0 , c_0 , g_0 are the chosen reference density, characteristic velocity scale, characteristic length scale, characteristic sound velocity, and body force (gravity) scale, respectively. Here, we choose ρ_0 as well as transport coefficients to be values of the He-Ar gas mixture with mass fraction $Y_1 = Y_2 = 0.5$ at STP. We choose U_0 to be the sound speed, and L_0 to be L_y . The transport coefficients are evaluated using the mass fraction and temperature dependent relationships for a binary gas mixture given in [12] that are also summarized in the appendix of [4]. The Boltzmann number is given by 1. Here, the Knudsen number alone is not a result of the non-dimensionalization of the governing equations, and is listed so as to ensure that we are within the limits of application of the continuum hypothesis. As we decrease the length, Reynolds and Peclet number decrease and Knudsen number increases in magnitude. FCNS is a coarse-grained model that captures hydrodynamic fluctuations and is therefore valid at high Re, high Pe and low Kn. The systems considered satisfy these criteria. The significance of thermal fluctuations is therefore determined by the Boltzmann number as defined in Equation 1. Thermal fluctuations are insignificant if $Bo \ll 1$ and are of increasing significance as Bo gets closer to 1. Table 3 lists the value of the dimensionless numbers for the three categories of physical systems used. Systems AL1, AL2 and AL3 are in decreasing order of magnitude of characteristic length and increasing order of magnitude of Bo . Here, we caution that this does not imply an inversely proportional relationship between the two quantities. It is possible that as the characteristic length of the system decreases, Bo also decreases, as reported in our previous study [17]. From the expressions for the non-dimensional parameters Re , Pr , Sc , and Bo , it can be concluded that Bo can be changed independent of the other three provided ρ_0 , $\rho_0 L_0$, L_0 and the transport coefficients are held constant. We see that Bo for Case AL3 is close to 1, which explains the departure from deterministic mixing behavior. In addition, the non-dimensional time of the simulation is less than 0.2 indicating linear stage growth, and in that regime, we see that there

is an enhanced role of fluctuation-driven diffusion. This is consistent with the analysis in [3], where the early time limit of the amplitude growth was shown to be proportional to $\sqrt{2t}$ with the proportionality constant depending on the intensity of the fluctuation, which is in our case, dependent on Bo .

Group	L1	L2	L3
Re	44602	4460	446
Pr	1.33	1.33	1.33
Sc	0.82	0.82	0.82
Bo	0.004	0.032	0.25
Kn	4.3×10^{-5}	4.3×10^{-4}	4.3×10^{-3}
Pe	36476	3648	365
Fr	7.2	22.8	72.1

Table 3: Non-dimensional parameters for the fluctuating RTI systems considered. The systems are categorized according to the length-based groups in Table 2. Values listed are for He-Ar mixture with specie fraction $Y_1 = Y_2 = 0.5$.

4. Conclusion

In conclusion, our main physical insight is that thermal fluctuations can trigger the onset of RTI at an initially unperturbed fluid-fluid interface and subsequently lead to mixing of multi-mode character, and presently this is demonstrated via numerical simulations of FCNS. In addition, we find that for systems with Boltzmann number much smaller than unity, the quantitative effect of thermal fluctuations on the mixing behavior is negligible and the behavior is the average of the outcome from several stochastic instances. However, when the system is such that the Boltzmann number is of order unity, we find that thermal fluctuations can significantly affect the mixing behavior; the ensemble-averaged solution shows a departure from the deterministic solution at late times. We conclude that for such systems, it is important to account for thermal fluctuations in order to correctly capture their physical behavior.

4.1. Acknowledgment

All simulations were performed on the CRAY XC-40 Shaheen-II at the KAUST Supercomputing Core Laboratory.

Figure 7: Amplitude evolution for fluctuating (FCNS) and deterministic (CNS) simulations of systems with progressively increasing Boltzmann number: (a) Case AL1 with $L_y = 10^{-3} \text{m}$ and $Bo=0.004$, (b) Case AL2 with $L_y = 10^{-4} \text{m}$ and $Bo=0.032$, (c) Case AL3 with $L_y = 10^{-5} \text{m}$ and $Bo=0.25$. FCNS results shown are ensemble averages. Bounds represent extrema from among all fluctuating instances at each simulation time step.

Appendix A. Governing Equations

We consider a binary mixture of two perfectly miscible ideal gases denoted as specie 1 and specie 2 of molecular mass m_1 and m_2 , respectively. We denote the total number of species in the mixture as $N_s = 2$ and the set of species indices as $S = [1, 2]$. The mass density of the mixture is ρ is the sum of individual specie densities, i.e., $\rho = \sum_{k \in S} \rho_k$. The mass fraction of the specie k is Y_k which is defined through $\rho_k = Y_k \rho$, with $\sum_{k \in S} Y_k = 1$. There are $N_s - 1 = 1$ independent specie mass densities and therefore for the binary mixture, we only consider the evolution of specie 1.

The fluctuating compressible Navier-Stokes equations for the mixture may be written in conservative form as

$$\mathcal{U}_t + \nabla \cdot \mathcal{F}_H = \nabla \cdot \mathcal{F}_G + \nabla \cdot \mathcal{F}_Z + \mathcal{F}_B, \quad (\text{A.1})$$

where $\mathcal{U} = \mathcal{U}(\mathbf{x}, t, \mathbf{N})$ is the vector of conserved quantities namely mass, momentum total energy and mass of specie 1, and is given by $\mathcal{U} = (\rho, \rho \mathbf{u}, E, \rho Y_1)^T$; $\mathbf{u}$ is the velocity and E is the total energy. The addition of a stochastic flux term makes the equations an SPDE system and therefore $\mathcal{U}$ is a multi-variate process. Here $\mathbf{x}$ is the spatial coordinate, t is the time and $\mathbf{N} \equiv \mathbf{N}(\mathbf{x}, t)$ is a Gaussian noise field which is formally the derivative of a Wiener sheet. $\mathcal{F}_H$, $\mathcal{F}_G$, $\mathcal{F}_B$, and $\mathcal{F}_Z$ are the hyperbolic or advective flux, diffusive or dissipative flux, flux due to body force (gravity), and stochastic or fluctuating flux, respectively. At equilibrium, a fluctuation-dissipation condition is satisfied when the system has reached a statistical steady state. It should be noted that the stochastic forcing in Equation A.1 is the divergence of white noise that models conservative, intrinsic, i.e., thermal fluctuations. We have that

$$\mathcal{F}_H = \begin{pmatrix} \rho \mathbf{u} \\ \rho \mathbf{u} \mathbf{u} + P \mathbf{I} \\ (E + P) \mathbf{u} \\ \rho \mathbf{u} Y_1 \end{pmatrix}, \mathcal{F}_G = \begin{pmatrix} 0 \\ \boldsymbol{\tau} \\ \boldsymbol{\tau} \cdot \mathbf{u} + \mathbf{q} + G \mathbf{f}_1 \\ \mathbf{f}_1 \end{pmatrix}, \mathcal{F}_B = \begin{pmatrix} 0 \\ \rho \mathbf{g} \\ \rho \mathbf{u} \cdot \mathbf{g} \\ 0 \end{pmatrix}, \quad (\text{A.2})$$

and the stochastic flux is given by

$$\mathcal{F}_Z = \begin{pmatrix} 0 \\ \tilde{\boldsymbol{\tau}} \\ \mathbf{u} \cdot \tilde{\boldsymbol{\tau}} + \tilde{\mathbf{q}} + G\tilde{\mathbf{f}}_1 \\ \tilde{\mathbf{f}}_1 \end{pmatrix}. \quad (\text{A.3})$$

Here, $\boldsymbol{\tau}$ is the shear stress, $\mathbf{q}$ is the heat flux vector, $\mathbf{f}_1$ is the barocentric diffusion flux with $\mathbf{f}_1 + \mathbf{f}_2 = 0$, $\tilde{\boldsymbol{\tau}}$, $\tilde{\mathbf{g}}$ is the acceleration due to gravity, and $\tilde{\mathbf{q}}$ are the stochastic stress tensor and heat flux, respectively, P is the thermodynamic pressure, and $\tilde{\mathbf{f}}_1$ is the stochastic diffusion flux of specie 1 with $\tilde{\mathbf{f}}_1 + \tilde{\mathbf{f}}_2 = 0$. Also, for a binary mixture of ideal gases we have that [4, 12]

$$G = S_T \frac{k_B T}{(m_1 Y_2 + m_2 Y_1)} + (C_{p1} - C_{p2}) T. \quad (\text{A.4})$$

Appendix A.1. Thermodynamic pressure

The pressure P is given by

$$P = \rho R_u T \sum_{k=1}^{N_s} \frac{Y_k}{w_k}, \quad (\text{A.5})$$

where $R_u = k_B N_A$ is the universal gas constant, where k_B is Boltzmann's constant and N_A is Avogadro's number; $w_k = m_k N_A$ is the molecular weight of specie k , where m_k is the molecular mass of that specie. The total energy is

$$E = \rho e(T, Y_k) + \frac{1}{2} \rho |\mathbf{u}|^2, \quad (\text{A.6})$$

where the internal energy per unit mass of the mixture is $e(T, Y_k) = \sum_{k=1}^{N_s} Y_k e_k(T)$, where $e_k = C_{v,k} T$, $C_{v,k}$ being the specific heat for constant volume of specie k . The enthalpy per unit mass of the mixture is given by $h(T, Y_k) = \sum_{k=1}^{N_s} Y_k h_k(T)$, where $h_k = e_k + R_u T / w_k$. The sound speed of the mixture is $c = \sqrt{\gamma P / \rho}$.

Appendix A.2. Deterministic diffusion fluxes

It can be shown that for the binary mixture of ideal gases that

$$\mathbf{f}_1 = \rho D \left(\nabla Y_1 + S_T Y_1 Y_2 \frac{\nabla T}{T} + (X_1 - Y_1) \frac{\nabla P}{P} \right), \quad (\text{A.7})$$

where the Soret coefficient S_T is usually assumed to be independent of concentration [4, 12] and D is the diffusion coefficient. The heat flux vector $\mathbf{q}$ is given by $\mathbf{q} = \kappa \nabla T$. The shear stress tensor τ is given by

$$\tau = \eta (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \left(\zeta - \frac{2}{3} \eta \right) (\nabla \cdot \mathbf{u}) \mathbf{I}, \quad (\text{A.8})$$

where η is the dynamic shear viscosity coefficient and ζ is the bulk viscosity coefficient. All transport coefficients are evaluated using the mass fraction and temperature dependent relationships for a binary gas mixture given in [12] that are also summarized in the appendix of [4].

Appendix A.3. Stochastic fluxes

$\tilde{\tau}$, $\tilde{\mathbf{q}}$, and $\tilde{\mathbf{f}}_k$ are Gaussian random fields with zero mean, i.e., $\langle \tilde{\tau} \rangle = 0$, $\langle \tilde{\mathbf{q}} \rangle = 0$, and $\langle \tilde{\mathbf{f}}_k \rangle = 0$ where $\langle \cdot \rangle$ denotes a suitable defined ensemble average. The cross-correlations are zero, i.e., $\langle \tilde{\tau}, \tilde{\mathbf{q}} \rangle = \langle \tilde{\tau}, \tilde{\mathbf{f}}_k \rangle = \langle \tilde{\mathbf{q}}, \tilde{\mathbf{f}}_k \rangle = 0$. The stochastic stress tensor can be written as

$$\tilde{\tau}(\mathbf{x}, t) = \sqrt{2k_B \eta T} \tilde{\mathbf{N}}^s + \left(\sqrt{\frac{k_B \zeta T}{3}} - \sqrt{\frac{2k_B \eta T}{3}} \right) Tr(\tilde{\mathbf{N}}^s) \mathbf{I}, \quad (\text{A.9})$$

where k_B is Boltzmann's constant, and $\tilde{\mathbf{N}}^s(\mathbf{x}, t) = \frac{(\mathbf{N}^s(\mathbf{x}, t) + (\mathbf{N}^s(\mathbf{x}, t))^T)}{\sqrt{2}}$ is a symmetric Gaussian random tensor field; the $\sqrt{2}$ in the denominator accounts for the reduction in variance due to averaging. The covariance of $\mathbf{N}^s(\mathbf{x}, t)$ is given by $\langle N_{ij}^s(\mathbf{x}, t) N_{kl}^s(\mathbf{x}', t') \rangle = \delta_{ij}^K \delta_{kl}^K \delta(\mathbf{x} - \mathbf{x}') \delta(t - t')$, i.e., it has zero mean and variance equal to 1, where δ is the Dirac delta and δ^K is the Kronecker delta. The stochastic diffusion flux of the k -th specie is given by

$$\tilde{\mathbf{f}}_1 = \sqrt{2D\rho[Y_1 Y_2 (m_1 Y_2 + m_2 Y_1)]} \mathbf{N}^{Y_1}, \quad (\text{A.10})$$

where $\mathbf{N}^{Y_1}(\mathbf{x}, t)$ is such that $\langle N_i^{Y_1}(\mathbf{x}, t) N_j^{Y_1}(\mathbf{x}', t') \rangle = \delta_{ij}^K \delta(\mathbf{x} - \mathbf{x}') \delta(t - t')$. The stochastic heat flux vector is a can be written as

$$\tilde{\mathbf{q}}(\mathbf{x}, t) = T \sqrt{2k_B \kappa} \mathbf{N}^q, \quad (\text{A.11})$$

where $\mathbf{N}^q(\mathbf{x}, t)$ is a white-noise random Gaussian vector field, whose covariance is given by $\langle N_i^q(\mathbf{x}, t) N_j^q(\mathbf{x}', t') \rangle = \delta_{ij}^K \delta(\mathbf{x} - \mathbf{x}') \delta(t - t')$.

- [1] Abarzhi, S. I., 2010. Review of theoretical modelling approaches of Rayleigh-Taylor instabilities and turbulent mixing. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* 368 (1916), 1809–1828.
- [2] Alda, W., Dzwinel, W., Kitowski, J., Moscinski, J., Pogoda, M., Yuen, D. A., 1998. Complex fluid-dynamical phenomena modeled by large-scale molecular-dynamics simulations. *Computers in physics* 12 (6), 595–600.
- [3] Barber, J. L., Kadau, K., Germann, T. C., Alder, B. J., 2008. Initial growth of the Rayleigh-Taylor instability via molecular dynamics. *European Physical Journal B* 64 (2), 271–276.
- [4] Bell, J., Garcia, A., Williams, S., 2010. Computational fluctuating fluid dynamics. *ESAIM: Mathematical Modelling and Numerical Analysis* 44, 1085–1105.
- [5] Chandrasekhar, S., 1961. *Hydrodynamic and magnetohydrodynamic stability*. Oxford University Press.
- [6] Cook, A., Dimotakis, P., 2001. Transition stages of Rayleigh-Taylor instability between miscible fluids. *Journal of Fluid Mechanics* 443, 69–99.
- [7] Detcheverry, F., Bocquet, L., 2012. Thermal fluctuations in nanofluidic transport. *Physical Review Letters* 109 (2), 1–5.
- [8] Dimonte, G., Youngs, D. L., Dimits, A., Weber, S., Marinak, M., Wunsch, S., Garasi, C., Robinson, A., Andrews, M. J., Ramaprabhu, P., Calder, A. C., Fryxell, B., Biello, J., Dursi, L., MacNeice, P., Olson, K., Ricker, P., Rosner, R., Timmes, F., Tufo, H., Young, Y. N., Zingale, M., 2004. A comparative study of the turbulent Rayleigh-Taylor instability using high-resolution three-dimensional numerical simulations. The Alpha-Group collaboration. *Physics of Fluids* 16 (5), 1668–1693.

- [9] Español, P., jan 1998. Stochastic differential equations for non-linear hydrodynamics. *Physica A: Statistical Mechanics and its Applications* 248 (1-2), 77–96.
- [10] Gallis, M. A., Koehler, T. P., Torczynski, J. R., Plimpton, S. J., 2016. Direct simulation Monte Carlo investigation of the Rayleigh-Taylor instability. *Physical Review Fluids* 043403 (1), 1–20.
- [11] Garcia, A. L., Mansour, M. M., Lie, G. C., Cementi, E., 1987. Numerical integration of the fluctuating hydrodynamic equations. *Journal of Statistical Physics* 47 (1-2), 209–228.
- [12] Hirshfelder, J., Curtis, C., Bird, R., 1954. *Molecular theory of gases and liquids*. John Wiley and Sons, Inc.
- [13] Kadau, K., Germann, T. C., Hadjiconstantinou, N. G., Lomdahl, P. S., Dimonte, G., Holian, B. L., Alder, B. J., apr 2004. Nanohydrodynamics simulations: an atomistic view of the Rayleigh-Taylor instability. *Proceedings of the National Academy of Sciences of the United States of America* 101 (16), 5851–5.
URL <http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=395886&tool=pmcentrez&rendition=abstract>
- [14] Kadau, K., Rosenblatt, C., Barber, J. L., Germann, T. C., Huang, Z., Carles, P., Holian, B. L., Alder, B. J., 2007. The importance of fluctuations in fluid mixing. *Proc. Nat. Acad. Sci. U.S.A.* 104, 7741–7746.
- [15] Livescu, D., 2013. Numerical simulations of two-fluid turbulent mixing at large density ratios and applications to the Rayleigh Taylor instability. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* 371 (20120185), 1–23.
- [16] Mikaelian, K. O., 2014. Solution to Rayleigh-Taylor instabilities : Bubbles , spikes , and their scalings. *Physical Review E* 89, 053009.

- [17] Narayanan, K., Samtaney, R., 2018. Appearance of deterministic mixing behavior from ensembles of fluctuating hydrodynamics simulations of the Richtmyer-Meshkov instability. *Physical Review E* 043111 (97), 1–17.
- [18] Sharp, D. H., 1984. An overview of Rayleigh-Taylor instability. *Physica D: Nonlinear Phenomena* 12 (1-3), 3–18.
- [19] Taylor, G. I., 1950. The instability of liquid surfaces when accelerated in a direction perpendicular to their planes. I. *Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences* 201 (1065), 192 LP – 196.
- [20] Wei, T., Livescu, D., 2012. Late-time quadratic growth in single-mode Rayleigh-Taylor instability. *Physical Review E* 046405 (October), 1–14.
- [21] Zhou, Y., 2017. Rayleigh Taylor and Richtmyer Meshkov instability induced flow , turbulence , and mixing . I. *Physics Reports* 720-722, 1–136.
URL <http://dx.doi.org/10.1016/j.physrep.2017.07.005>
- [22] Zhou, Y., 2017. Rayleigh Taylor and Richtmyer Meshkov instability induced flow , turbulence , and mixing . II. *Physics Reports* 723-725, 1–160.
URL <https://doi.org/10.1016/j.physrep.2017.07.008>