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Title: Generalized Cahn-Hilliard Navier-Stokes Equations for Numerical Simulations of Multicomponent Immiscible Flows

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Generalized Cahn-Hilliard Navier-Stokes Equations for Numerical Simulations of Multicomponent Immiscible Flows

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Objectives and Approach

- **Current numerical approaches and their limitations**
  - Sharp Interface Methods (zero interface thickness)
    - Front tracking
    - Volume of Fluid (VOF)
    - Level Set Method
  - Diffuse Interface Methods (non-zero interface thickness)
    - Conventional Cahn-Hilliard (CH) Method
  - Non-continuum Methods
    - Lattice-Boltzmann Method
- **Generalized Cahn-Hilliard Navier-Stokes equations**
  - Mathematical derivation of general equations (**physically consistent**) describing multi-component ($N \geq 2$) compressible flows from basic thermodynamics (CGCHNS-Compressible Generalized Cahn-Hilliard Navier-Stokes equations)
  - Rigorous derivation of general equations describing multi-component ($N \geq 2$) incompressible flows by taking the incompressible limit of the above compressible equations (IGCHNS-Incompressible Generalized Cahn-Hilliard Navier-Stokes equations)
  - Both CGCHNS and IGCHNS equations can describe flows with arbitrary density ratios, such as the Rayleigh-Taylor instability, and naturally handle complex topological changes of the interface.

Mathematical Development

❖**General Conservation Equations for Multi-Component Flows**

Continuity : $$\frac{\partial \rho}{\partial t} + \nabla \cdot \left(\rho \vec{V}\right) = 0$$

Momentum : $$\frac{\partial}{\partial t}\left(\rho \vec{V}\right) + \nabla \cdot \left(\rho \vec{V} \otimes \vec{V}\right) = \nabla \cdot \underline{\underline{\sigma}} + \rho \sum_{\alpha=1}^{N} Y_\alpha \vec{F}_\alpha$$

Energy : $$\frac{\partial}{\partial t}\left(\rho e\right) + \nabla \cdot \left(\rho e \vec{V}\right) = \underline{\underline{\sigma}} : \nabla \vec{V} - \nabla \cdot \vec{q} + \rho \sum_{\alpha=1}^{N} Y_\alpha \vec{F}_\alpha \cdot \vec{V}_\alpha$$

Species mass fraction : $$\frac{\partial}{\partial t}\left(\rho Y_\alpha\right) + \nabla \cdot \left(\rho Y_\alpha \vec{V}\right) = -\nabla \cdot \vec{J}_\alpha = -\nabla \cdot \left(\rho Y_\alpha \vec{V}_\alpha\right)$$

Entropy : $$\frac{\partial}{\partial t}\left(\rho s\right) + \nabla \cdot \left(\rho s \vec{V}\right) = -\nabla \cdot \vec{q}_s + \Delta_s$$

❖**Specific Helmholtz Free Energy Accounting for the Presence of Interfaces**

$$f = f^0\left(v, T, Y\right) + vAf^I\left(T, Y\right) + \frac{1}{2} v \sum_{\alpha=1}^{N} \lambda_\alpha\left(T\right)\left(\nabla Y_\alpha\right)^2$$

The expression can be derived, using statistical mechanics considerations, directly from the partition function.

Bulk contribution, related to the potential between like-molecules

Interface contribution, related to the repulsive potential between the immiscible components

First-order non-local contribution (Cahn-Hilliard), related to the attractive potential between like molecules.

Mathematical Development (*Cont.*)

❖ **General or Extended Thermodynamics**

$$e = e\left(v, s, Y_{1,2,\ldots,N}, \nabla Y_{1,2,\ldots,N}\right)$$

$$de = -Pdv + Tds + \sum_{\alpha=1}^{N} \mu_\alpha^0 dY_\alpha + \sum_{\alpha=1}^{N} \vec{\phi}_\alpha \cdot d\left(\nabla Y_\alpha\right)$$

- Replace **d*v*/dt, d*e*/dt, d(Y$_\alpha$)/dt** and **d*s*/dt** using continuity, energy, scalar and entropy equations into the above differential equation for **de**, together with mathematical relation $d\left(\nabla Y_\alpha\right)/dt = \nabla\left(dY_\alpha / dt\right) - \nabla Y_\alpha \cdot \nabla \vec{V}$
  Finally, we have

$$\Delta_s = \nabla \cdot \left(\vec{q}_s - \frac{1}{T}\vec{q} + \frac{1}{T}\sum_{\alpha=1}^{N} \vec{\phi}_\alpha \nabla \cdot \vec{J}_\alpha + \frac{1}{T}\sum_{\alpha=1}^{N} \mu_\alpha \vec{J}_\alpha \right) - \sum_{\alpha=1}^{N} \vec{J}_\alpha \cdot \nabla\left(\frac{\mu_\alpha}{T}\right) + \left(\vec{q} - \sum_{\alpha=1}^{N} \vec{\phi}_\alpha \nabla \cdot \vec{J}_\alpha\right) \cdot \nabla\left(\frac{1}{T}\right)$$

$$\frac{1}{T}\left[\underline{\underline{\sigma}}^r + P\underline{\underline{I}} + \rho\sum_{\alpha=1}^{N}\left(\nabla Y_\alpha \otimes \vec{\phi}_\alpha\right)\right] : \nabla\vec{V} + \frac{1}{T}\sum_{\alpha=1}^{N} \vec{F}_\alpha \cdot \vec{J}_\alpha + \frac{1}{T}\underline{\underline{\sigma}}^v : \nabla\vec{V}$$

where $\mu_\alpha = \mu_\alpha^0 - \nabla \cdot \left(\rho \vec{\phi}_\alpha\right)/\rho$ is the generalized chemical potential

- **Second-Law of thermodynamics** requires $\Delta_s \geq 0$ for any process. Since the viscous stress tensor should satisfy $\underline{\underline{\sigma}}^v : \nabla\vec{V} \geq 0$, after some manipulations, the condition can be written like this:

$$\left(\vec{q} - \sum_{\alpha=1}^{N} \vec{\phi}_\alpha \nabla \cdot \vec{J}_\alpha - \sum_{\alpha=1}^{N} \vec{J}_\alpha \mu_\alpha\right) \cdot \left(\frac{\nabla T}{T}\right) + \sum_{\alpha=1}^{N} \vec{J}_\alpha \cdot \left(\nabla \mu_\alpha - \vec{F}_\alpha\right) \leq 0$$

Mathematical Development (*Cont.*)

- With the generalized Helmholtz free energy (*f*), we can rewrite the inequality equation required by second-law of thermodynamics, after separating individual contributions to the chemical potential

$$\vec{J}_q \cdot \left(\frac{\nabla T}{T}\right) + \sum_{\alpha=1}^{N} \vec{J}_\alpha \cdot \left[\left(\nabla \mu_\alpha\right)_T - \vec{F}_\alpha\right] \le 0$$

where $\vec{J}_q = \vec{q} - \sum_{\alpha=1}^{N} \lambda_\alpha \left(\nabla Y_\alpha\right) \nabla \cdot \vec{J}_\alpha - \sum_{\alpha=1}^{N} \vec{J}_\alpha \left[\mu_\alpha^{02} - T\left(\partial \mu_\alpha^{02} / \partial T\right)_{v,Y,\nabla Y_\alpha}\right] - \sum_{\alpha=1}^{N} \vec{J}_\alpha \left[\mu_\alpha^{01} - T\left(\partial \mu_\alpha^{01} / \partial T\right)_{p,Y}\right]$ is the generalize heat flux. $\mu_\alpha^{01} = \left(\partial f^0 / \partial Y_\alpha\right)_{v,T,Y_{\beta \ne \alpha}}$ is the classical chemical potential and $\mu_\alpha^{02} = v\left[A \partial f^I \left(T, Y\right) / \partial Y_\alpha - \lambda_\alpha \nabla^2 Y_\alpha - \nabla \lambda_\alpha \cdot \nabla Y_\alpha\right]$ is the chemical potential related to the interface and Cahn-Hilliard contributions. The generalized chemical potential is $\mu_\alpha = \mu_\alpha^{01}\left(v,T,Y\right) + \mu_\alpha^{02}\left(v,T,Y,\nabla Y_\alpha\right)$

❖ **General Near-Equilibrium Solutions of the inequality equation**

$$\vec{J}_\alpha = -\sum_{k=1}^{N} a_{\alpha k} \left[\left(\nabla \mu_k\right)_T - \vec{F}_k\right] - D_\alpha^T \left(\frac{\nabla T}{T}\right) \quad \text{and} \quad \vec{J}_q = -\sum_{k=1}^{N} D_k^T \left[\left(\nabla \mu_k\right)_T - \vec{F}_k\right] - r\left(\frac{\nabla T}{T}\right)$$

(***Onsager relations***)

- The second law imposes restrictions on the values of the coefficients.
- After using basic thermodynamic relations, Onsager's symmetry principle, and a series of mathematical manipulations, finally we have the expression for species mass flux $\vec{J}_\alpha$ and heat flux $\vec{q}$

Mathematical Development (*Cont.*)

$$X_\alpha \frac{\left(\nabla a_\alpha\right)_{T,p}}{a_\alpha} = \sum_{\beta=1}^{N} \frac{X_\alpha X_\beta}{D_{\alpha\beta}}\left(\vec{V}_\beta^* - \vec{V}_\alpha^*\right) + \frac{p}{nR^0T}\left(1-\rho v_\alpha\right)Y_\alpha\left(\frac{\nabla p}{p}\right) + \left(\frac{\rho}{nR^0T}\right)\sum_{\beta=1}^{N} Y_\alpha Y_\beta\left(\vec{F}_\alpha - \vec{F}_\beta\right) + \sum_{\beta=1}^{N}\frac{X_\alpha X_\beta}{\rho D_{\alpha\beta}}\left(\frac{D_\beta^T}{Y_\beta} - \frac{D_\alpha^T}{Y_\alpha}\right)\left(\frac{\nabla T}{T}\right)$$

$$\vec{J}_\alpha = \vec{J}_\alpha^* - \sum_{k=1}^{N} a_{\alpha k}\left(\nabla \mu_k^{02}\right)_T \quad \text{and} \quad \vec{V}_\alpha^* = \vec{J}_\alpha^* / \left(\rho Y_\alpha\right)$$

$$\vec{q} = R^0 T \sum_{\alpha=1}^{N}\sum_{\beta=1}^{N} \frac{X_\beta D_\alpha^T}{W_\alpha D_{\alpha\beta}}\left(\vec{V}_\alpha^* - \vec{V}_\beta^*\right) + \sum_{\alpha=1}^{N}\vec{J}_\alpha\left[\mu_\alpha^{01} - T\left(\partial\mu_\alpha^{01}/\partial T\right)_{p,Y}\right] - \kappa\nabla T$$

$$+v\sum_{\alpha=1}^{N}\lambda_\alpha\cdot\left(\nabla Y_\alpha\right)\left(\nabla\cdot\vec{J}_\alpha\right) + \sum_{\alpha=1}^{N}\vec{J}_\alpha\left[\mu_\alpha^{02} - T\left(\partial\mu_\alpha^{02}/\partial T\right)_{v,Y,\nabla Y_\alpha}\right] - \sum_{\alpha=1}^{N} D_\alpha^T\left(\nabla\mu_\alpha^{02}\right)_T + Tv\sum_{\alpha=1}^{N}\vec{J}_\alpha\left[O\left(\partial f^I/\partial T\right) + O\left(d\lambda_\alpha/dT\right)\right]$$

- **Compressible Generalized Cahn-Hilliard Navier-Stokes Equations (CGCHNS)**

Continuity :

$$\frac{\partial\rho}{\partial t} + \nabla\cdot\left(\rho\vec{V}\right) = 0$$

Momentum :

$$\frac{\partial}{\partial t}\left(\rho\vec{V}\right) + \nabla\cdot\left(\rho\vec{V}\otimes\vec{V}\right) = -\nabla p + \sum_{\alpha=1}^{N}\rho\mu_\alpha^{02}\nabla Y_\alpha + \nabla\cdot\underline{\underline{\sigma}}^v + \rho\sum_{\alpha=1}^{N} Y_\alpha\vec{F}_\alpha + \left[O\left(\frac{\partial f^I}{\partial T}\right) + O\left(\frac{\partial\lambda_\alpha}{\partial T}\right)\right]$$

Energy :

$$\frac{\partial}{\partial t}\left(\rho e^0\right) + \nabla\cdot\left(\rho e^0\vec{V}\right) = -\nabla\cdot\left(\vec{q} - v\sum_{\alpha=1}^{N}\lambda_\alpha\nabla Y_\alpha\nabla\cdot\vec{J}_\alpha\right) - p\nabla\cdot\vec{V} + \sum_{\alpha=1}^{N}\mu_\alpha^{02}\nabla\cdot\vec{J}_\alpha + \underline{\underline{\sigma}}^v : \nabla\vec{V} + \sum_{\alpha=1}^{N}\vec{F}_\alpha\cdot\vec{J}_\alpha$$

$$+T\left[O\left(\partial f^I/\partial T\right) + O\left(\partial^2 f^I/\partial T^2\right) + O\left(d\lambda_\alpha/\partial T\right) + O\left(d^2\lambda_\alpha/\partial T^2\right)\right]$$

Species mass fraction :

$$\frac{\partial}{\partial t}\left(\rho Y_\alpha\right) + \nabla\cdot\left(\rho Y_\alpha\vec{V}\right) = -\nabla\cdot\vec{J}_\alpha$$

Mathematical Development (*Cont.*)

❖ **Incompressible Generalized Cahn-Hilliard Navier-Stokes Equations (IGCHNS)**

- The incompressible limit is obtained as the rigorous infinite sound of speed limit. Here, we choose $T \to \infty$ and $\partial p / \partial T \to const$ (**Livescu, Phil. Trans. R. Soc. A, 2013**). Also, the interfacial Helmholtz free energy loses temperature dependence, i.e. $f^I \to f^{I*}(Y)$ and $\lambda_\alpha \to \lambda_\alpha^*$
- The Continuity and Momentum equations maintain the same formulations as in compressible flows.
- The internal energy equation and species mass fraction equations are reduced to an identical expression for the velocity divergence :

$$\nabla \cdot \vec{V} = -\nabla \cdot \left[\sum_{\alpha=1}^{N} \left(\frac{\vec{J}_\alpha}{\rho_\alpha^*} \right) \right]$$

- For simplicity, assume ideal-gas equation of state (EOS). This is then reduced to

$$\frac{1}{\rho} = \sum_{\alpha=1}^{N} \frac{Y_\alpha}{\rho_\alpha^*}$$

where $\rho_\alpha^* = W_\alpha \left(\partial p / \partial T \right) / R^0$ is the micro-density of species α

- The number of species mass fraction equations needed to be considered is (N-2) and the formulation for the species mass flux is reduced to:

$$\vec{J}_\alpha = \vec{J}_\alpha^* - \sum_{k=1}^{N} a_{\alpha k} \left(\nabla \mu_k^{02} \right)_T$$

$$\nabla X_\alpha = \sum_{\beta=1}^{N} \frac{X_\alpha X_\beta}{D_{\alpha\beta}} \left(\vec{V}_\beta^* - \vec{V}_\alpha^* \right) \quad \text{and} \quad \vec{V}_\alpha^* = \vec{J}_\alpha^* / \left(\rho Y_\alpha \right)$$

Mathematical Development (*Cont.*)

- **Incompressible Cahn-Hilliard Navier-Stokes Equations for Binary Fluids**

(unlike previous studies, these equations allow arbitrary density ratios).

Continuity : $$\frac{\partial \rho}{\partial t} + \nabla \cdot \left(\rho \vec{V}\right) = 0$$

Momentum : $$\frac{\partial}{\partial t}\left(\rho \vec{V}\right) + \nabla \cdot \left(\rho \vec{V} \otimes \vec{V}\right) = -\nabla p + A\left(\frac{\rho_1^* \rho_2^*}{\rho_2^* - \rho_1^*}\right)\tilde{\mu} \nabla \left(\frac{1}{\rho}\right) + \nabla \cdot \underline{\underline{\sigma}}^v + \rho \vec{g}$$

Divergence condition: $$\nabla \cdot \vec{V} = -\nabla \cdot \left[D^m \nabla \ln \rho\right] + \left(\frac{\rho_2^* - \rho_1^*}{\rho_1^* \rho_2^*}\right) \nabla \cdot \left[\bar{D}^I \nabla \left(\frac{1}{\rho}\tilde{\mu}\right)\right]$$

Pure miscible flow: $\bar{D}^I = 0$ Pure immiscible flow: $D^m = 0$

- A simple model for the repulsive interface free energy $f^{I*}\left(Y\right)$ is a double-well function, for which the corresponding chemical potential is

$$\tilde{\mu} = Y_1\left(Y_1 - 1\right)\left(Y_1 - 1/2\right) - \bar{\lambda}\nabla^2 Y_1$$

- The species mass fraction equation for pure immiscible flow becomes:

$$\frac{dY_1}{dt} = \frac{1}{\rho}\nabla \cdot \left[\bar{D}^I \nabla \left(\frac{1}{\rho}\tilde{\mu}\right)\right]$$

For arbitrary density and mobility coefficient $\bar{D}^I$, the equilibrium condition is $\tilde{\mu}_{eq} = 0$

- **Sharp Interface analysis for Binary immiscible Fluids**
  - The species equation for immiscible flows has an near equilibrium solution which converges to the sharp interface equilibrium solution as the interface thickness goes to zero $\left(\varepsilon \rightarrow 0\right)$

Mathematical Development (*Cont.*)

$$Y_{eq} = \frac{1}{2}\left[1 + \tanh\left(\frac{\phi(\underline{x})}{\varepsilon}\right)\right] \qquad |\phi(\underline{x})| = 1 \qquad \varepsilon = 2\sqrt{2\bar{\lambda}}$$

Signed distance function

Parameter control interface thickness

- And the corresponding chemical potential is

$$\tilde{\mu}_{eq} = -\varepsilon \cdot \kappa(\phi)\left[1 - \tanh^2\left(\phi(\underline{x})/\varepsilon\right)\right]/16 \to 0 \quad as \quad \varepsilon \to 0$$

- By implementing the equilibrium solution into the incompressible Cahn-Hilliard Navier-Stokes equation, the divergence condition approaches the divergence free condition:

$$\nabla \cdot \vec{V} = O(\varepsilon) \to 0 \quad as \quad \varepsilon \to 0$$

- and the continuity equation for the equilibrium solution approaches the classical level set equation:

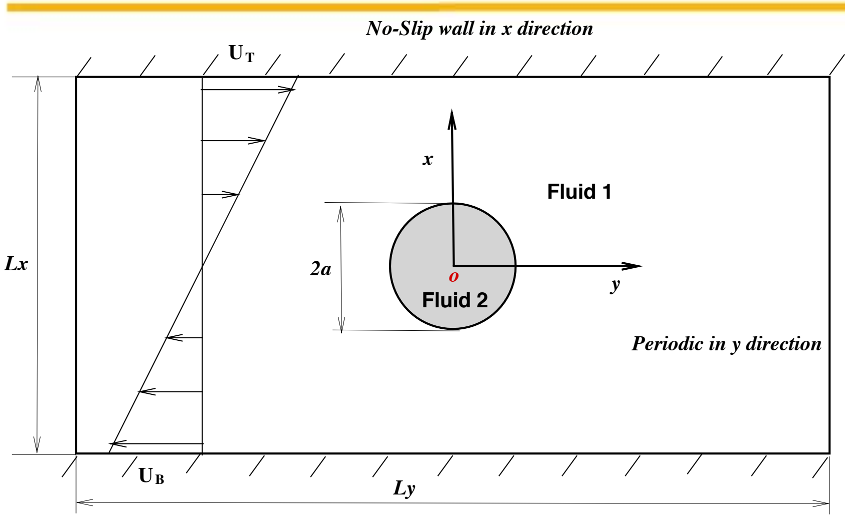
$$\frac{\partial \phi}{\partial t} + (\vec{V} \cdot \nabla)\phi = O(\varepsilon) \to 0 \quad as \quad \varepsilon \to 0$$

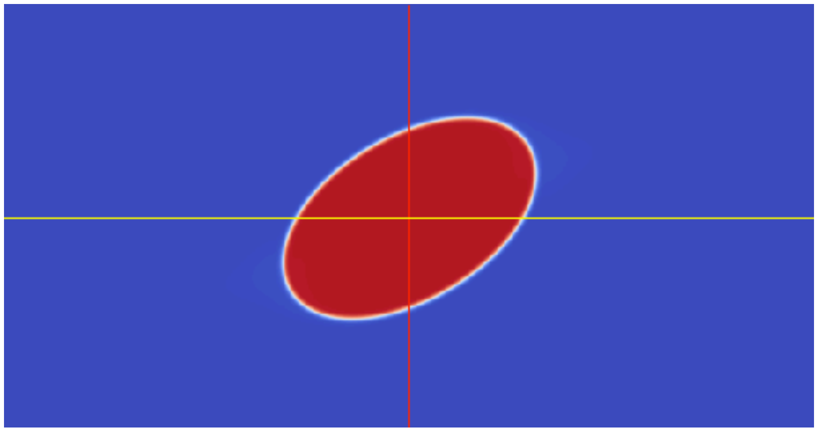
- For the near-equilibrium solution, the surface tension force in the momentum equation can be rewritten as:

$$\vec{F}_S = A\left(\frac{\rho_1^* \rho_2^*}{\rho_2^* - \rho_1^*}\right)\tilde{\mu}\nabla\left(\frac{1}{\rho}\right) = -T_S \cdot \delta_\varepsilon \cdot \kappa(\phi)\nabla\phi$$

where $\kappa(\phi) = \nabla \cdot \left(\nabla\phi / |\nabla\phi|\right)$ is the interface curvature and $\delta_\varepsilon = 3\left[1 - \tanh^2(\phi/\varepsilon)\right]/(4\varepsilon)$ is an approximation of the Dirac delta function with properties: $\int_{-\infty}^{+\infty} \delta_\varepsilon(\phi)\, d\phi = 1$

Drop Deformation in Shear Flow





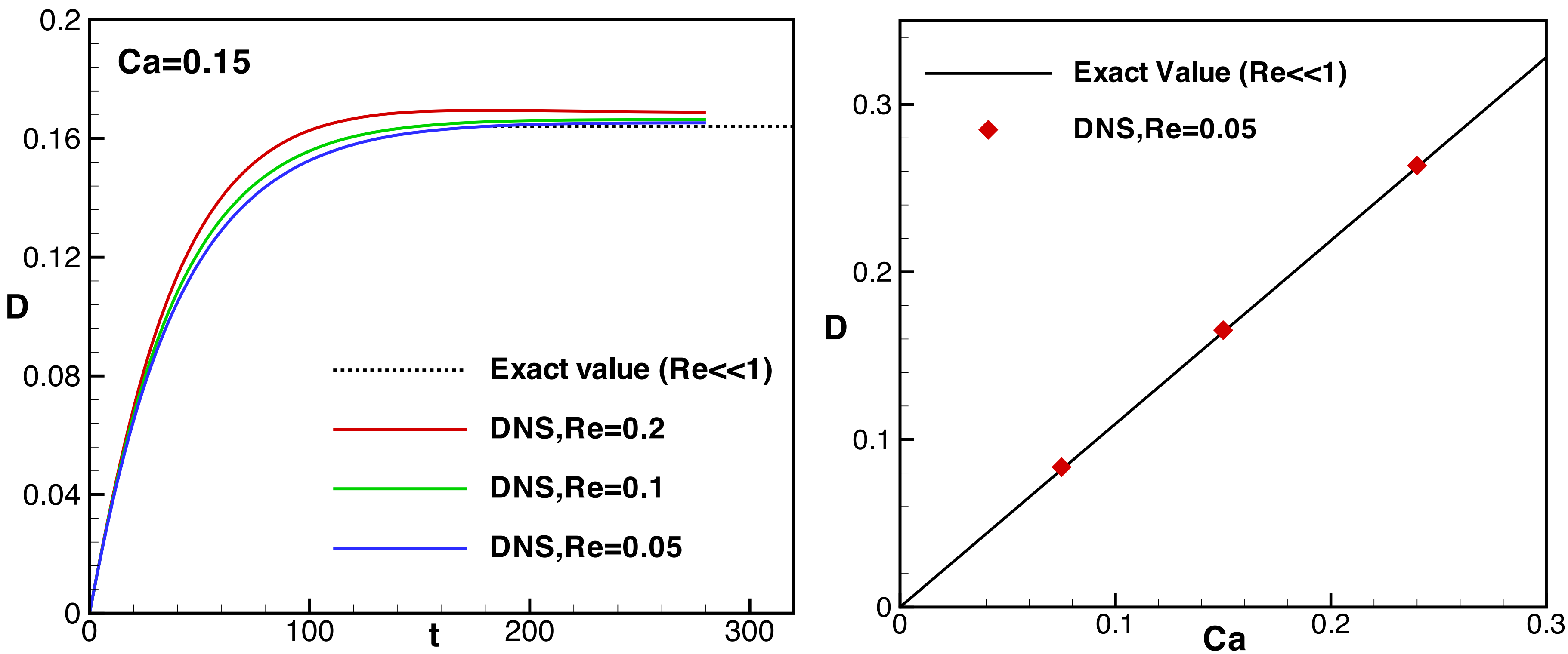
- For a given Reynolds number, there is a critical Capillary number Ca_{max} and the drop deforms and reaches a stationary deformed shape when $Ca < Ca_{max}$
- When the Reynolds is small enough ($Re << 1$), the flow becomes Stokes or creeping flow in which the drop deformation parameter D is related to the Capillary number Ca (**G. I. Taylor, Proc. R. Soc. Lond. A, 1934**) as:

$$D = \frac{L-B}{L+B} = \frac{19(\eta_2/\eta_1)+16}{16(\eta_2/\eta_1)+16} C_a$$

Where L and B are the longest and shortest axes of the ellipsoid (the steady deformed shape).

η_1 and η_2 are the viscosities of fluid 1 and fluid 2, respectively.

Drop Deformation (*Cont.*)



Incompressible & Immiscible Rayleigh-Taylor Instability

- **Linear Stability Theory (LST) for Rayleigh-Taylor Instability with zero-thickness**
  - If the initial perturbations of the interface are small, then, at early time, the mixing layer width grows exponentially with time

$$H(t) = h_0 \cosh\left[n(t-t_0)\right] + \frac{u_0}{n}\sinh\left[n(t-t_0)\right]$$

  - The exponential growth rate of immiscible Rayleigh-Taylor instability is given implicitly in an algebraic equation (**Chandrasekhar, Hydrodynamics and Hydromagnetic Stability, 1981**)

$$-\left\{\frac{gk}{n^2}\left[\left(\alpha_1-\alpha_2\right)+\frac{k^2 T_S}{g\left(\rho_1+\rho_2\right)}\right]+1\right\}\left(q-k\right)-\frac{4k^2 v}{n}\left(\alpha_1-\alpha_2\right)^2\left(q-k\right)+$$

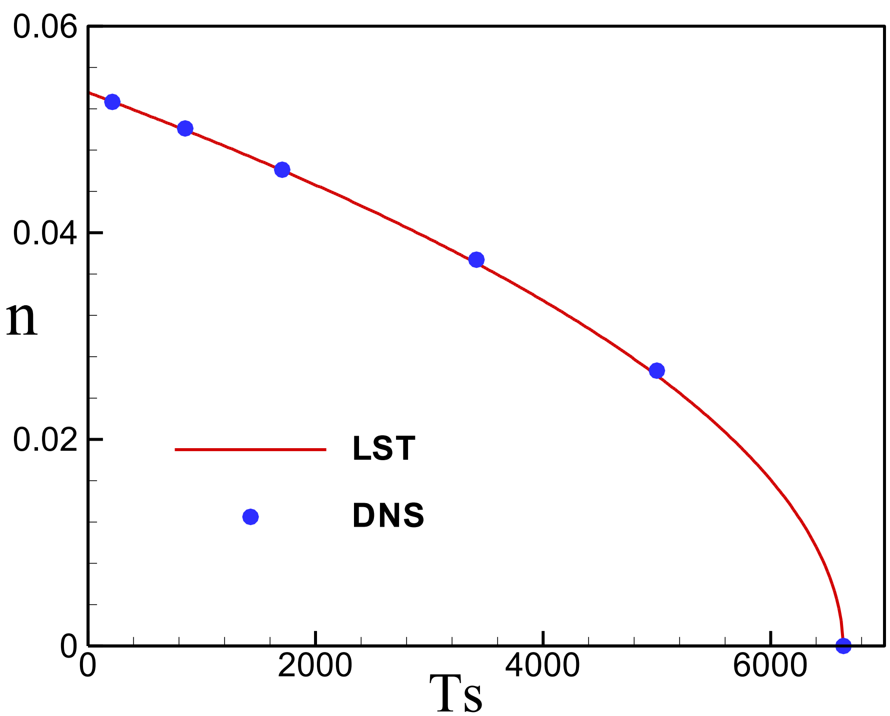
$$\frac{4k^3 v^2}{n^2}\left(\alpha_1-\alpha_2\right)^3\left(q-k\right)^2-4k\alpha_1\alpha_2=0$$

- **Direct Numerical Simulation (DNS) using incompressible Cahn-Hilliard Navier-Stokes equations**
  - Fixed Density ratio $\rho_2/\rho_1 = 3$ or Atwood number $A = 0.5$
  - The ratio of mesh grid in vertical direction to horizontal direction is $\Delta_v/\Delta_h = 0.8$ and the initial interface thickness is $\delta_0 = 8\Delta_v$
  - The resolution is chosen based on the condition: the ratio of interface thickness to wavelength

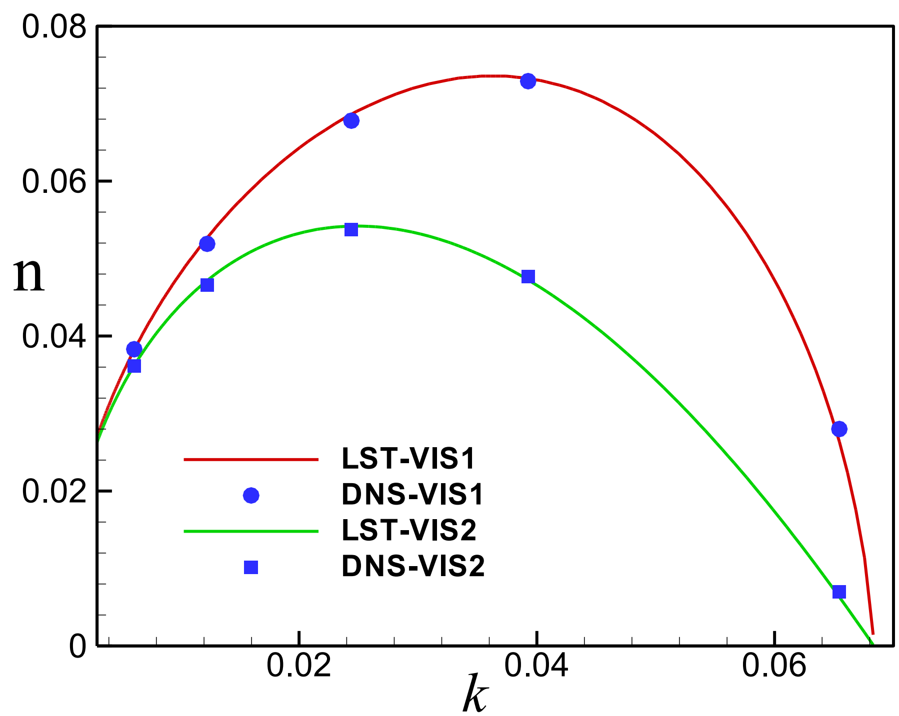
$$\delta_0/\lambda_L \le 1\%$$

Rayleigh-Taylor Instability

For fixed wavenumber *k* and viscosity *ν*

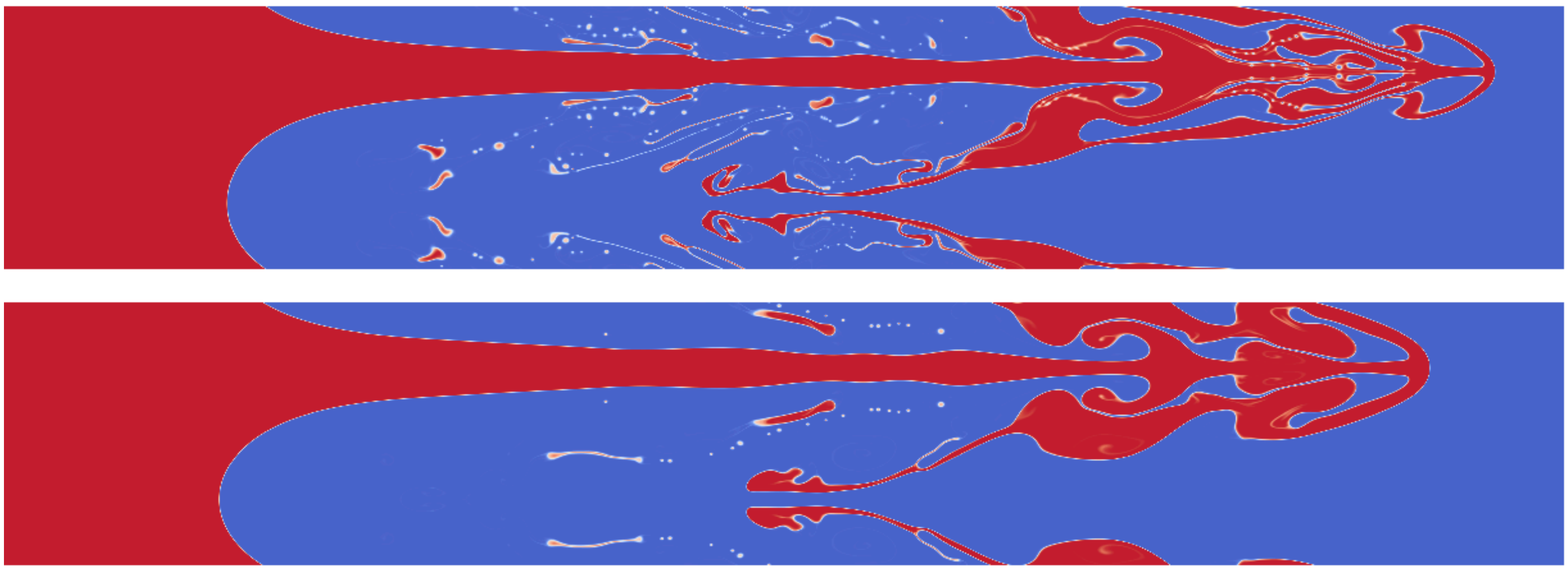


For fixed surface tension coefficient Ts



Rayleigh-Taylor Instability (*Cont.*)

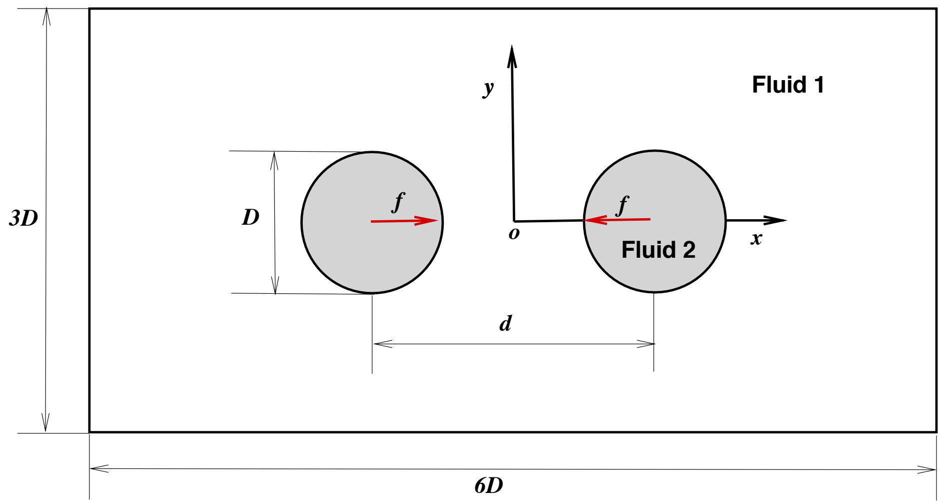
- **Late time Rayleigh-Taylor Instability results**



$T_S = 214$

$T_S = 856$

Head-on Coalescence of Drops

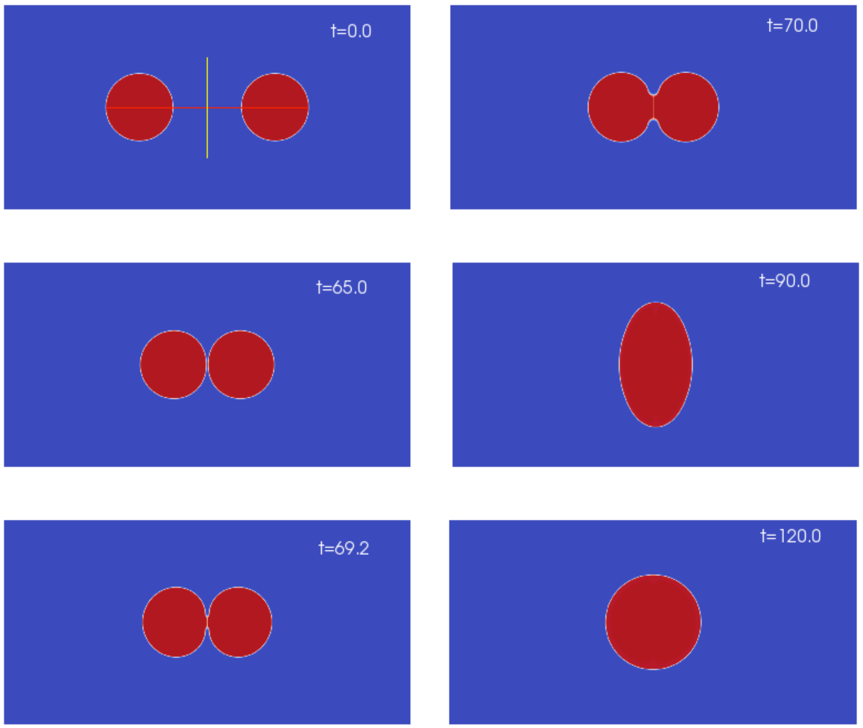


$\rho_2 / \rho_1 = 3.0$

- **Purpose**: **Show the capability of our approach in handling the singular topological changes during interface reconnection as well as breakup.**
- In reality, the rupture or reconnection of interface is the interplay between **repulsive** and **attractive forces between molecules** of the two drops, which are represented by ***f*^I** and **Cahn-Hilliard** terms in the **Helmholtz free energy (*f*)** given before.
- A external body force $f = -C_0 \rho sign(x - x_0)(1 - Y_1)$ is enforced to move the drops toward each other and turned off right before the drops approach together.

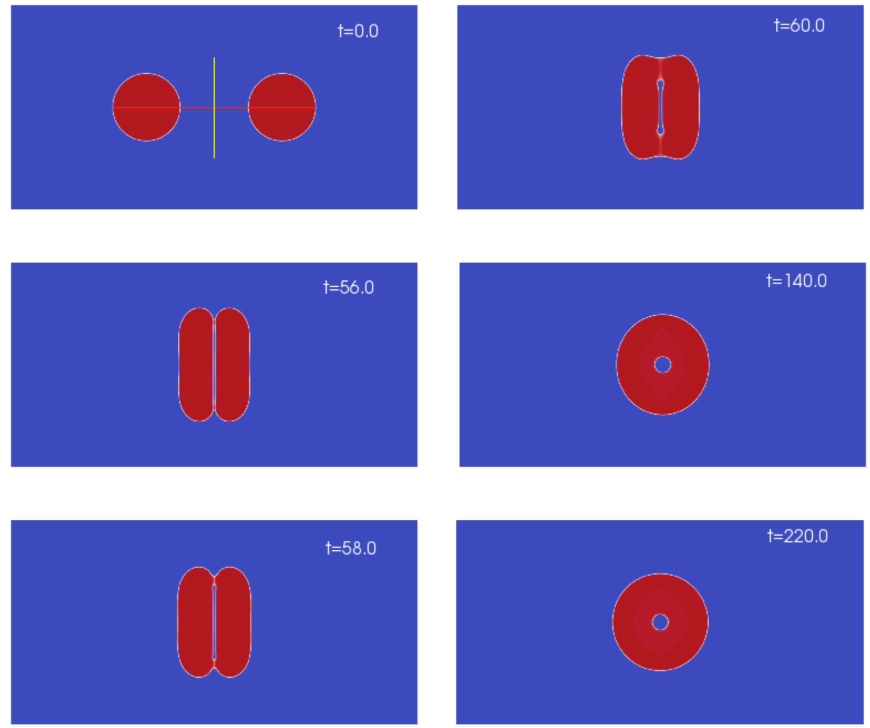
Head-on Coalescence of Drops Results

Re = 40.0, We=4.0



Previous studies with single point contact: **e.g. Nobari et al, Phys. Fluids, 1996, Front-tracking, artificial rupturing needed.**

Re = 40.0, We=16.0



Previous study with double-point contact: Yue et al, J. Fluid Mech., 2004, Diffuse-interface but for constant density only.

Non-diffuse interface cannot capture double point contact with trap of matrix fluid

Summary

- For the first time, the compressible generalized Cahn-Hilliard Navier-Stokes (CGCHNS) equations are derived from basic thermodynamics for multi-component (N ≥2) flows.
- For the first time, the incompressible generalized Cahn-Hilliard Navier-Stokes (IGCHNS) equations are rigorously derived as the incompressible limit of CGCHNS. The equations can address fluids with arbitrary density ratios.
- The Cahn-Hilliard Navier-Stokes equations can naturally handle complex interface deformation, including merging and breaking.
- Both the compressible and incompressible Cahn-Hilliard Navier-Stokes equations have been implemented into the CFDNS code.
- For the first time, extensive comparisons with Linear Stability Theory (LST) for the immiscible Rayleigh-Taylor instability are presented. Simulations using the incompressible Cahn-Hilliard Navier-Stokes equations reproduce the LST predictions. The equations have also been tested in several other immiscible flow problems.