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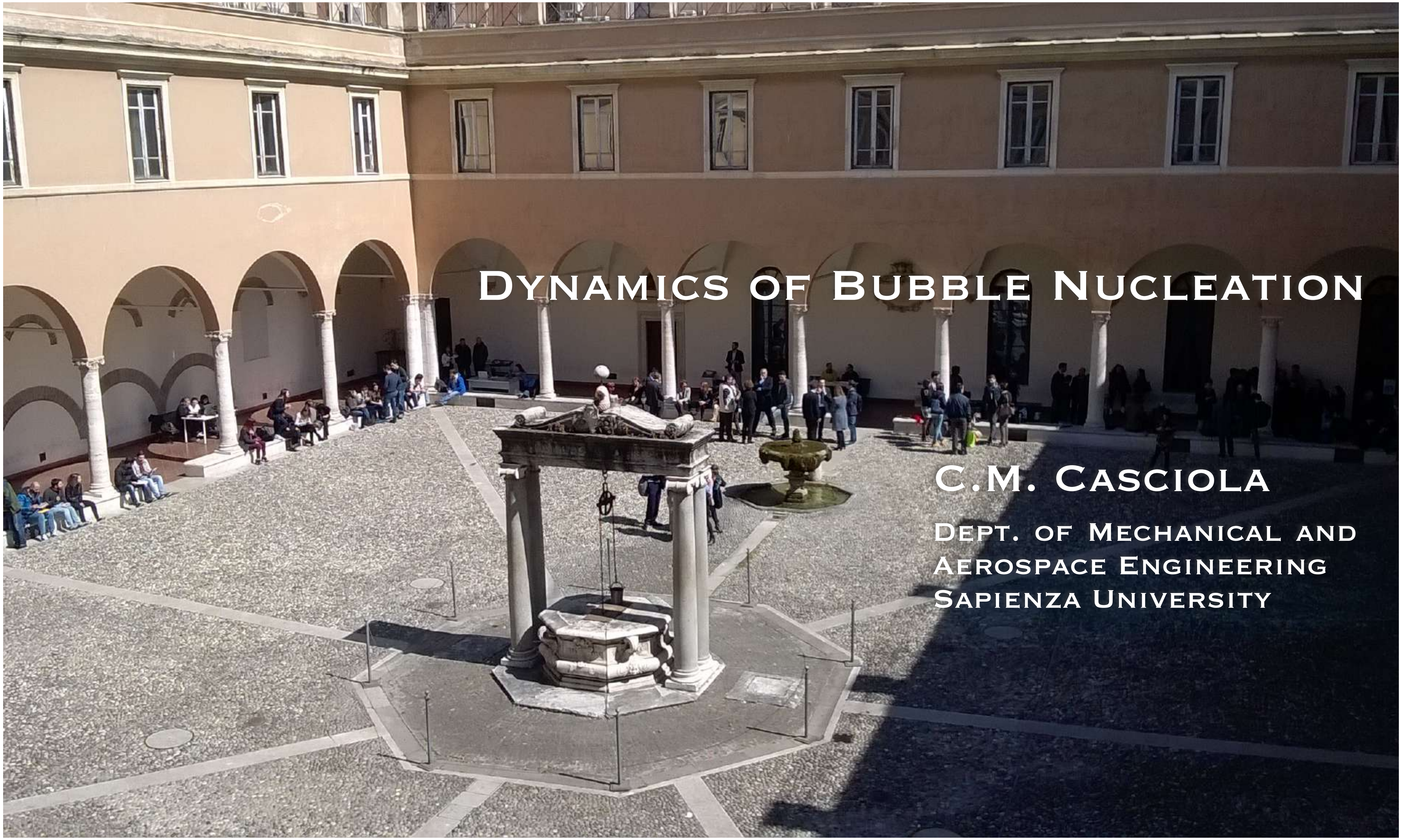

DYNAMICS OF BUBBLE NUCLEATION

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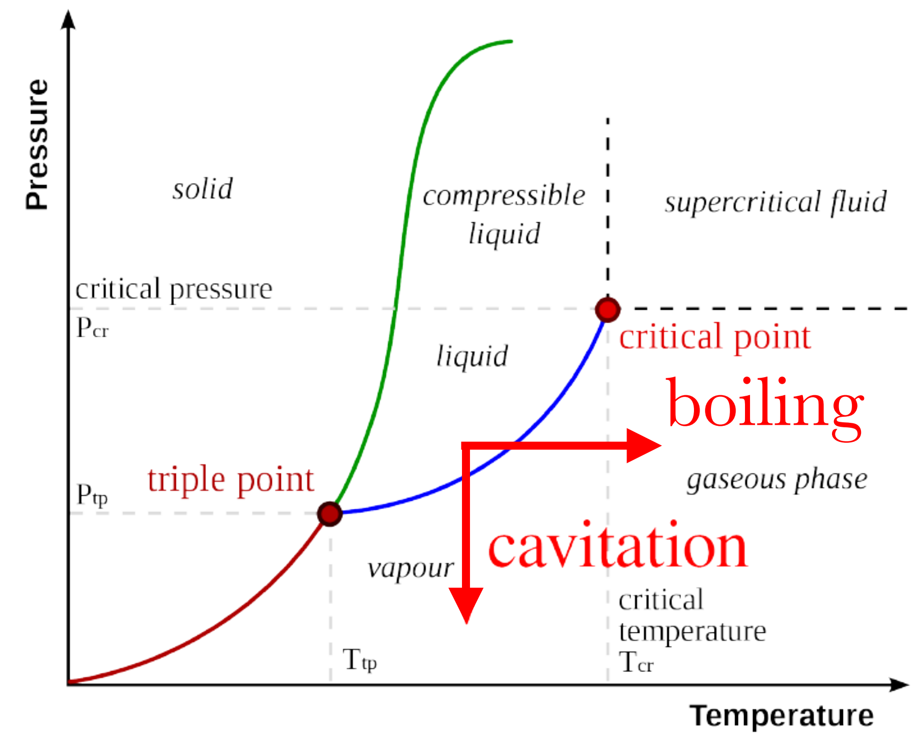
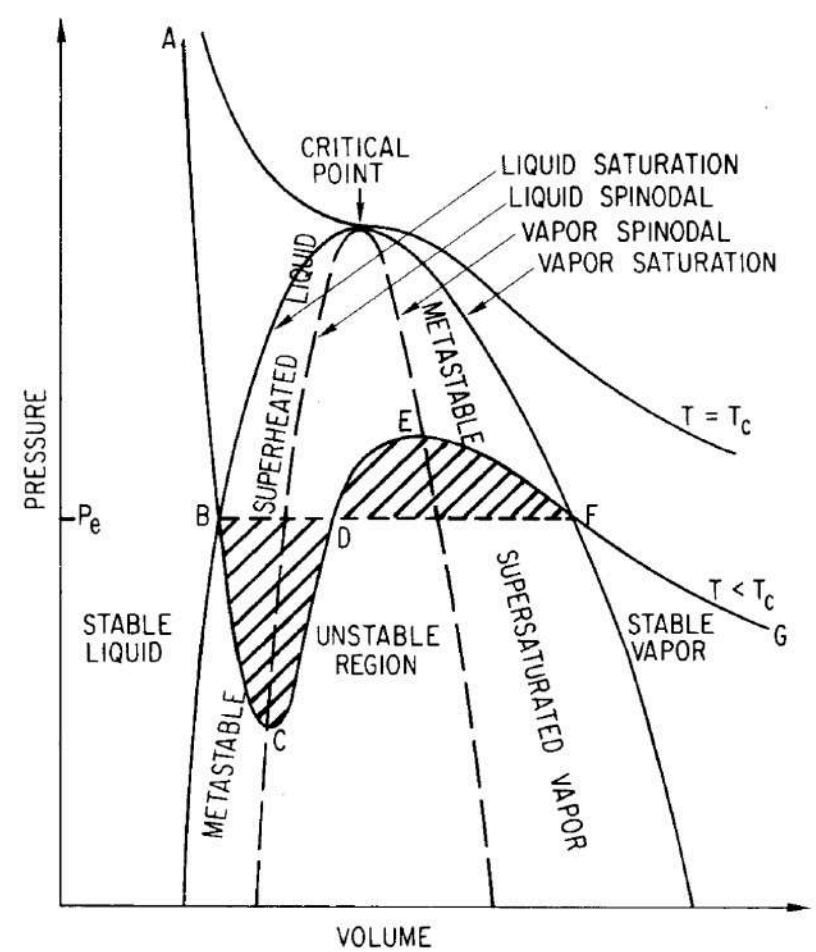
SAPIENZA UNIVERSITY

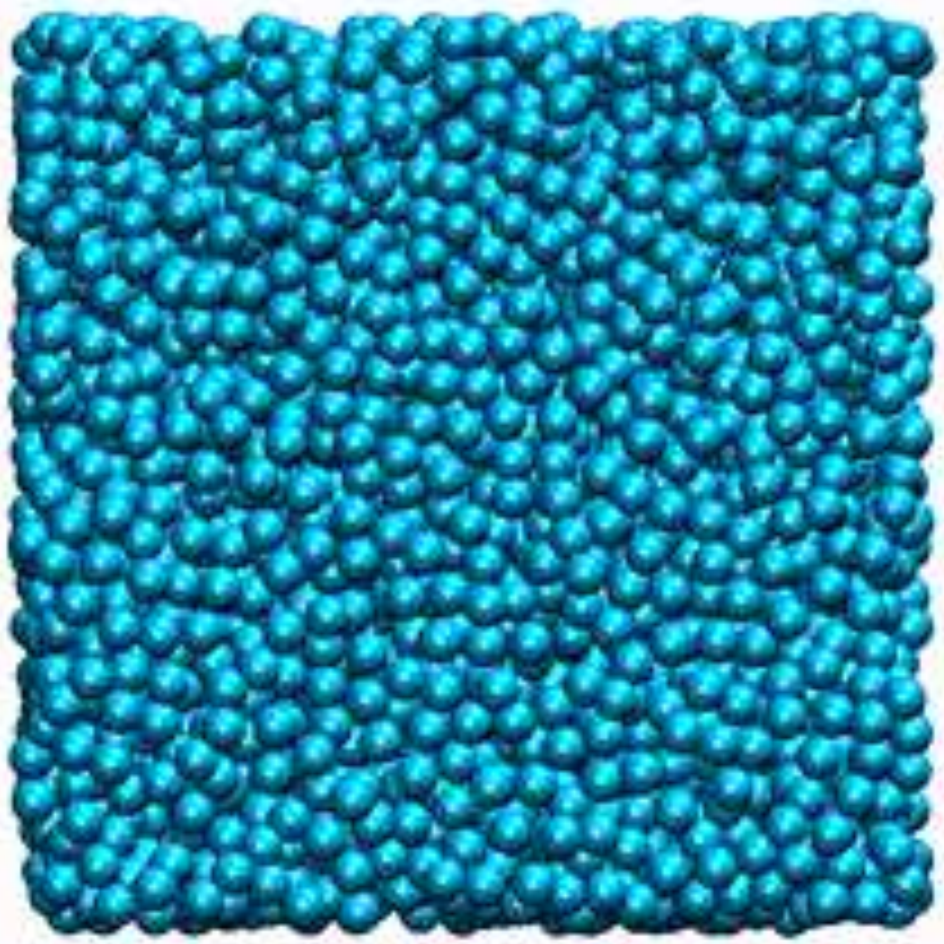
CaSToRC HPC National Competence Center Seminar Series
Cyprus, January 18th 2022

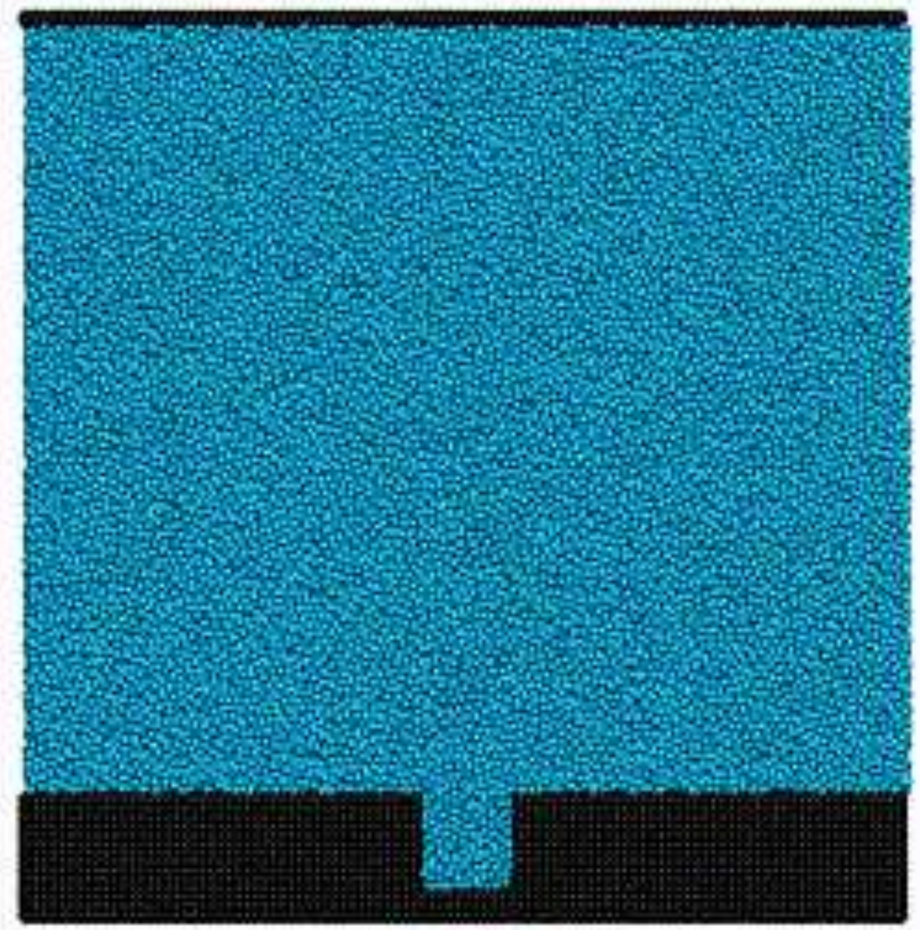
Introduction

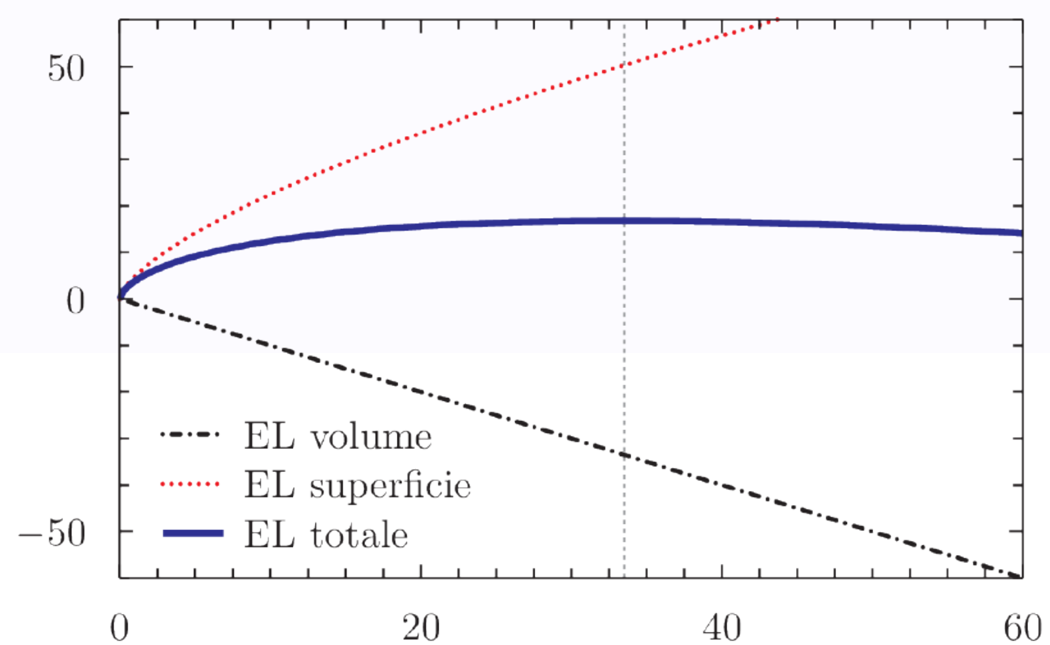




Bubble Nucleation







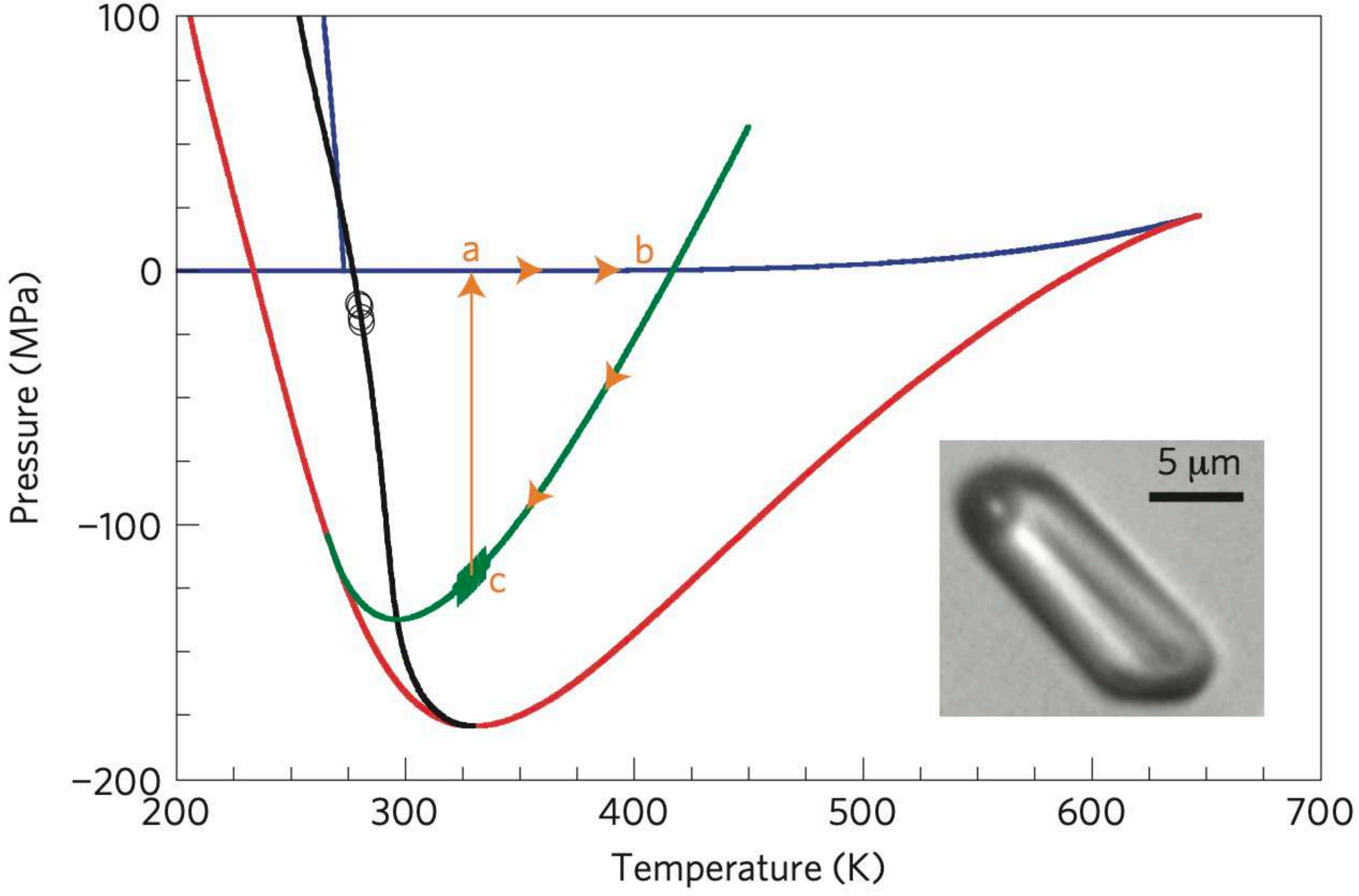
$$t = t_0 \exp\left(\frac{\Delta\Omega^{\dagger}}{k_B T}\right)$$

The Question is:

Can we bridge the gap between the (metastable) liquid and the successive (nonlinear) bubble dynamics phase, encompassing nucleation in the model?

EQUILIBRIUM

Experiments: Quartz Inclusions



Spinodal

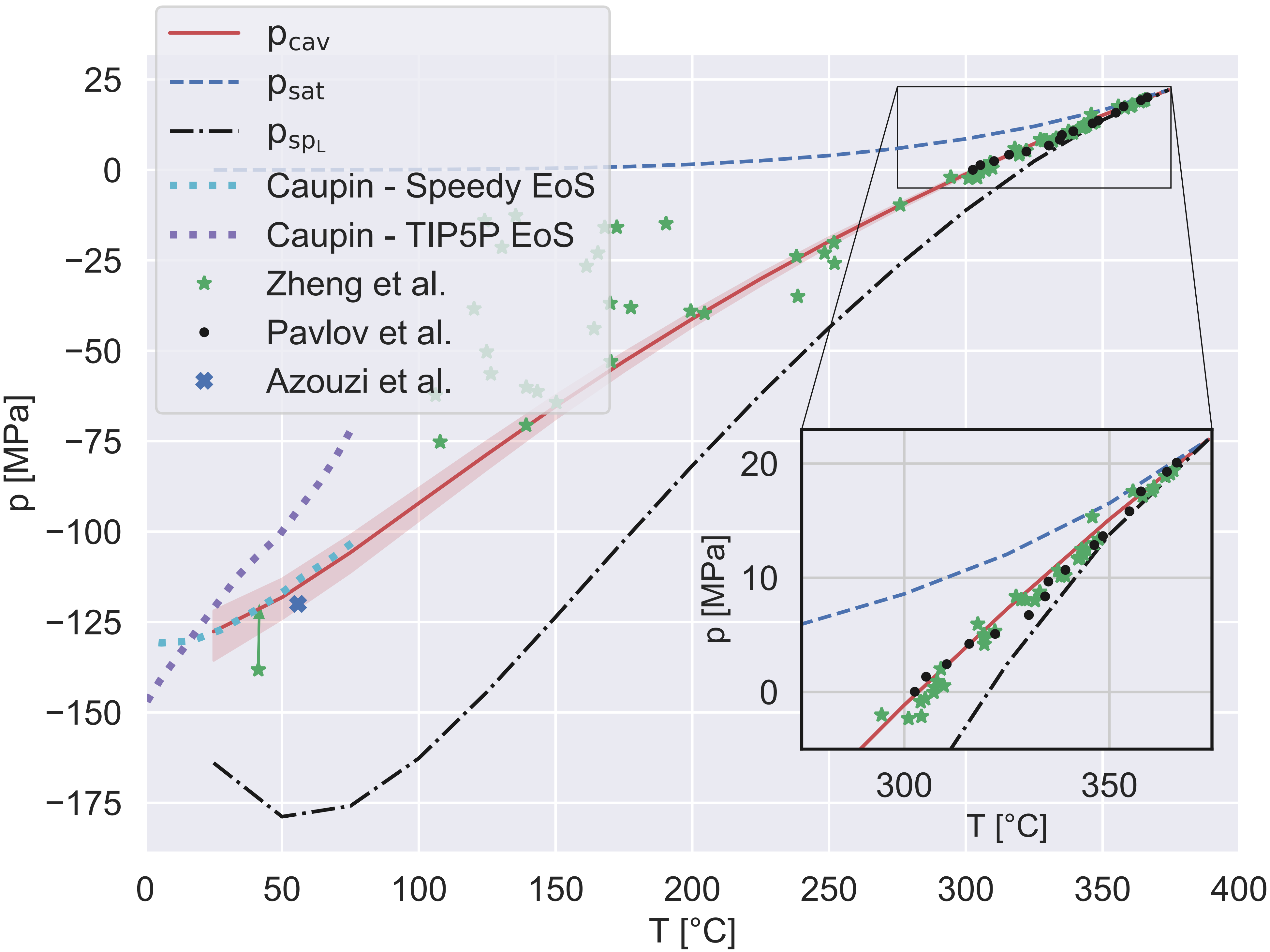
Isochore

Line Density Maxima

Liquid-Vapor Equilibrium

* Azouzi et al., Nat. Phy. 2012

Comparison with Experimental Data
Cavitation Pressure



VdW's Square Gradient Approximation

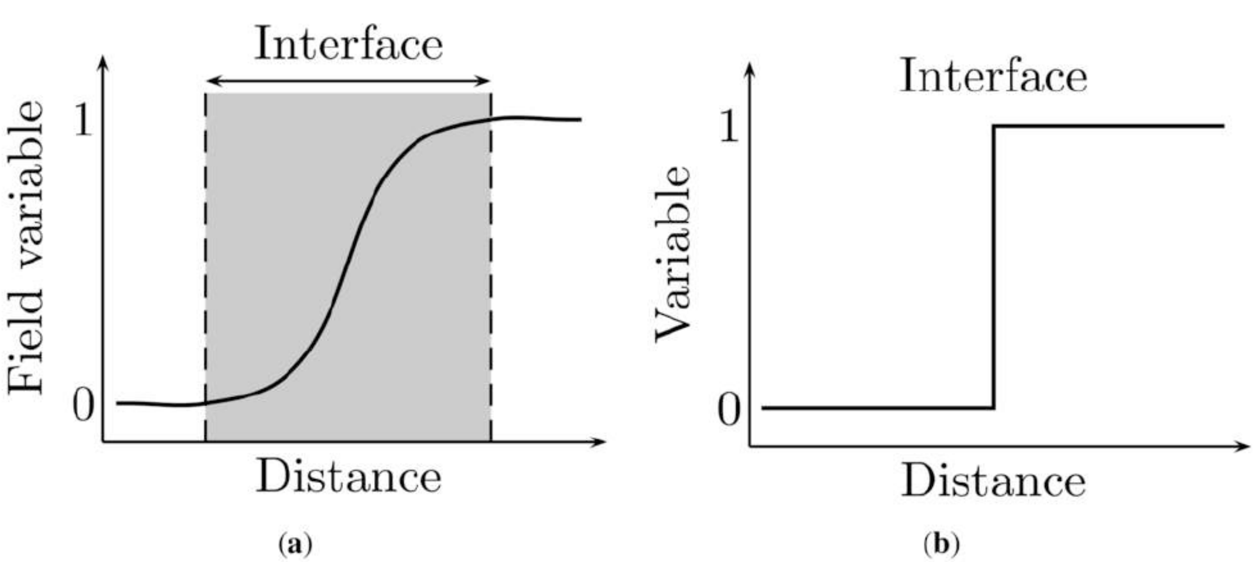
$$F[\rho,\theta] = \int_{\mathcal{D}} \hat{f}\, dV = \int_{\mathcal{D}} \left(\hat{f}_0(\rho,\theta) + \frac{\lambda}{2}|\nabla\rho|^2 \right) dv$$

Free-energy functional assumed to be given by two contributions

- bulk free-energy density of the homogeneous phases
- energy penalty associated with the interface

Minimization wrt density variations provides equilibrium conditions

$$\frac{\delta F}{\delta \rho} = \frac{\partial \hat{f}_0}{\partial \rho} - \nabla \cdot (\lambda \nabla \rho) = \mu_0$$



Free Energy Profile

In the transition between two (meta)stable states,

$$\rho_a(\mathbf{x}),\ \rho_b(\mathbf{x})$$

the system goes through a sequence of intermediate states
(a curve in the space of density fields)

$$\rho(\mathbf{x}, s),\ s \in [0, 1], \quad \rho(\mathbf{x}, 0) = \rho_a(\mathbf{x}),\ \ \rho(\mathbf{x}, 1) = \rho_b(\mathbf{x})$$

identifying a free energy profile

$$F(s) = \int_{\mathcal{D}} \left(\hat{f}_0(\rho(\mathbf{x}, s), \theta) + \lambda |\nabla \rho(\mathbf{x}, s)|^2 \right) dv$$

Minimum Free Energy Path

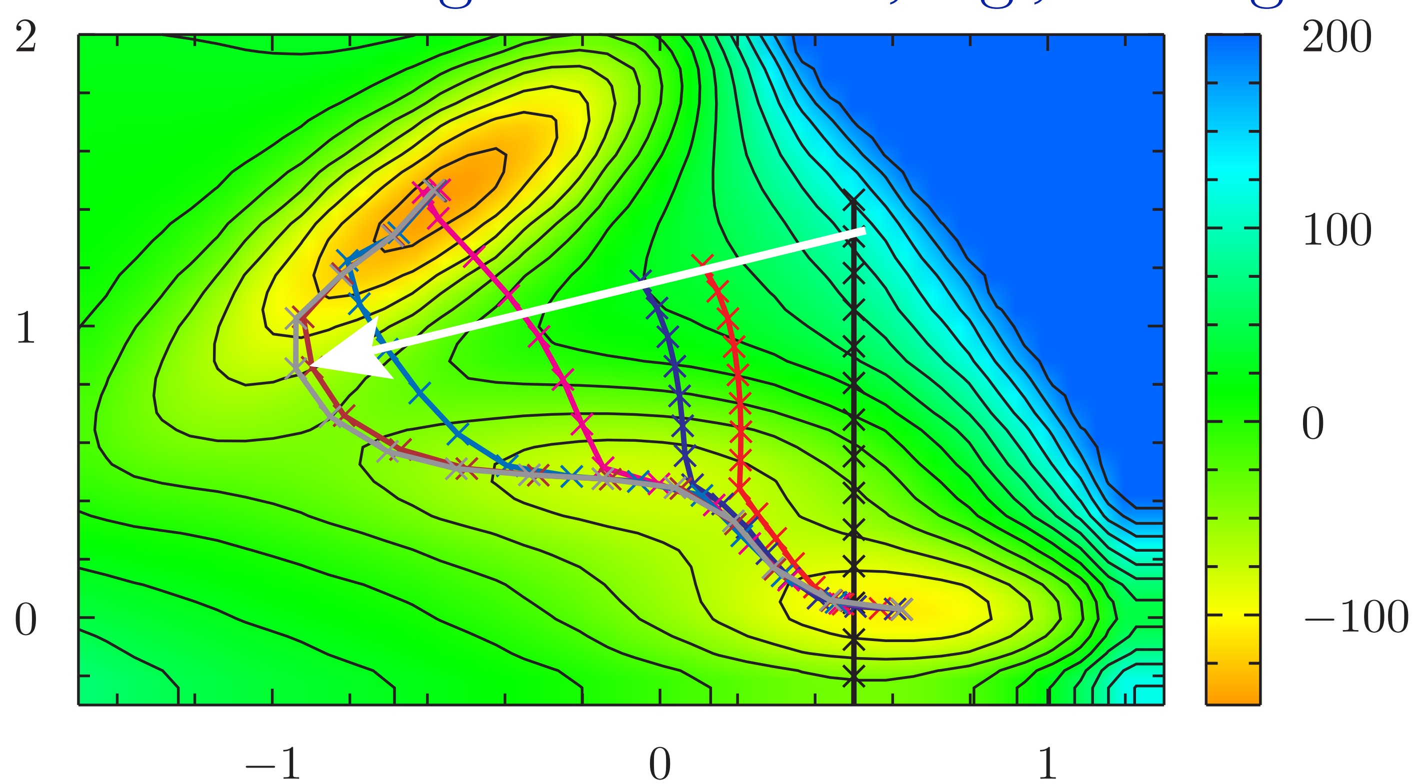
The Minimum Free Path is the curve

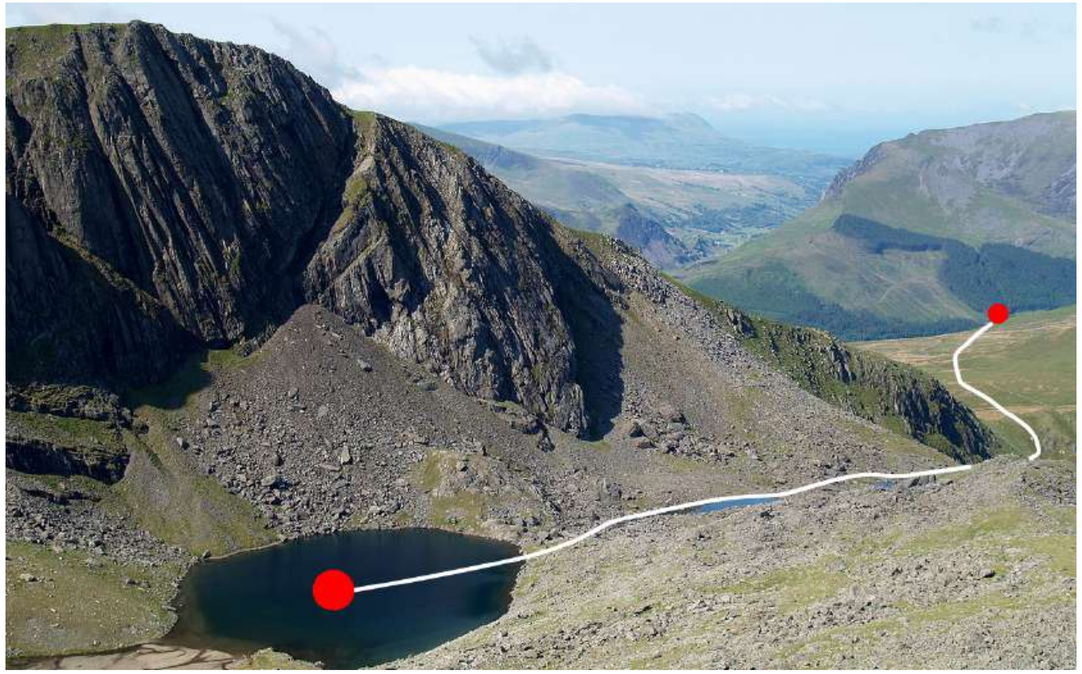
$$\rho(\mathbf{x}) = \rho_{MFEP}(\mathbf{x}, s)$$

that is everywhere tangent to free energy gradient

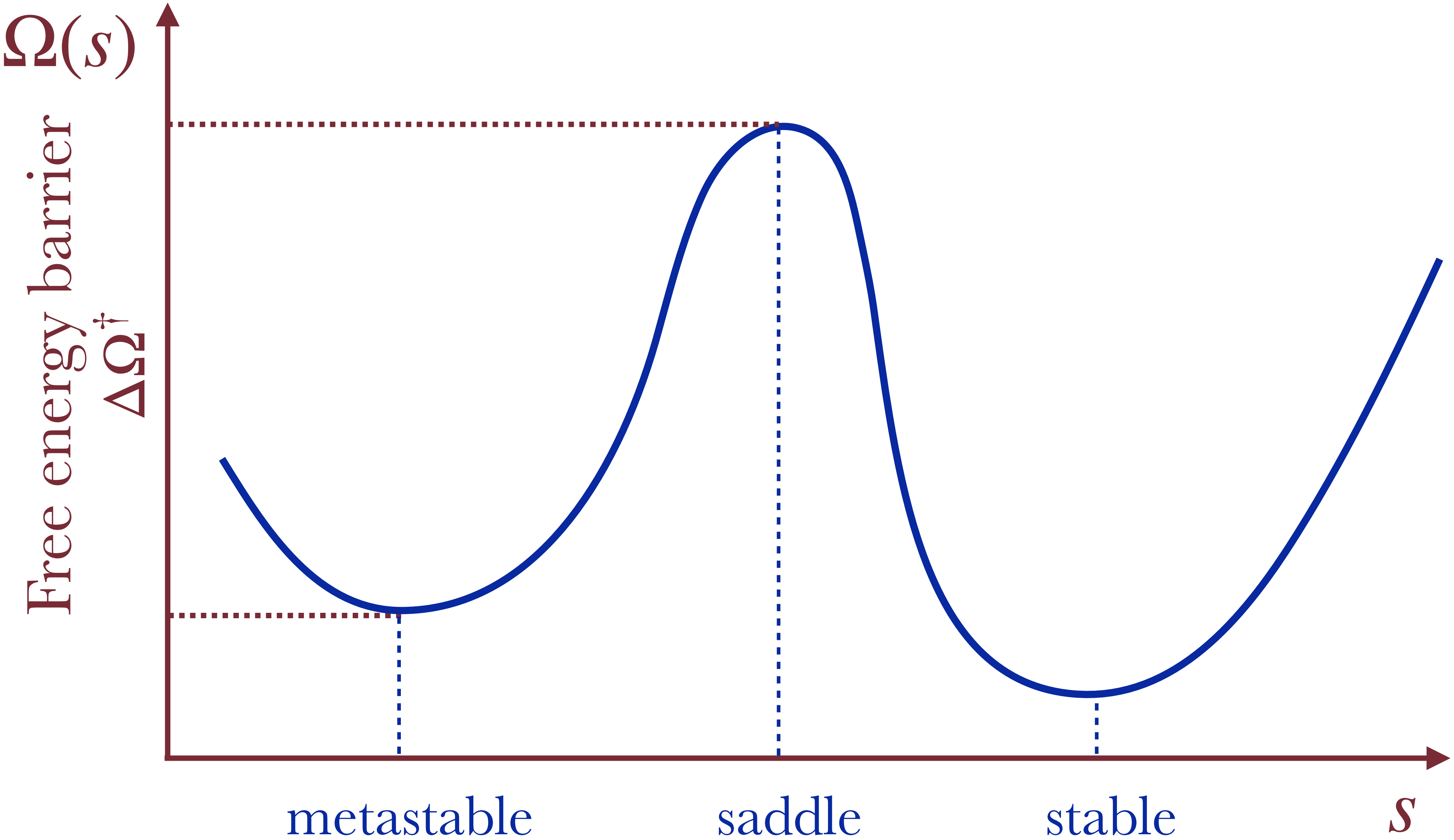
$$\left(\frac{\delta F[\rho(\mathbf{x}, s)]}{\delta \rho(s)} \right)^{\perp} = 0$$

String Method: see, e.g., Maragliano et al., J. Chem. Phys. 2006



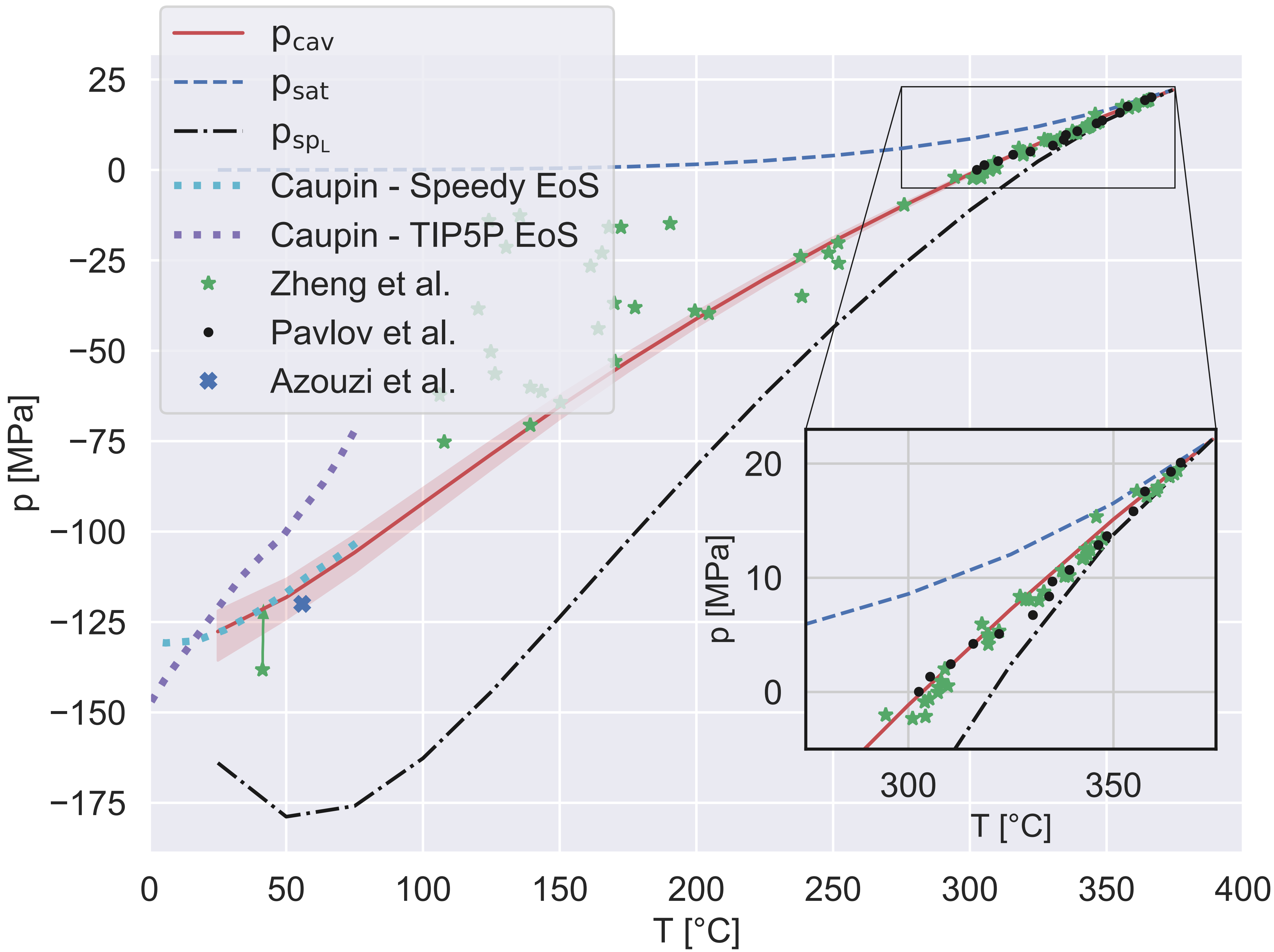


Free Energy Barrier



Transition time $\propto \exp(\Delta\Omega^\dagger / k_b\theta)$

Comparison with Experimental Data
Cavitation Pressure



Magaletti, Gallo, Casciola, Scientific Reports 2021

DYNAMICS

Governing Equations

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \qquad \text{(mass)}$$

$$\frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) = \nabla \cdot \boldsymbol{\tau} \qquad \text{(momentum)}$$

$$\frac{\partial E}{\partial t} + \nabla \cdot (\rho E) = \nabla \cdot (\mathbf{u} \cdot \boldsymbol{\tau} - \mathbf{q}_e) \qquad \text{(energy)}$$

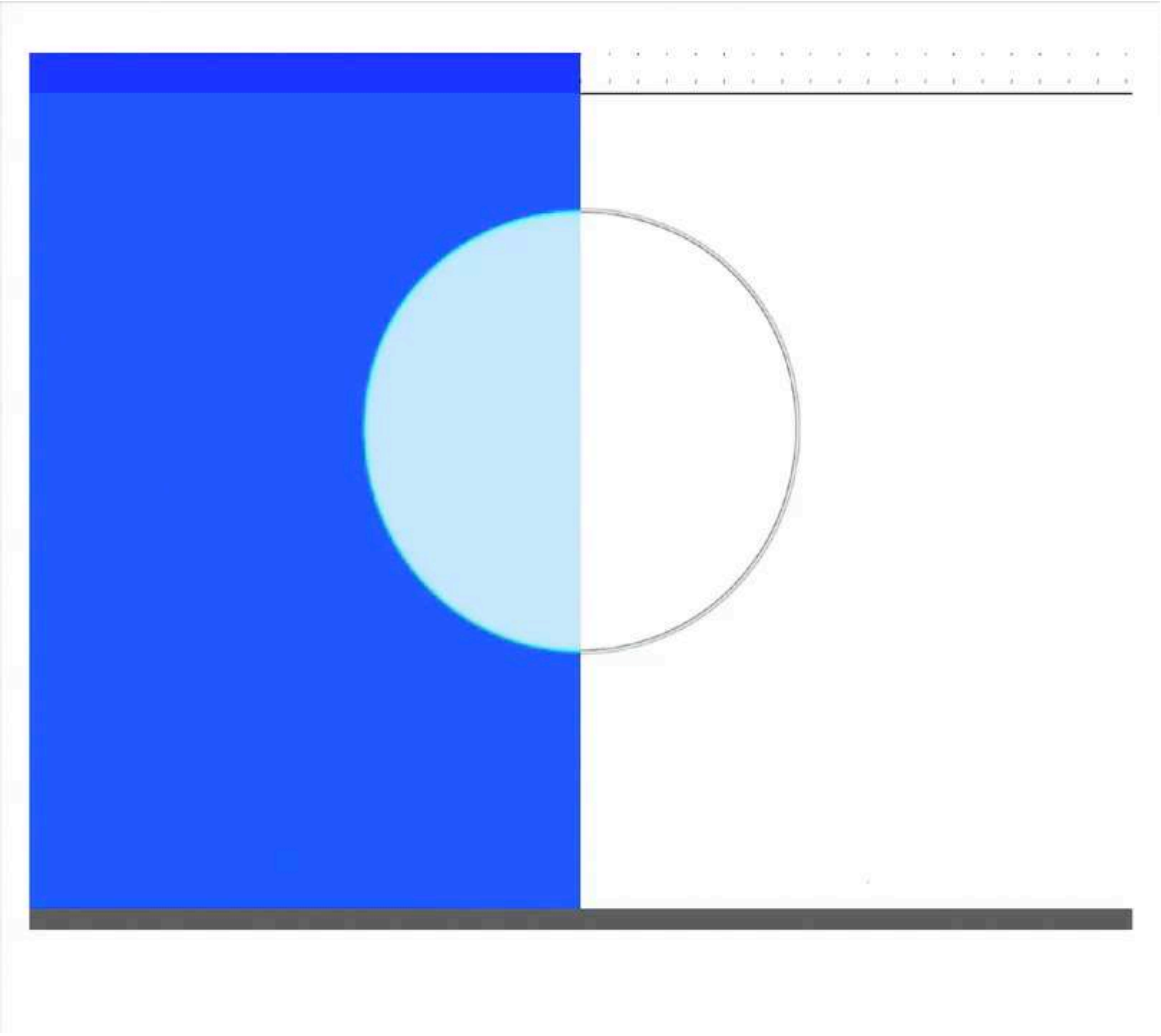
$$\boldsymbol{\tau} = -p_0 \boldsymbol{I} + \boldsymbol{\Sigma} = -p_0 \boldsymbol{I} + \mu \left[\left(\nabla \mathbf{u} + \nabla \mathbf{u}^T \right) - \frac{2}{3} \nabla \cdot \mathbf{u}\, \boldsymbol{I} \right] +$$

$$\boxed{\left[\frac{\lambda}{2} |\nabla \rho|^2 + \rho \nabla \cdot (\lambda \nabla \rho) \right] \boldsymbol{I} - \lambda \nabla \rho \otimes \nabla \rho}$$

Distributed capillarity

$$\mathbf{q}_e = \boxed{\lambda \rho \nabla \rho \nabla \cdot \mathbf{u}} - k \nabla \theta$$

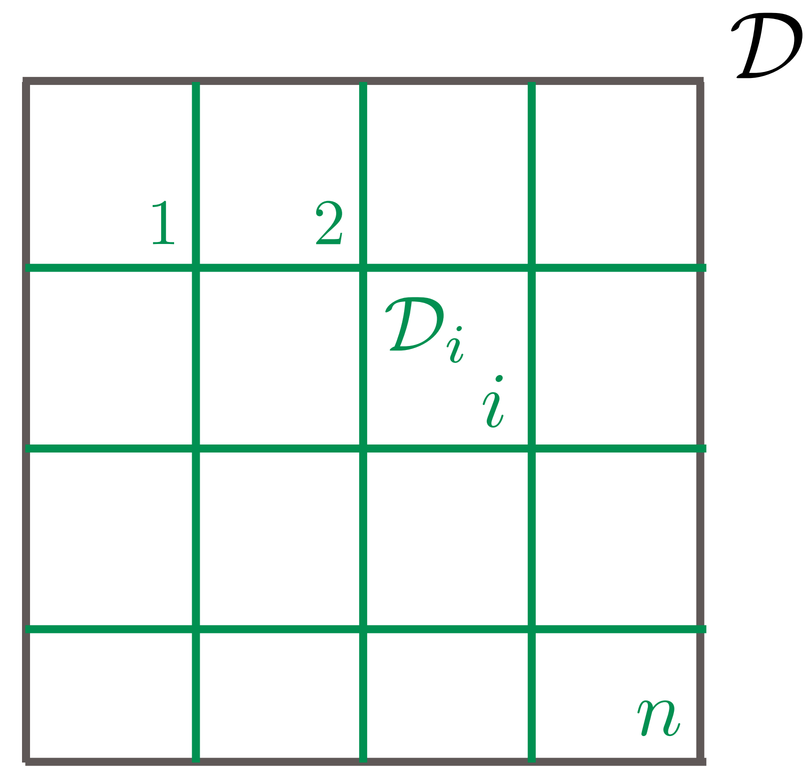
Diffuse Interface (Phase Field) Model



THERMAL FLUCTUATIONS

Coarse Graining & Collective Variables

Microcanonical system
(constant Energy, Volume, #particles)



$$f(\Gamma) = \frac{\delta\left[H(\Gamma) - E_T\right]}{N_T!\,\hbar^{3N_T} Z} \qquad \Gamma = (\mathbf{q}, \mathbf{p})$$

$$\Delta = (\delta\rho_1, \delta E_1, \ldots, \delta\rho_n, \delta E_n) \qquad \mathrm{p}(\Delta) = \langle \Delta - \hat{\Delta}(\Gamma) \rangle$$

Fluctuation wrt most probable state

Pdf of fluctuation

*

$$\mathrm{p}(\Delta) = \int f(\Gamma)\delta\left(\Delta - \hat{\Delta}(\Gamma)\right) d\Gamma = \exp\left(-\frac{S_0}{k_b}\right) \underbrace{\int d\Gamma\, \delta(E_0 - H(\Gamma))\Pi_{s=1}^{n}\delta(\rho - \hat{\rho}_s(\Gamma))\delta(E - \hat{E}_s(\Gamma))}_{\text{Fluctuation Entropy}}$$

Fluctuation Probability

$$p(\Delta) = \exp\left(\frac{S[\rho, E] - S_0}{k_b}\right)$$ Einstein-Boltzmann principle

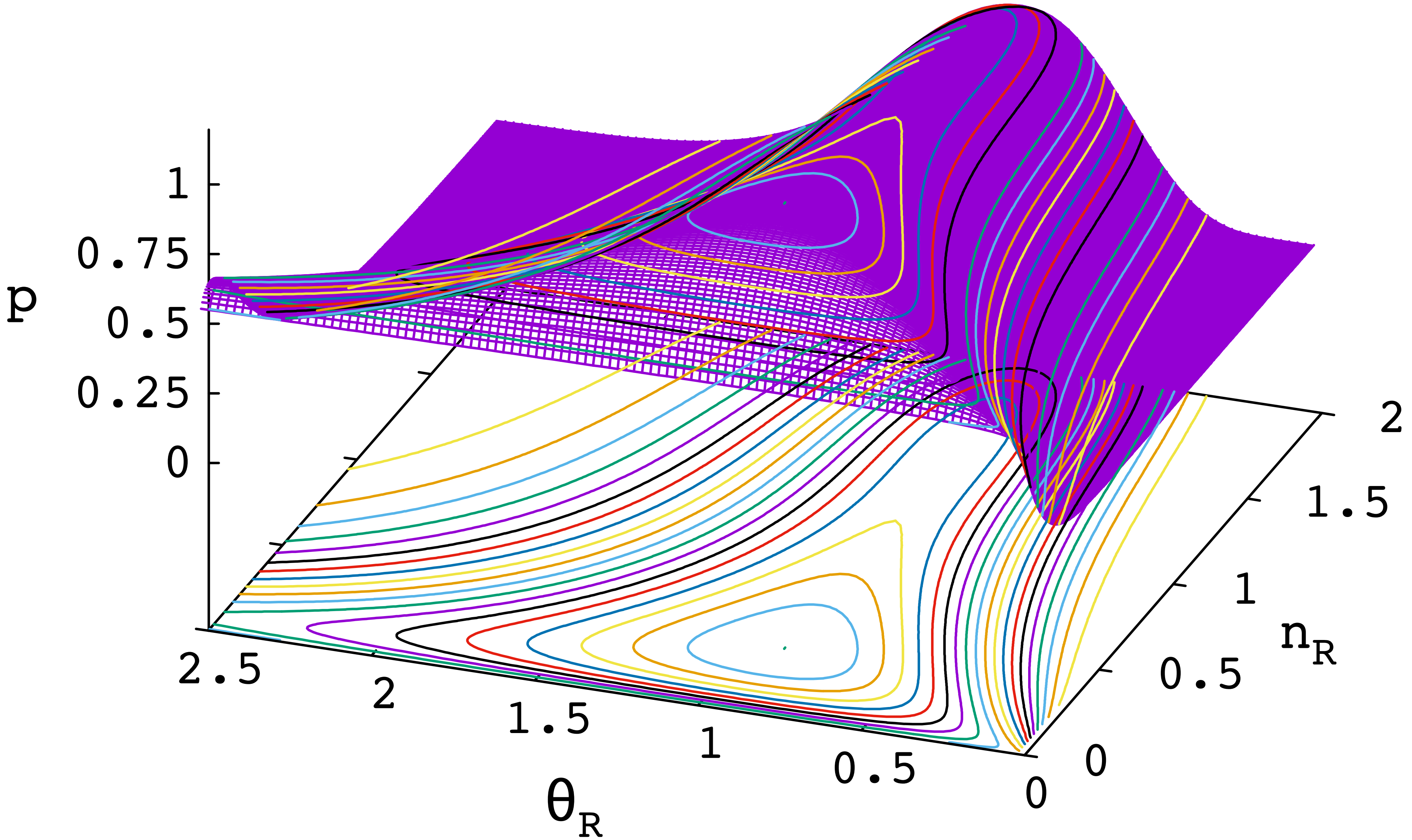
The pdf is a functional of the fluctuating field

From the VdW diffuse interface model:

$$S[\rho, E] = -\frac{\delta F[\rho, \theta]}{\delta \theta} = \int_{\mathcal{D}} \hat{s}_0(\rho, \theta) dv$$

$$p(\Delta) = \exp\left(\frac{1}{k_b}\int_{\mathcal{D}} \left(\hat{s}_0(\rho, \theta) - s_{eq}\right) dv\right)$$

Fluctuation probability



Correlations

$$\Delta S \simeq \Delta S_0 = -\frac{1}{2}\int_V dV \left[\frac{c_{T0}^2}{\theta_0\rho_0}\delta\rho^2 - \frac{\lambda}{\theta_0}\delta\rho\left(\nabla^2\delta\rho\right) + \frac{\rho_0}{\theta_0}\delta\mathbf{v}\cdot\delta\mathbf{v} + \frac{\rho_0 c_{v0}}{\theta_0^2}\delta\theta^2\right]$$

$$C_\Delta(\mathbf{x}) = \langle\Delta\otimes\Delta^\dagger\rangle = \frac{1}{Z}\int D\delta\rho D\delta\mathbf{v} D\delta\theta\,\Delta\otimes\Delta^\dagger\ \exp\left(\frac{1}{k_B}\int_V \Delta s(\delta\rho,\delta\mathbf{v},\delta\theta)\,dV\right)$$

* The correlation tensor is expressed as a path integral

$$C_{\delta\rho\delta\rho} = \frac{\mathrm{k}_B\theta_0}{4\pi\lambda\,|\hat{\mathbf{r}}-\tilde{\mathbf{r}}|}\exp\left(-|\hat{\mathbf{r}}-\tilde{\mathbf{r}}|\sqrt{\frac{c_T^2}{\rho_0\lambda}}\right)\quad C_{\delta\theta\delta\theta} = \frac{k_B\theta_0^2}{\rho_0 c_v}\delta\left(\hat{\mathbf{r}}-\tilde{\mathbf{r}}\right)\quad \mathbf{C}_{\delta\mathbf{v}\delta\mathbf{v}} = \frac{k_B\theta_0}{\rho_0}\mathbf{I}\delta\left(\hat{\mathbf{r}}-\tilde{\mathbf{r}}\right)$$

Landau-Lifschitz-Navier-Stokes equations

Stochastic terms included to force thermal fluctuations: SPDEs

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0$$

$$\frac{\partial \rho \mathbf{u}}{\partial t} + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) = -\nabla p + \nabla \cdot \Sigma + \boxed{\nabla \cdot \delta\Sigma}$$

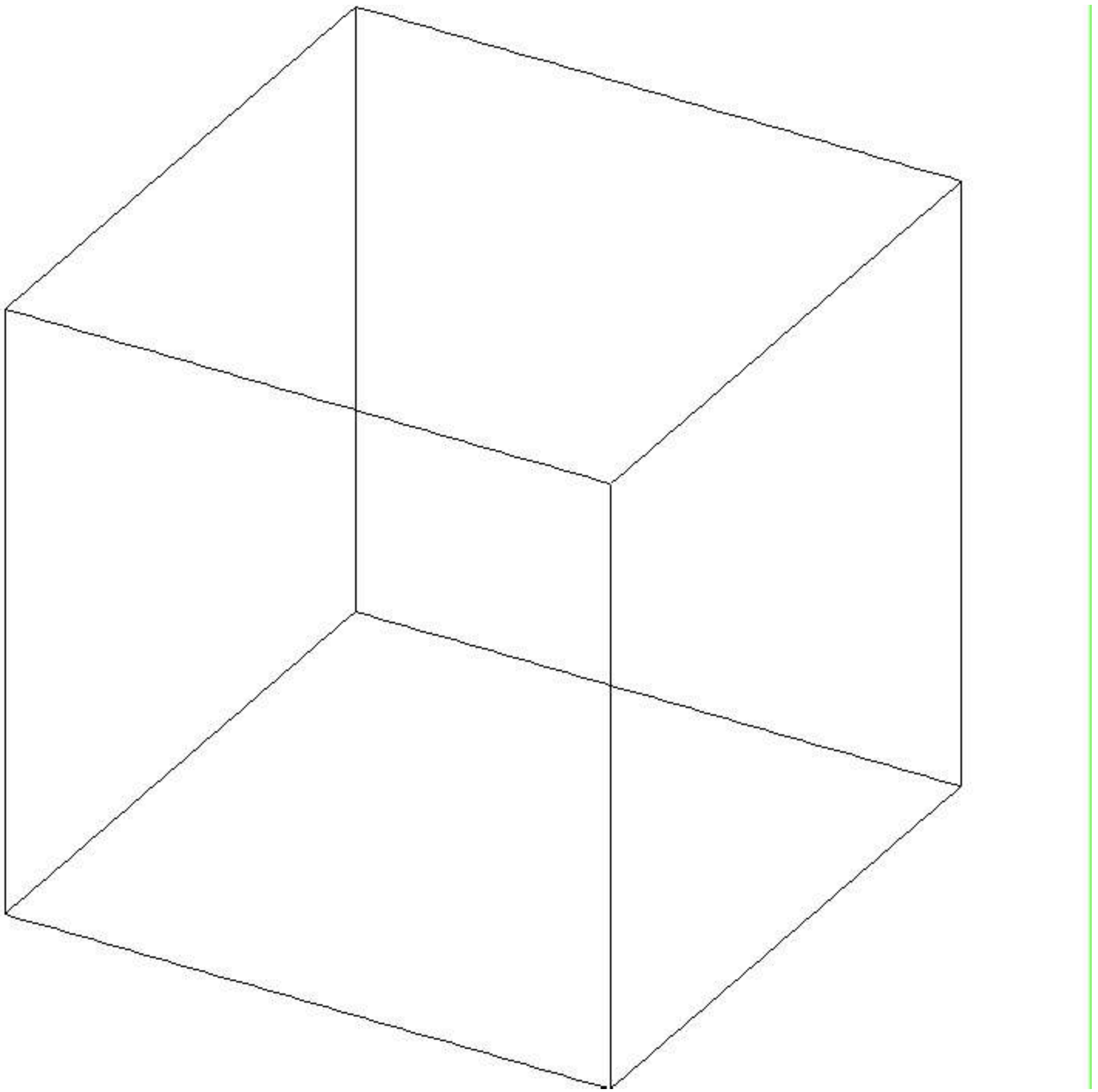
$$\frac{\partial E}{\partial t} + \nabla \cdot (\mathbf{u} E) = \nabla \cdot (-p\mathbf{u} + \mathbf{u} \cdot \Sigma - \mathbf{q}) + \boxed{\nabla \cdot (\mathbf{u} \cdot \delta\Sigma + \delta\mathbf{q})}$$

Solution should reproduce the equilibrium pdf (fluctuation-dissipation theorem) :
enforcing equilibrium **correlations determines noise** amplitude

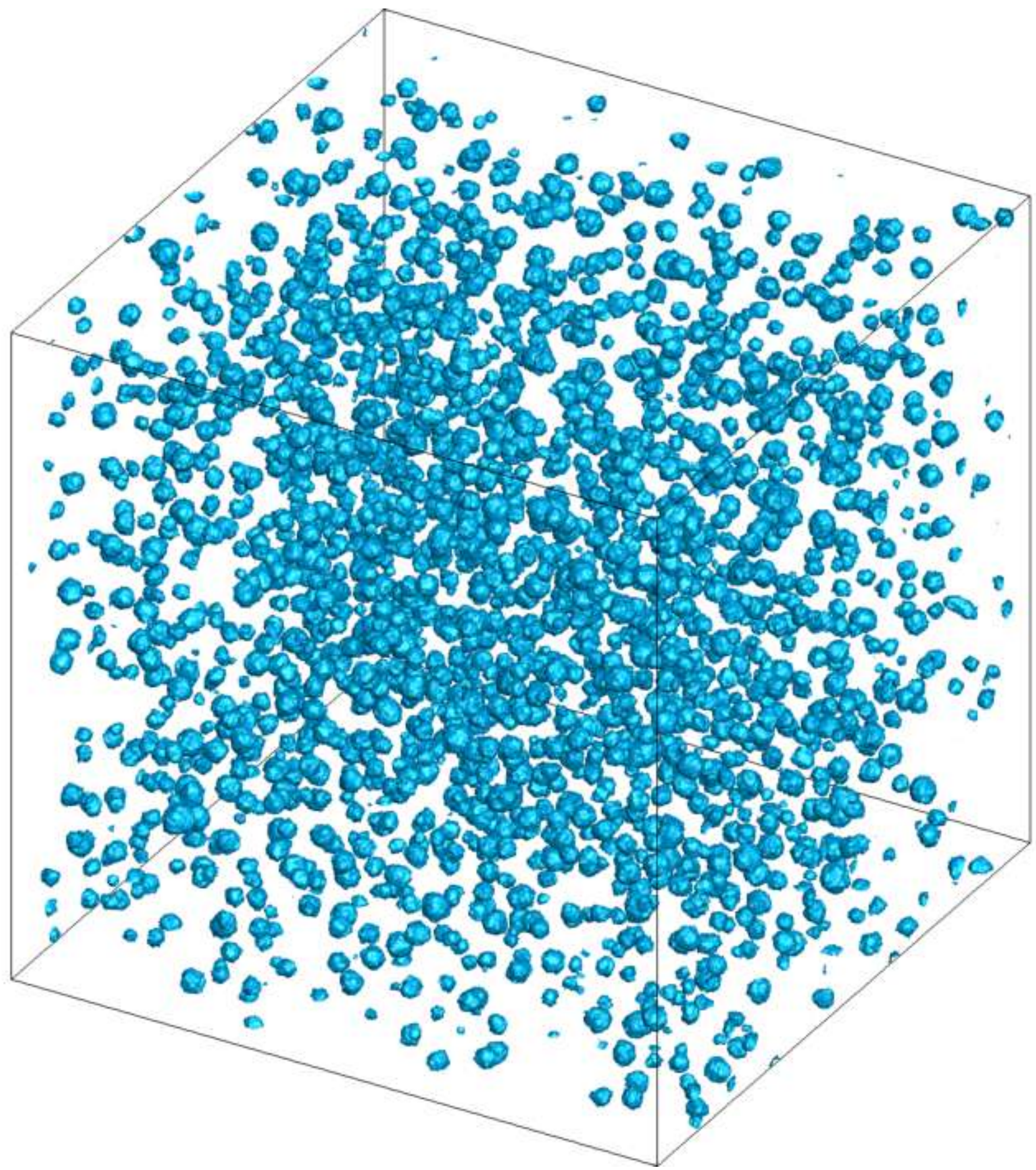
$$\boldsymbol{\delta\Sigma} = \sqrt{2\mu \mathrm{k}_B \theta}\,\boldsymbol{\tilde{W}^v} - \frac{1}{3}\sqrt{2\mu \mathrm{k}_B \theta}\,\mathrm{Tr}\left(\boldsymbol{\tilde{W}^v}\right)\boldsymbol{I}$$

$$\boldsymbol{\delta q} = \sqrt{2k \mathrm{k}_B \theta^2}\,\boldsymbol{\tilde{W}^E}$$

Bubble formation

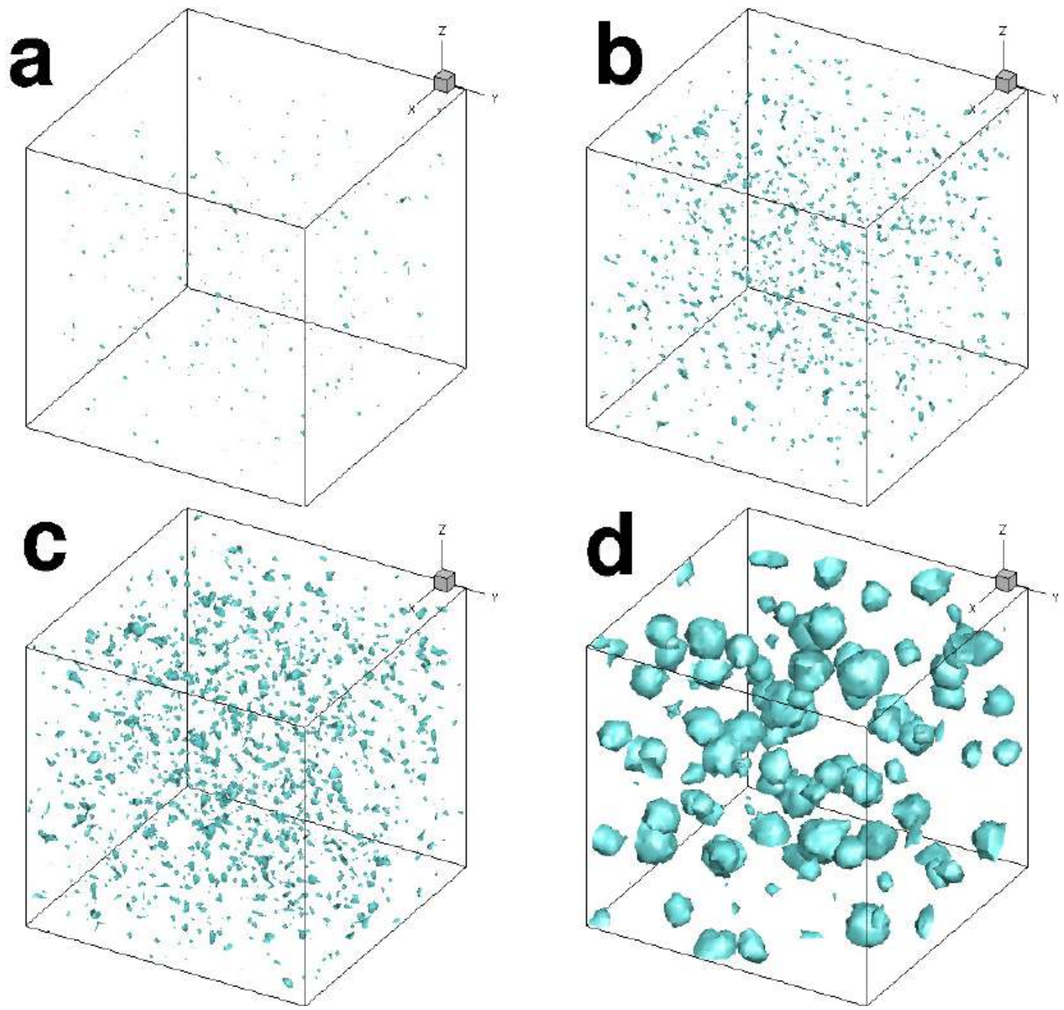


Bubbles

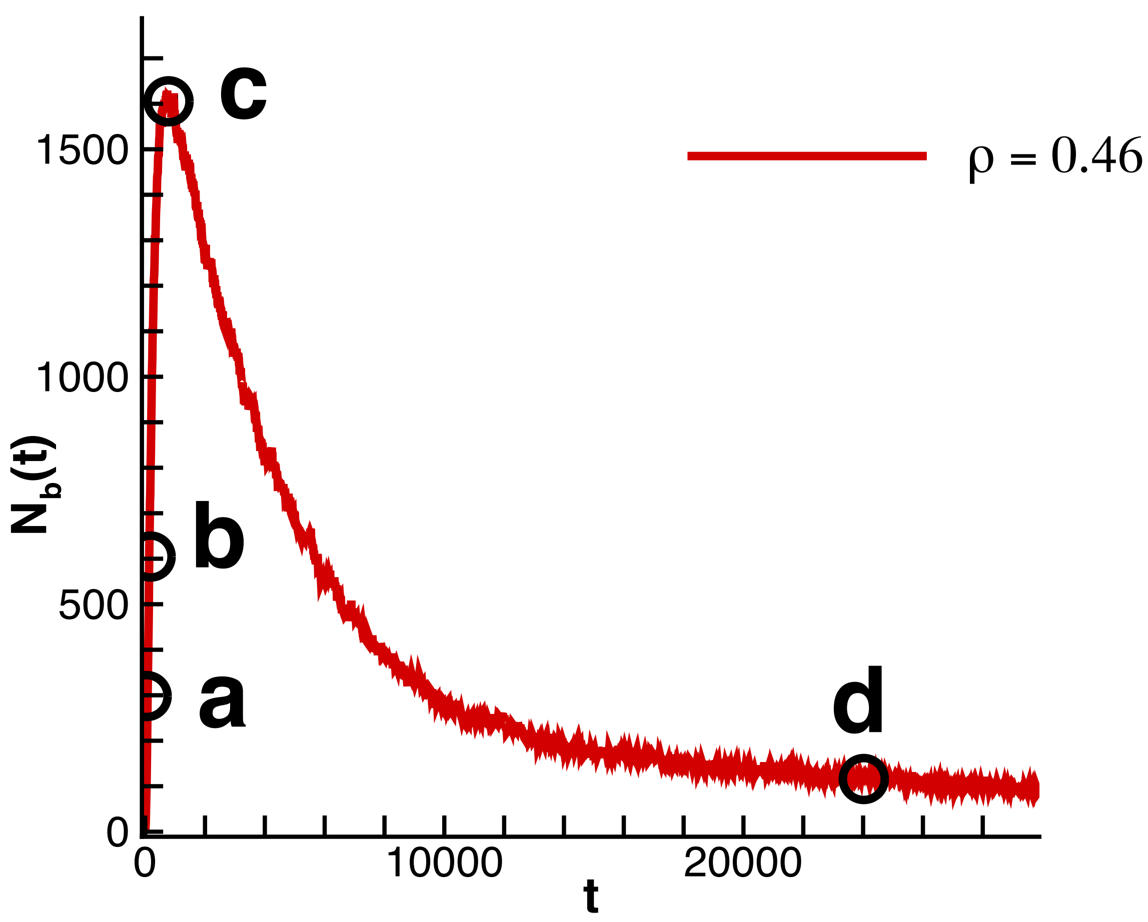


Bubble Nucleation in a Lennard-Jones fluid

Snapshots during the nucleation process



Number of stable bubbles vs time

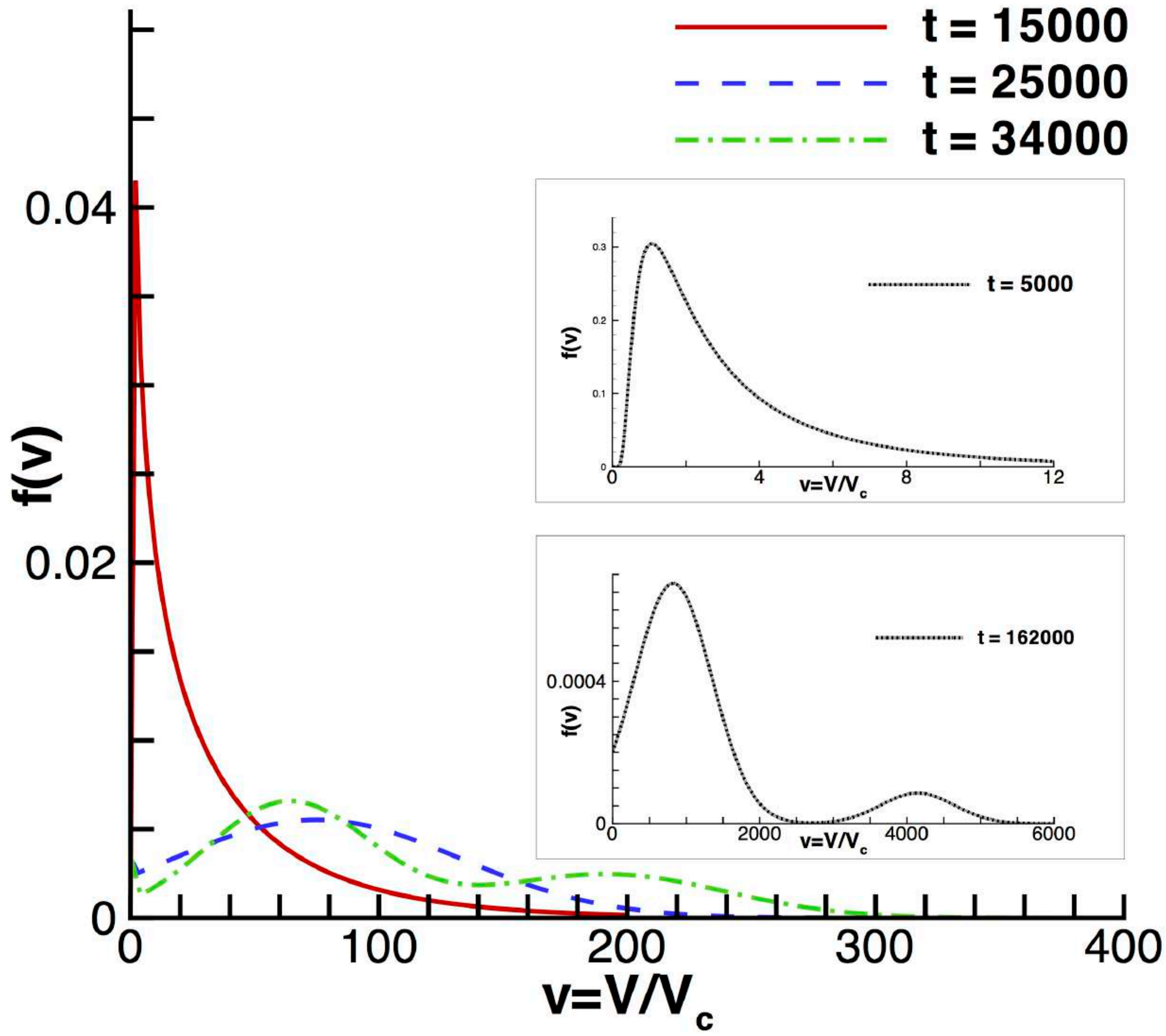


PHYSICAL REVIEW FLUIDS **00**, 003600 (2018)

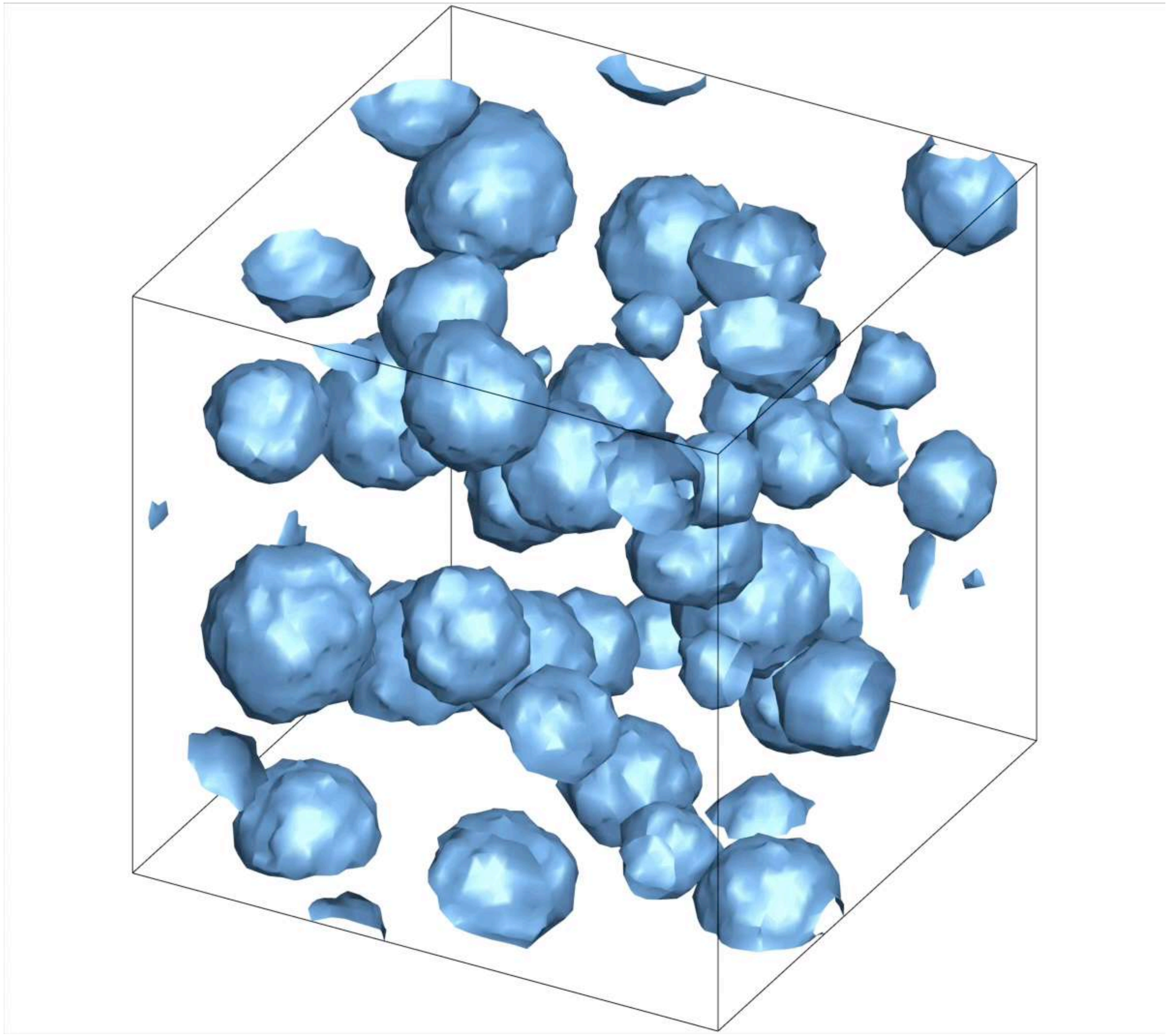
Thermally activated vapor bubble nucleation: The Landau-Lifshitz–Van der Waals approach

Mirko Gallo, Francesco Magaletti, and Carlo Massimo Casciola*
Department of Mechanical and Aerospace Engineering, Sapienza Università di Roma, Rome, Italy

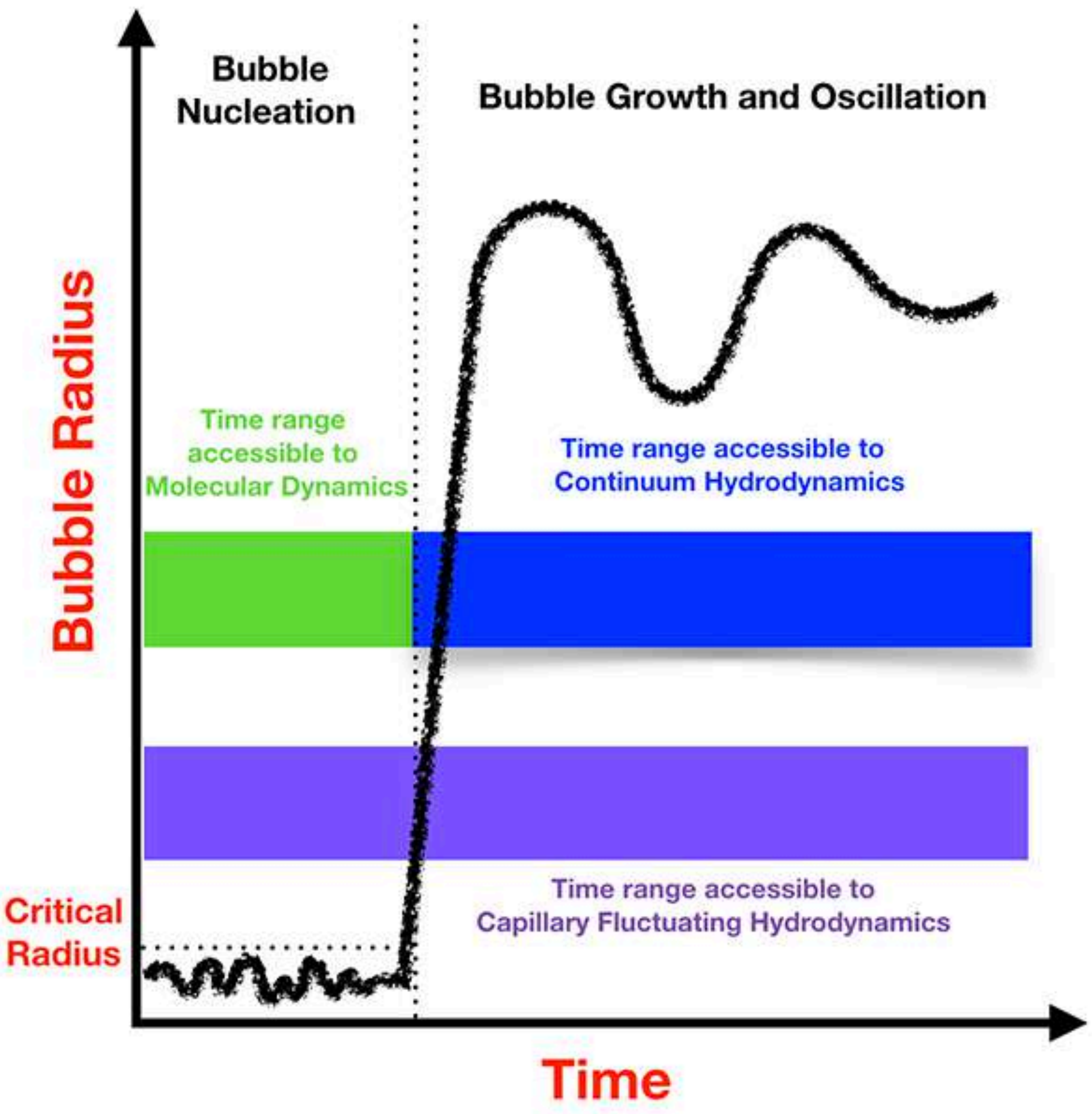
Bubble Size Distribution

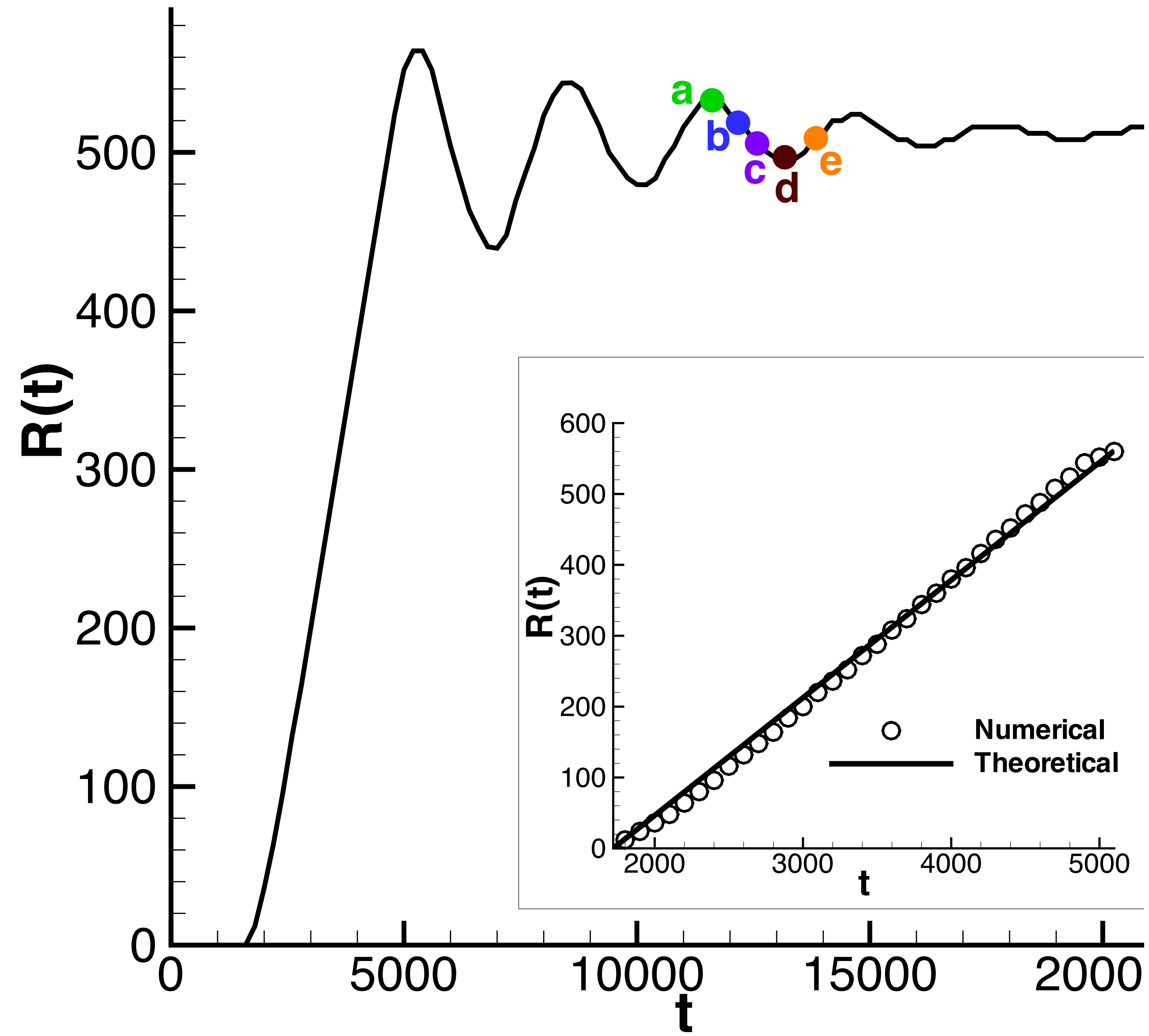


Bubble Cohalescence



From Nucleation to Nonlinear Bubble Dynamics





Nucleation and growth dynamics of vapor bubbles

Mirko Gallo[1], Francesco Magaletti[1,2], Davide Cocco[3] and Carlo Massimo Casciola[1]†

JFM 2019

WALL WETTABILITY

Heterogeneous Nucleation

$$F[\rho,\theta] = \int_{\mathcal{D}} \left(\hat{f}_0(\rho,\theta) + \frac{\lambda}{2}|\nabla\rho|^2 \right) dv + \int_{\partial D} \hat{f}_w(\rho,\theta) dS$$

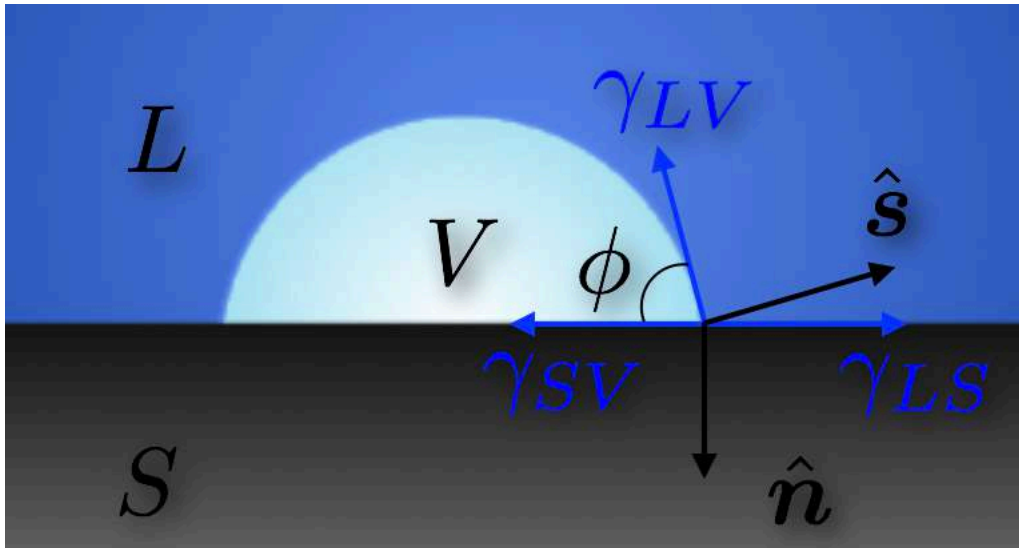
Free-energy functional assumed to be given by three contributions

- bulk free-energy density of the homogeneous phases
- energy penalty associated with the interface
- surface energy (fluid-wall interaction)

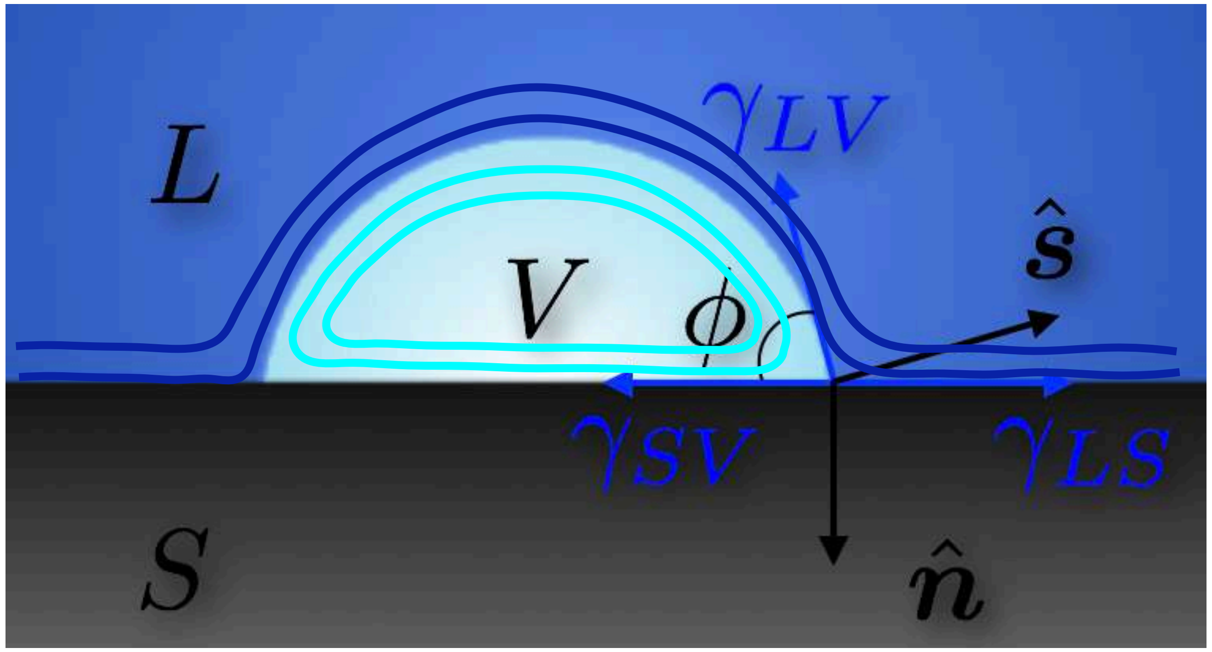
Equilibrium conditions

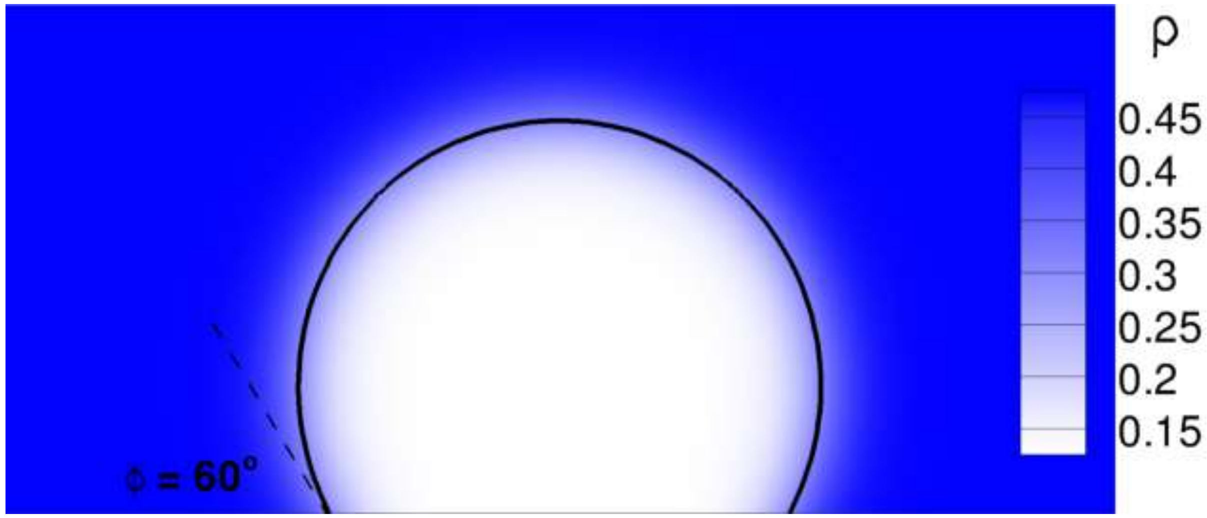
$$\frac{\partial \hat{f}_0}{\partial \rho} - \nabla \cdot (\lambda \nabla \rho) = 0$$ in the bulk fluid

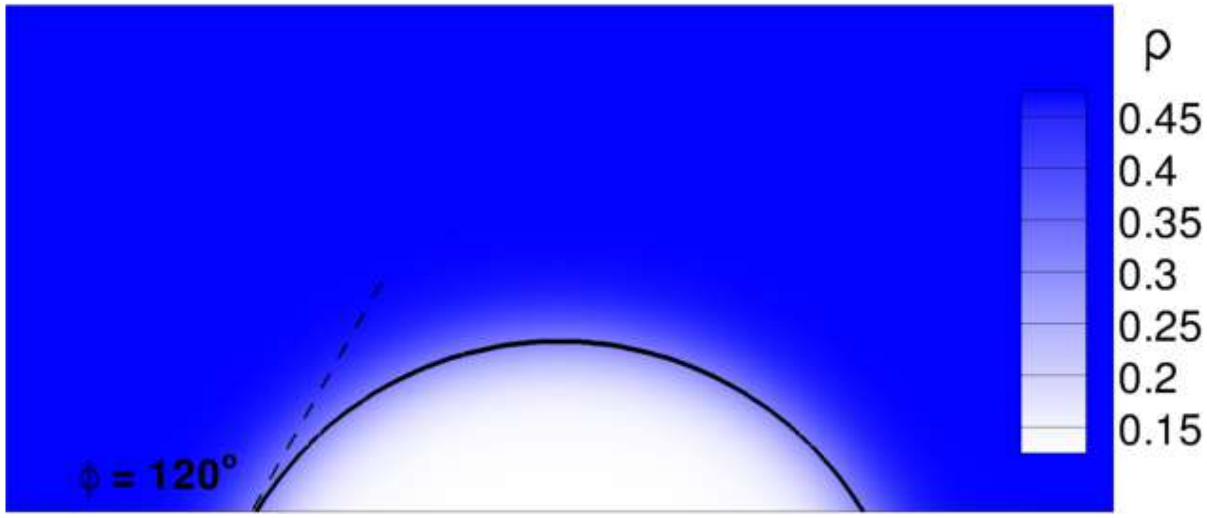
$$\lambda \frac{\partial \rho}{\partial n} + \frac{\partial \hat{f}_w}{\partial \rho} = 0$$ at the wall



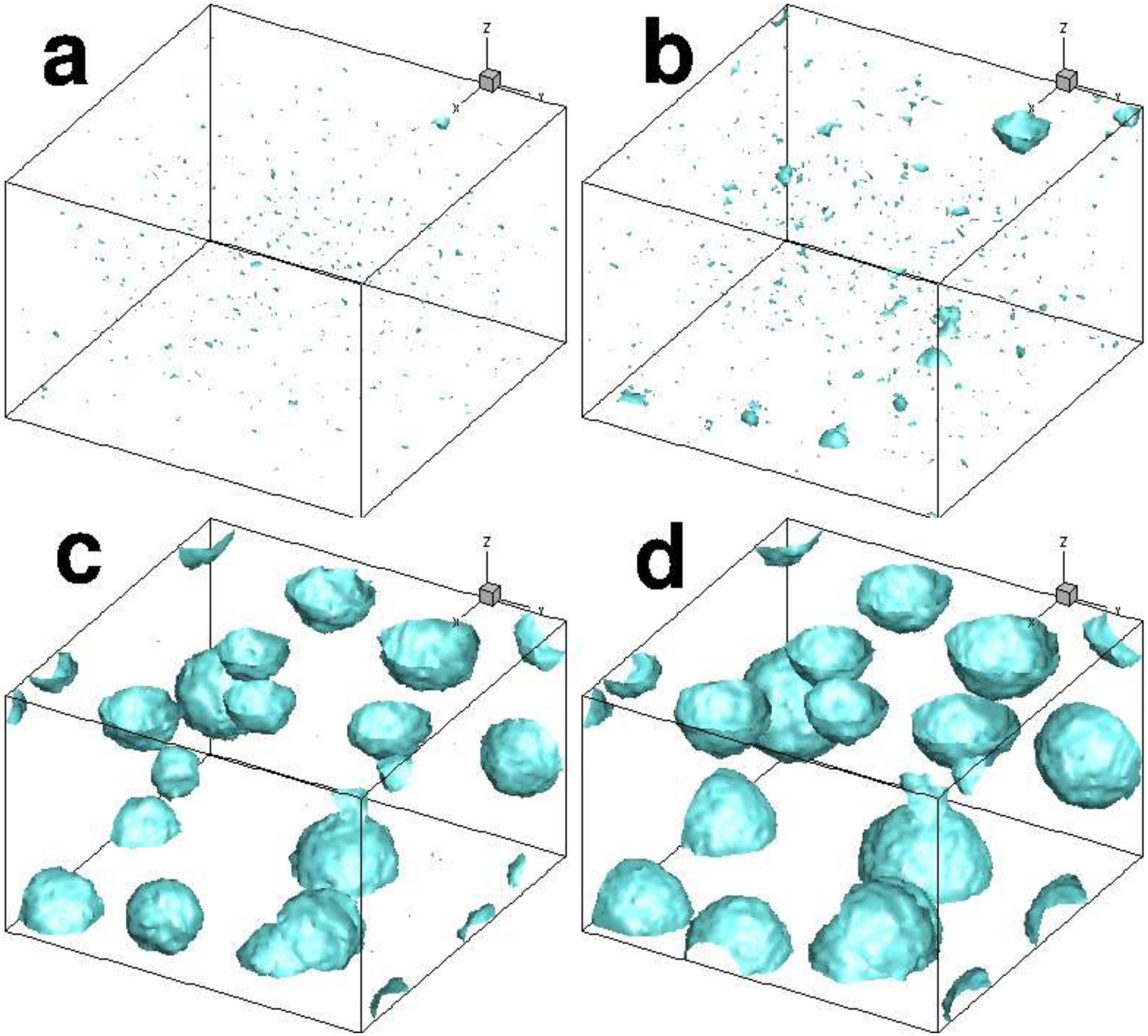
Sketch of Density at the Triple Line





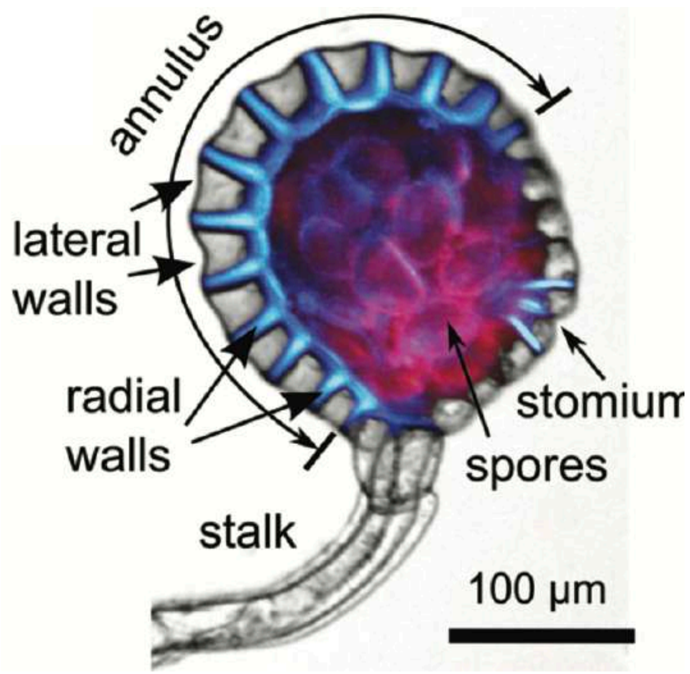


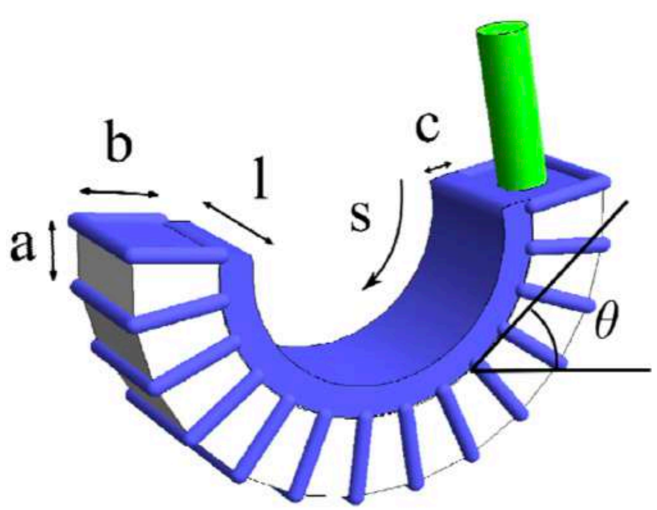
Bubble Nucleation at a Solid Surface



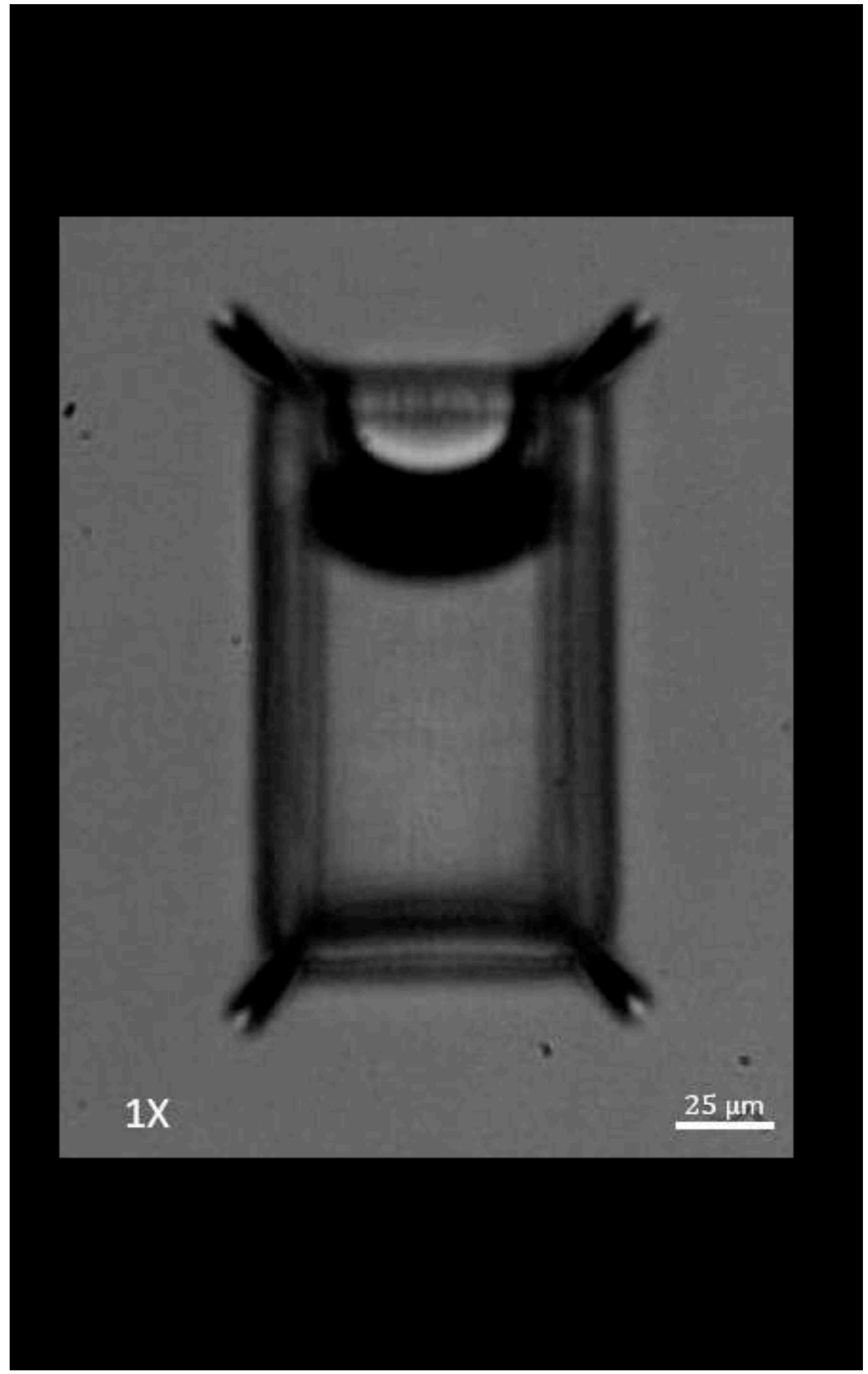
Gallo, Magaletti, Casciola, J. Fluid Mech. 2021

Confined Nucleation

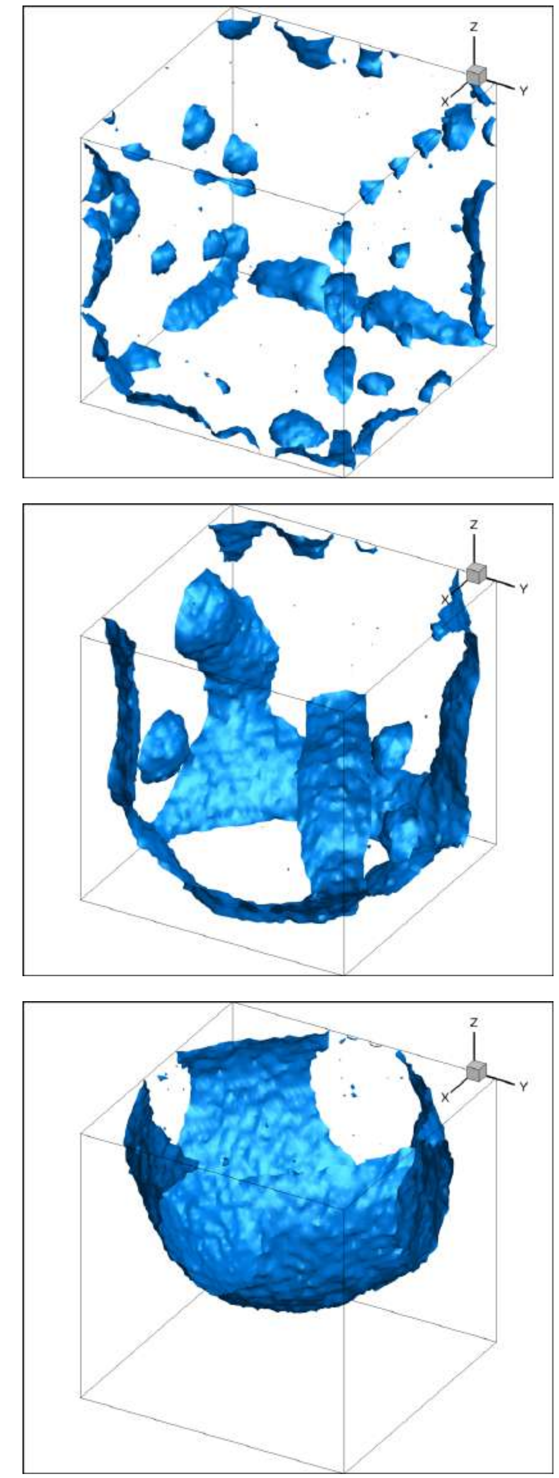




Noblin at al. Science 2021



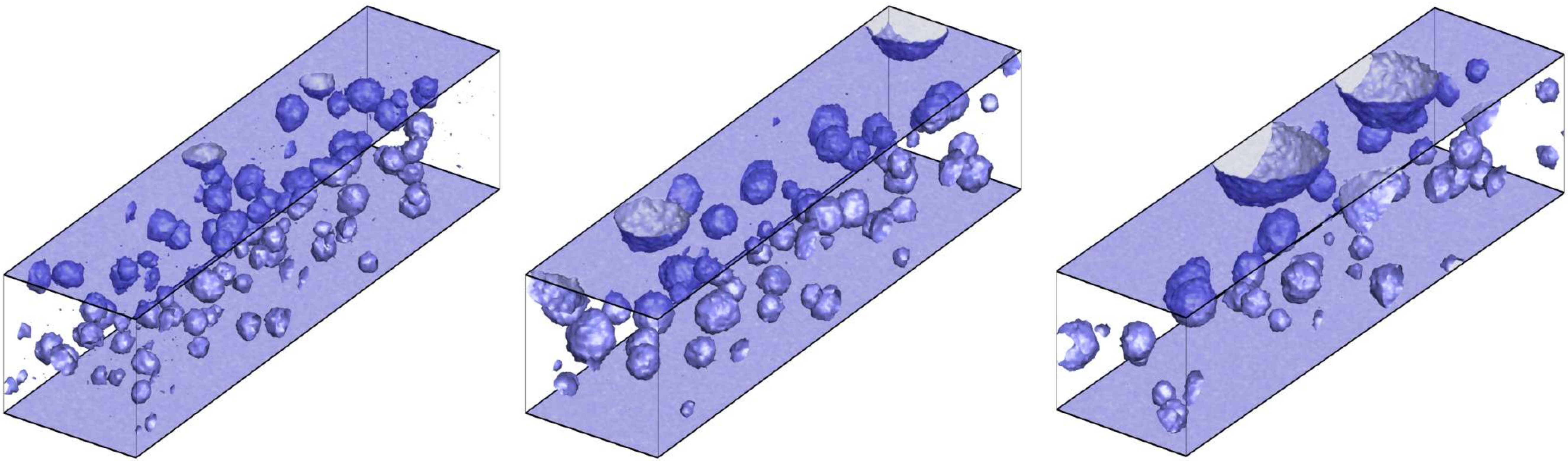
Courtesy of Barbara Mazzolai &
Andrea Montagna (IIT)

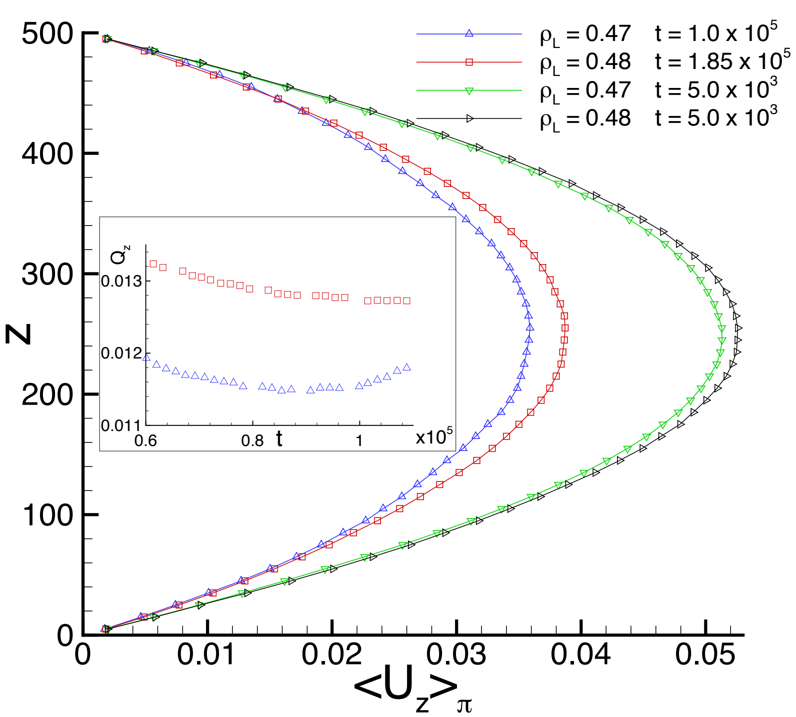


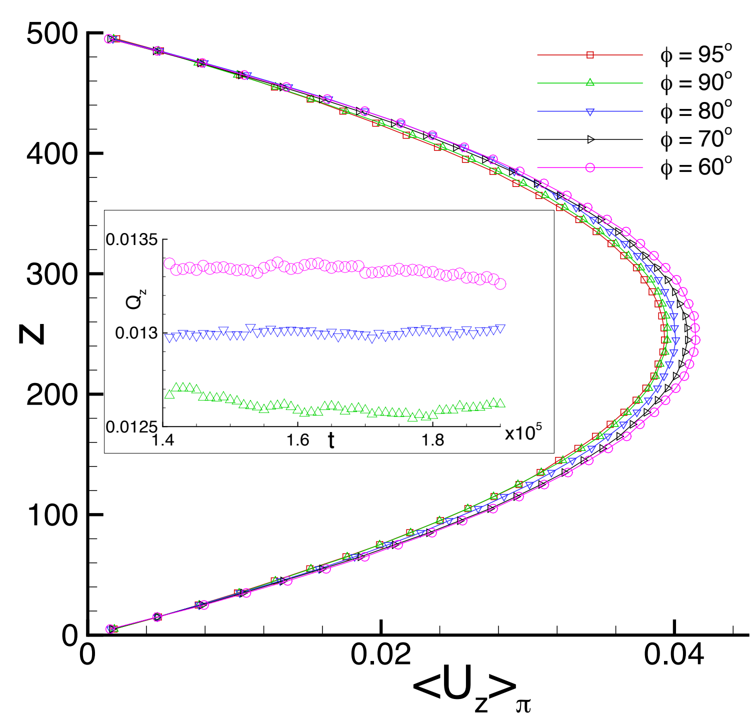
FHD-Simulation

NUCLEATION UNDER FLOW

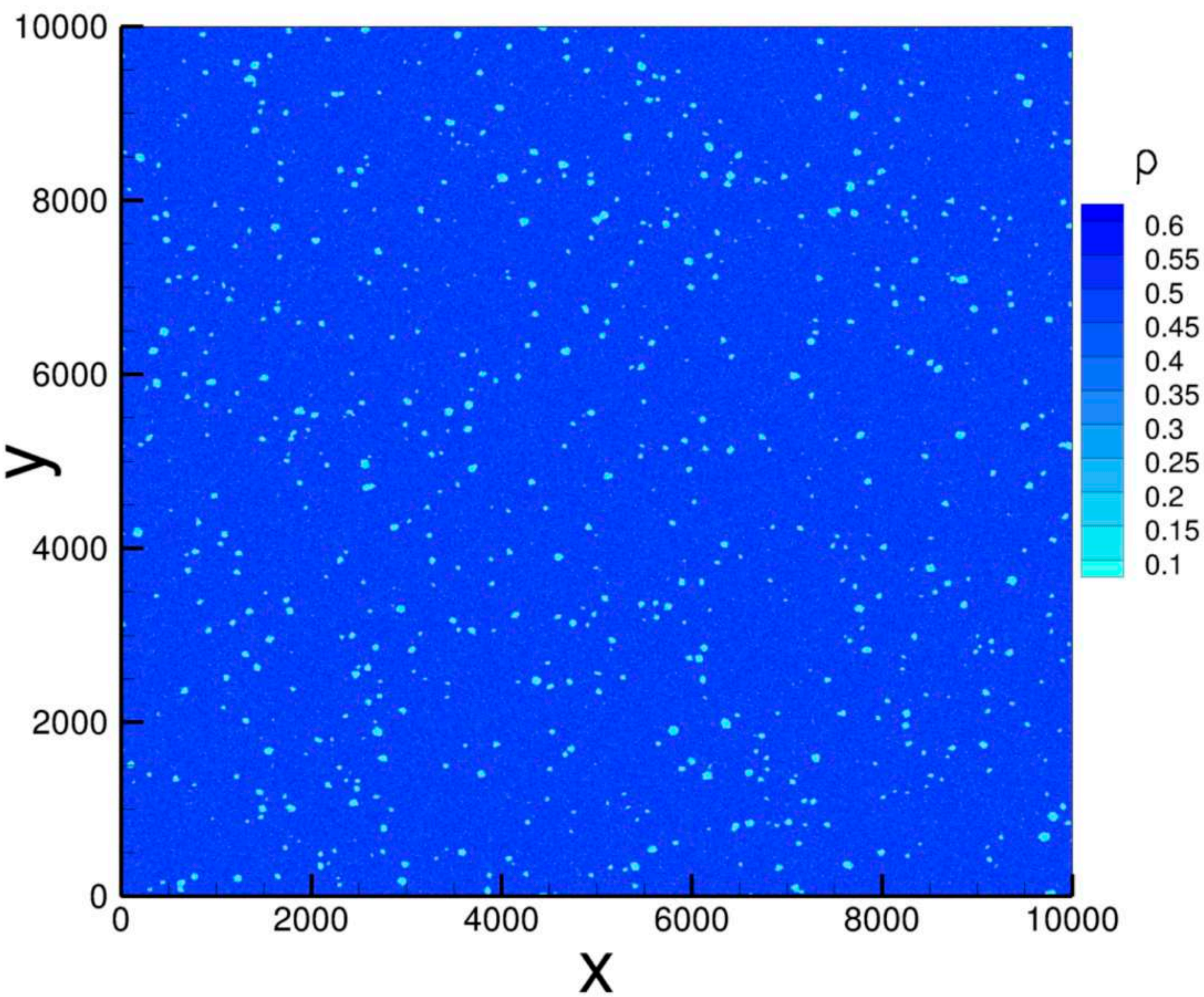
Bubble Nucleation Under Flow

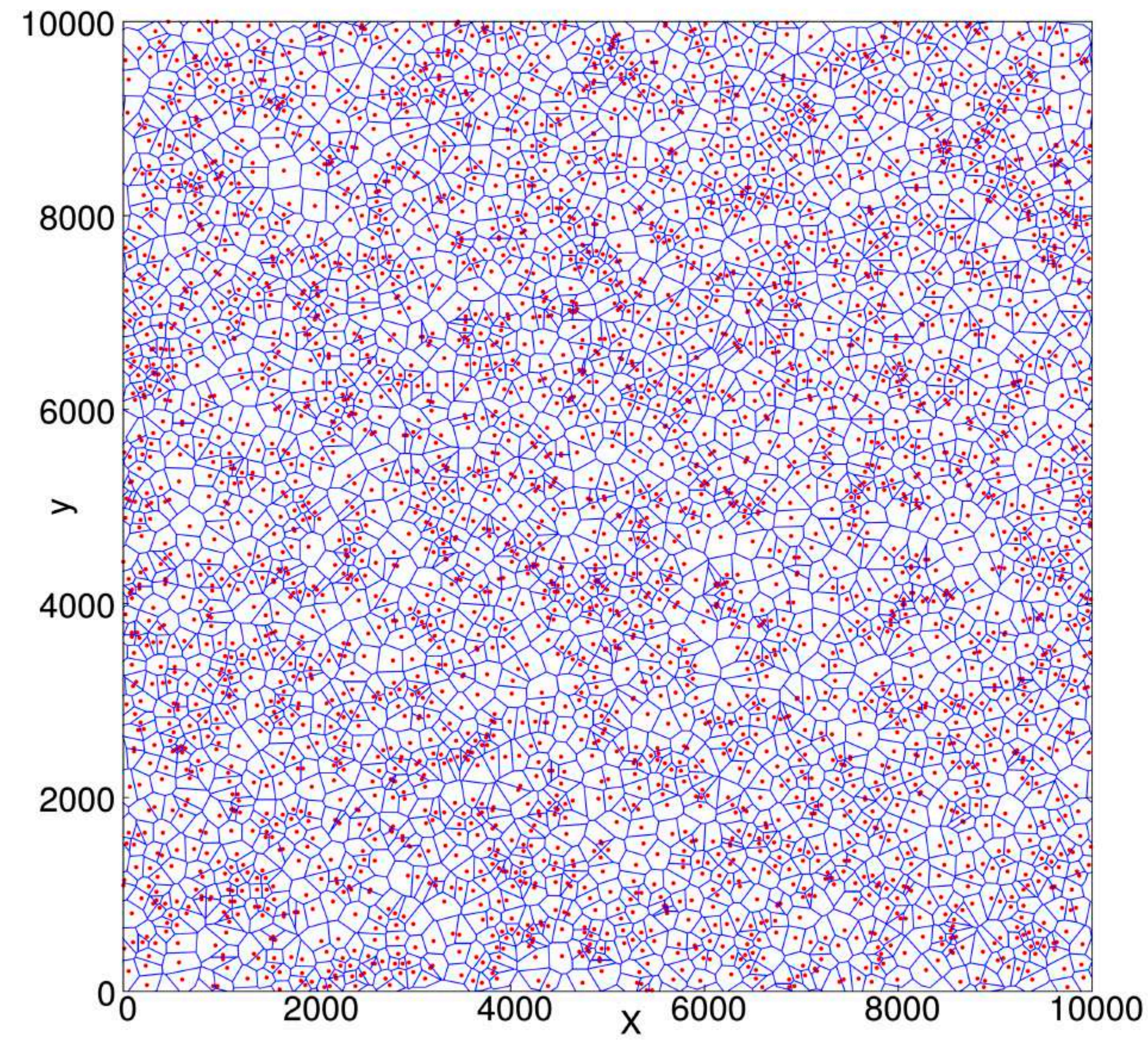




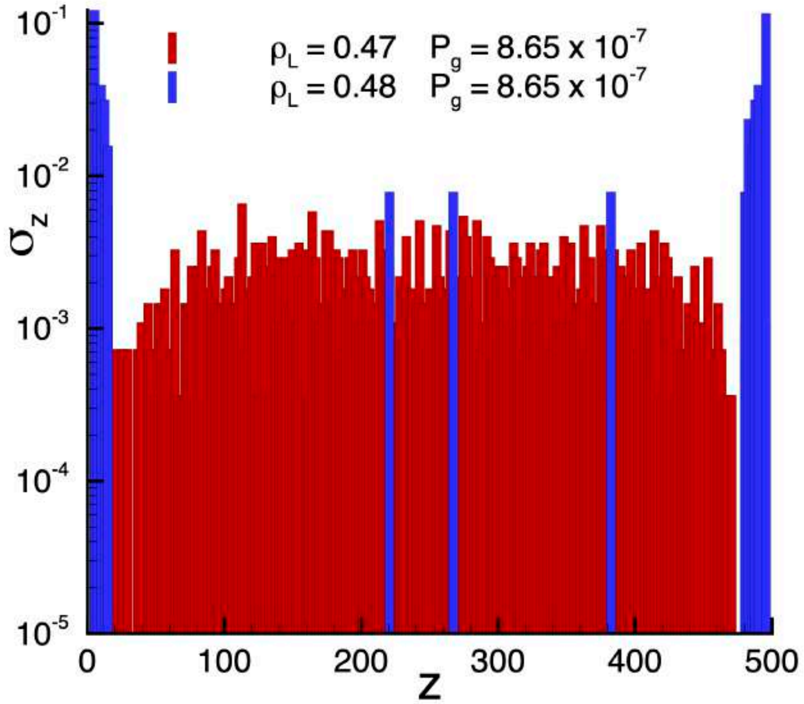


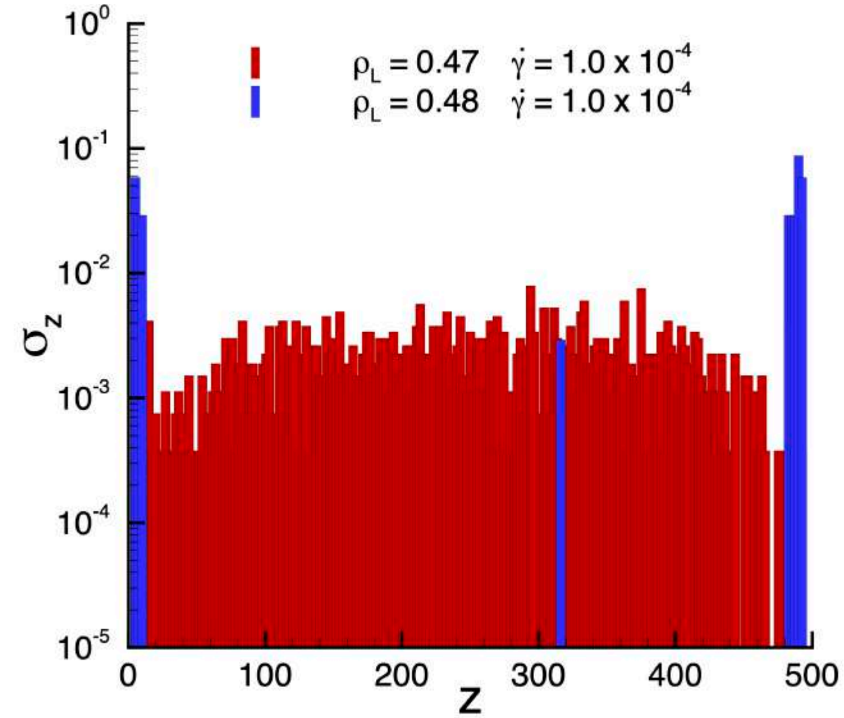
Bubble Nucleation under Flow

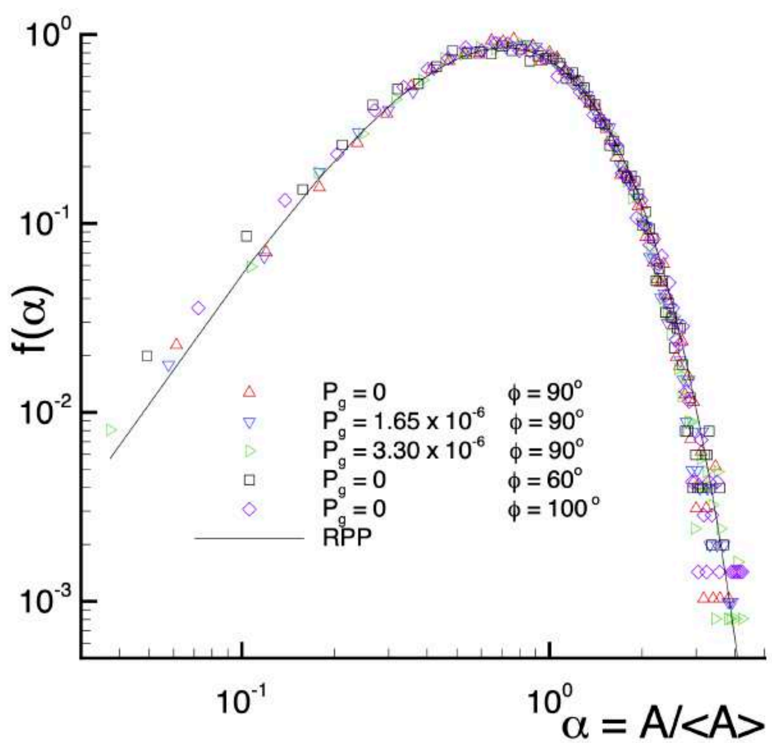


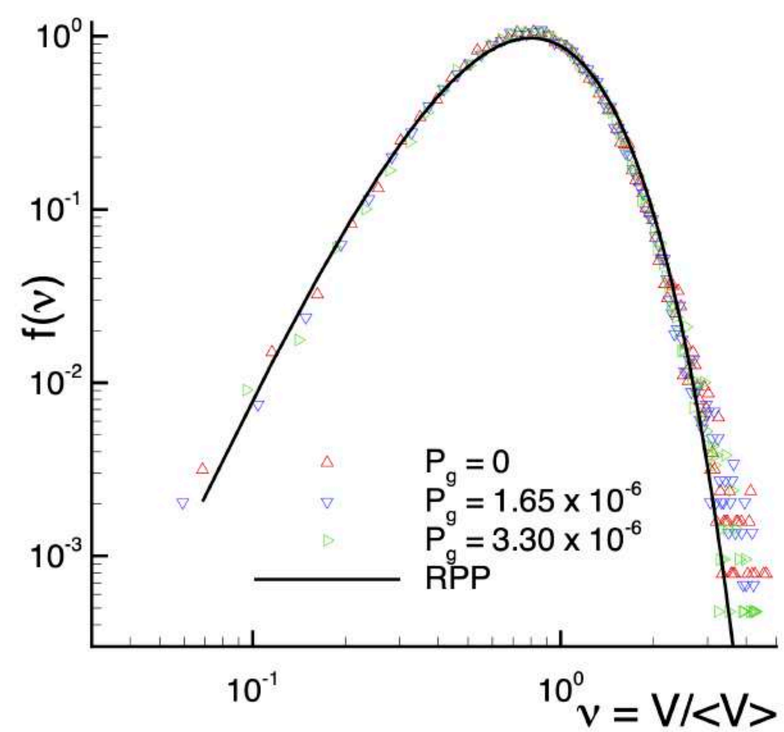


Heterogeneous Nucleation under Flow

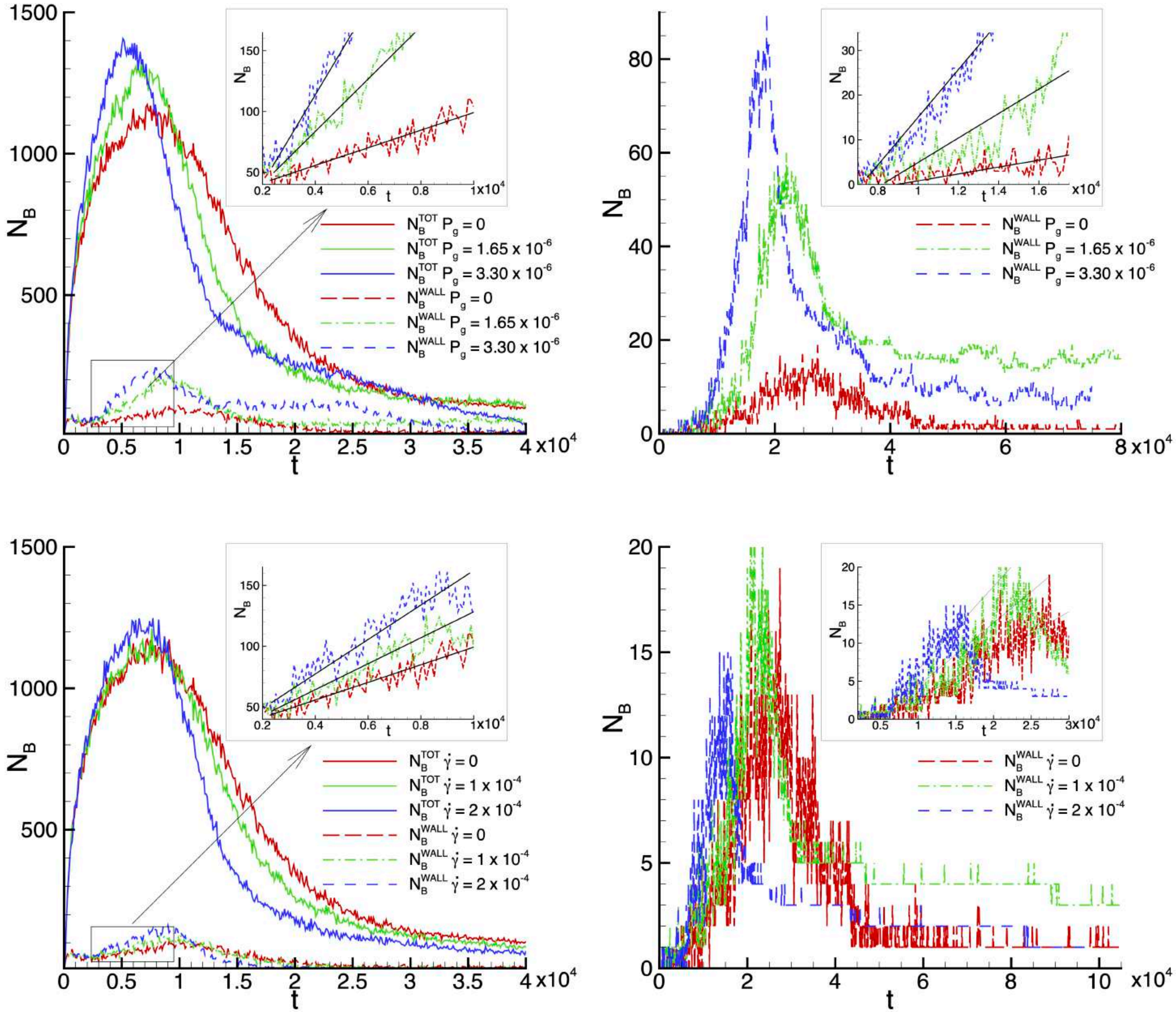








Heterogeneous Nucleation under Flow



Summary

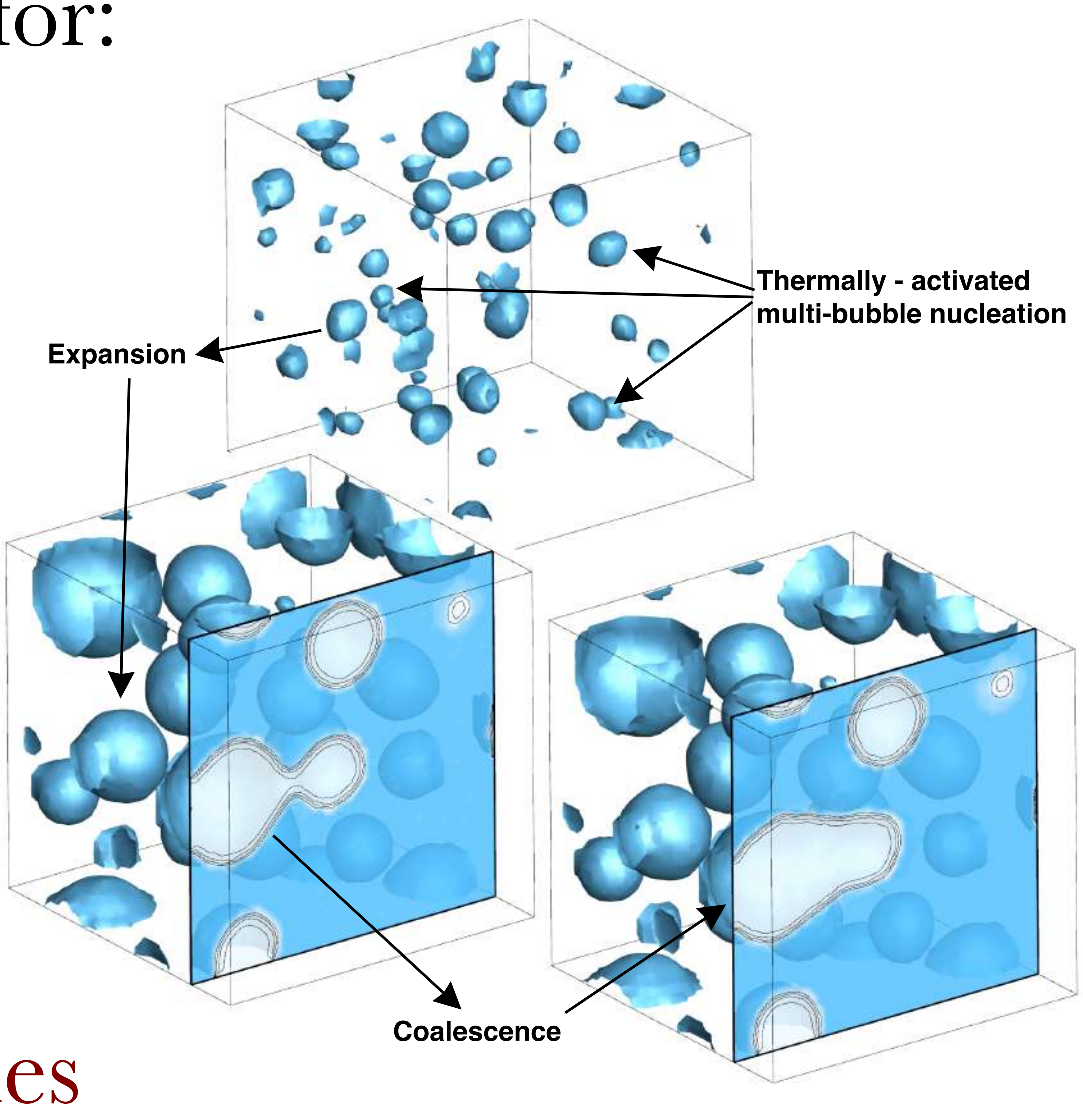
Stochastic pdes used to model vapour bubble nucleation
(Landau's FH + Van der Waals's capillarity + Navier-Stokes)

The deterministic component accounts for:

- phase change
- shock waves
- transition to/from supercritical state

The stochastic part features:

- bubble nucleation
- correct nucleation rate at changing thermodynamics conditions



- unprecedented time and length scales
- modelling of nucleation in dynamic conditions (flow induced cavitation)!

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Thank you for listening!