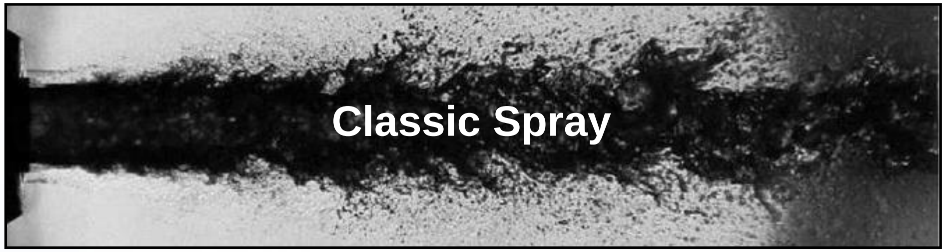
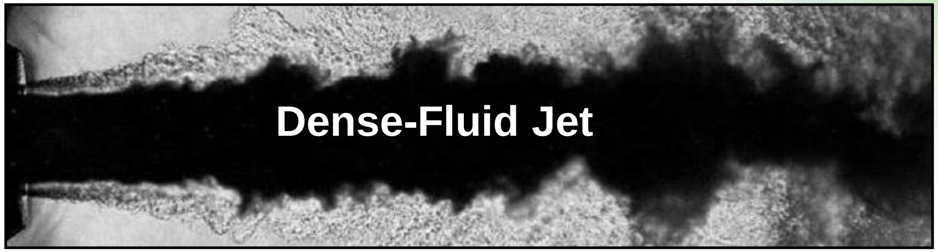
SAND2015-9247PE

The breakdown of classic spray atomization at engine-relevant conditions


Classic Spray


Dense-Fluid Jet

Rainer N. Dahms, Joseph C. Oefelein, Ahren Jasper, Julien Manin, and Lyle M. Pickett
Combustion Research Facility, Sandia National Laboratories, Livermore, CA

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences

Acknowledgement: Eric W. Lemmon, NIST, Boulder, CO
Cyril Crua, University of Brighton, Brighton, UK

Outline

Flow scales

Nano scales

☐ **Introduction & problem statement**

☐ **Real-fluid thermodynamics & mean-field capillary theory**

☐ **Breakdown of classic two-phase relations across fluid interfaces**
A new fundamental mechanism: Bridging sub- & supercritical limits

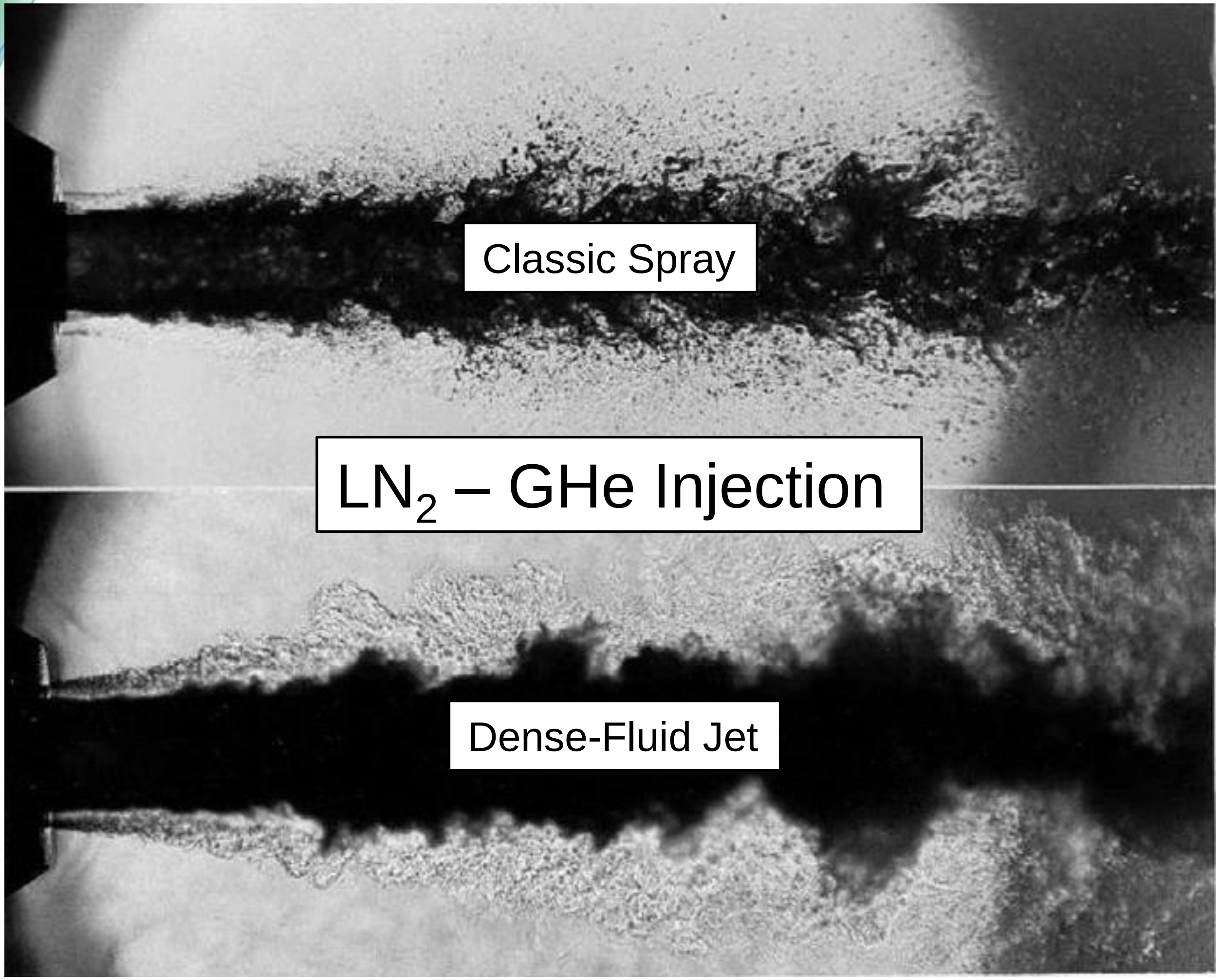
☐ **Non-equilibrium nano-scale dynamics within fluid interfaces**
Fundamentally understanding high-pressure gas-liquid mixing

☐ **A novel regime diagram for gas-liquid mixing dynamics**
Experimental validation (Pickett) & large-eddy simulation (Oefelein)

☐ **Cryogenic & hydrocarbon liquids: Fundamental differences**

☐ **Summary & Conclusions**

Classic sprays vs. dense-fluid jets from liquid rocket imaging



Low chamber pressure (10 bar)

High chamber pressure (60 bar)

W. Mayer, A. Schik, B.Vieille, C. Chaveau, I.Gökalp, D. Talley, and R.Woodward,
“Atomization and breakup of cryogenic propellants under high-pressure subcritical and supercritical conditions,”
J. Propul. Power **14** (1998) 835-842

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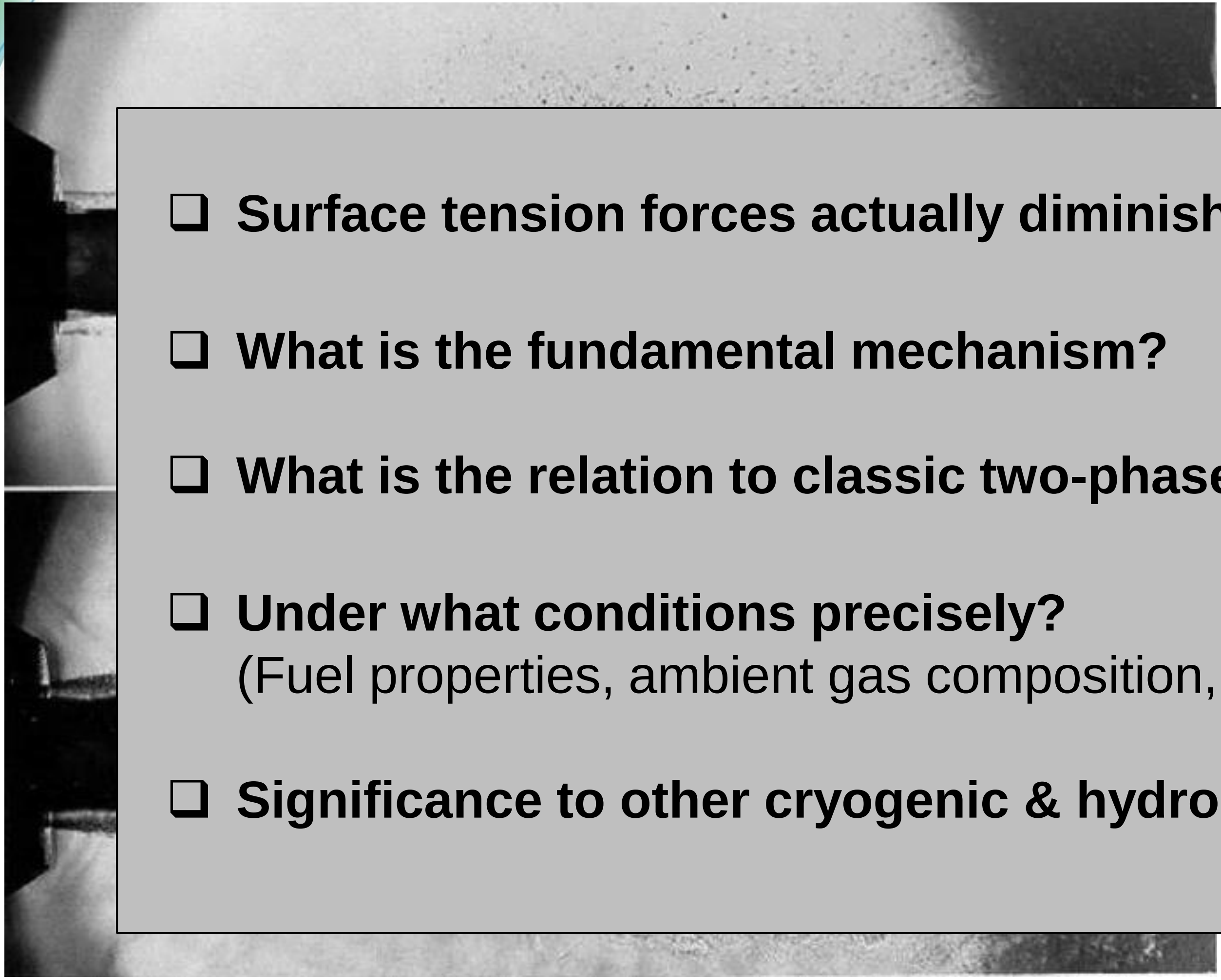

- ☐ **Surface tension forces actually diminished?**
- ☐ **What is the fundamental mechanism?**
- ☐ **What is the relation to classic two-phase theory?**
- ☐ **Under what conditions precisely?**
  (Fuel properties, ambient gas composition, etc.)
- ☐ **Significance to other cryogenic & hydrocarbon systems?**

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Real-fluid multi-component thermodynamics and transport

Eric Lemmon, NIST, Boulder

- **Helmholtz energy (A) EoS for each species!**

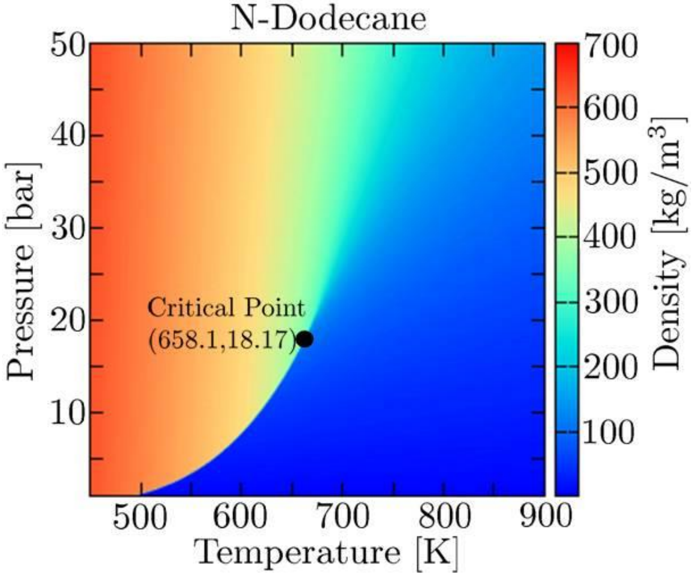
$$\frac{A}{RT} = \alpha^0(\delta,\tau) + \alpha^r(\delta,\tau)$$

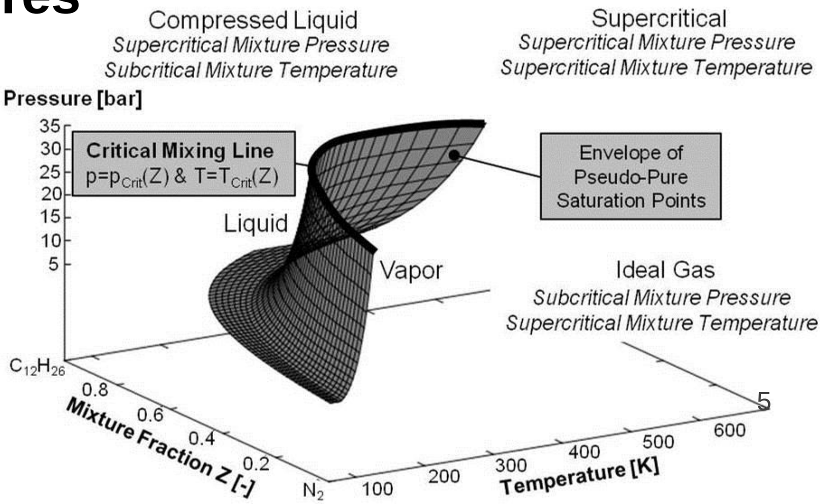
All thermodynamic properties derivable from A

- **Non-linear combination rules for mixtures**
  Reducing functions vary with composition

- **Applies to (arbitrary) liquid & gas mixtures at all relevant pressures and temperatures**
  (incl. near-critical and supercritical)

- **Exhibits desirable behavior over entire two-phase regime of fluid densities**
  (including meta-stable, unstable regions)





Mean-field capillary theory for molecular gas-liquid interfaces

- ☐ **Theory foundation:**
  - Established by van der Waals in 1894
  - Reformulated by Cahn & Hilliard in 1958

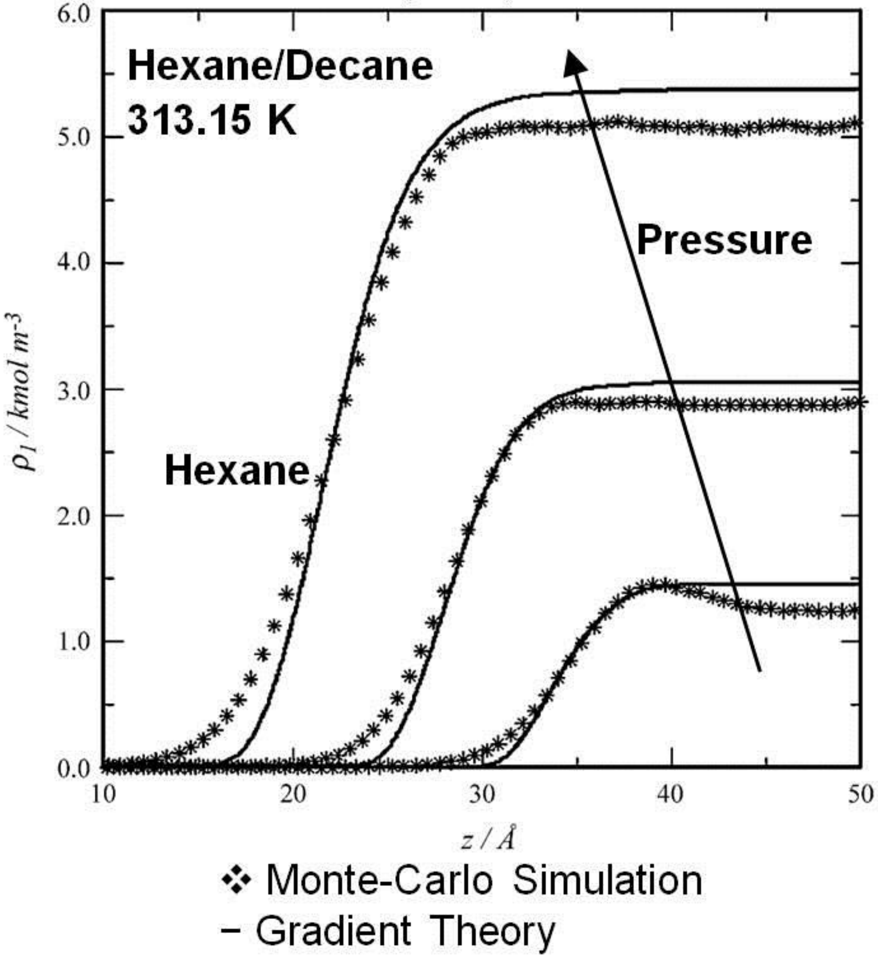
$$\boxed{\sum\sum\frac{1}{2}\kappa_{ij}\frac{d\varrho_i}{dz}\frac{\varrho_j}{dz}=\bar{\omega}(\varrho)-\bar{\omega}_s}$$

- ☐ **Thermo-mechanical model of continuous fluid media**
  - Surface tension & interfacial profiles as function of multi-component mixture

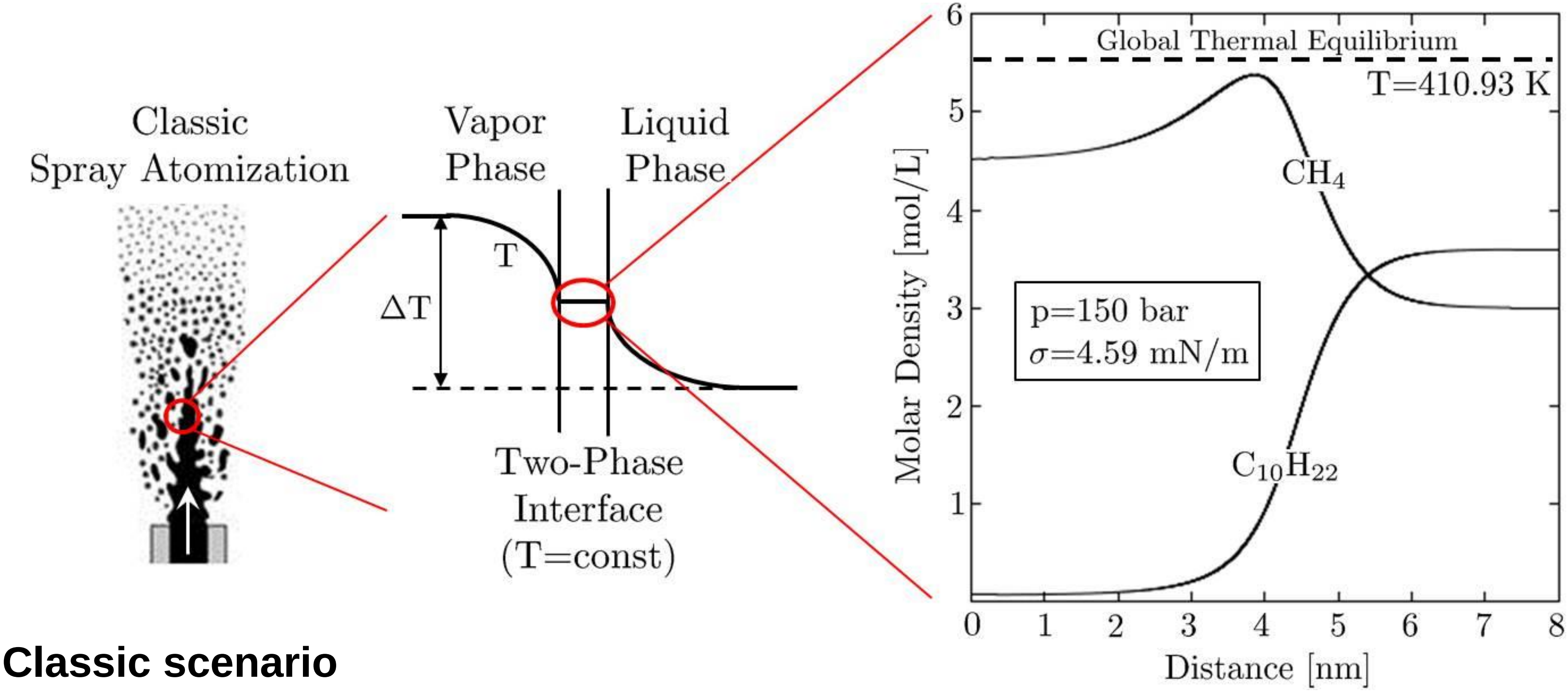
- ☐ **Applied to**
  - Hydrocarbon mixtures
  - Polar compounds
  - Vapor-liquid, liquid-liquid interfaces

- ☐ **Mean-field solution consistent with averaged Monte-Carlo simulations**

Müller et al., Fluid Phase Equilibria 282 (2009) 68-81

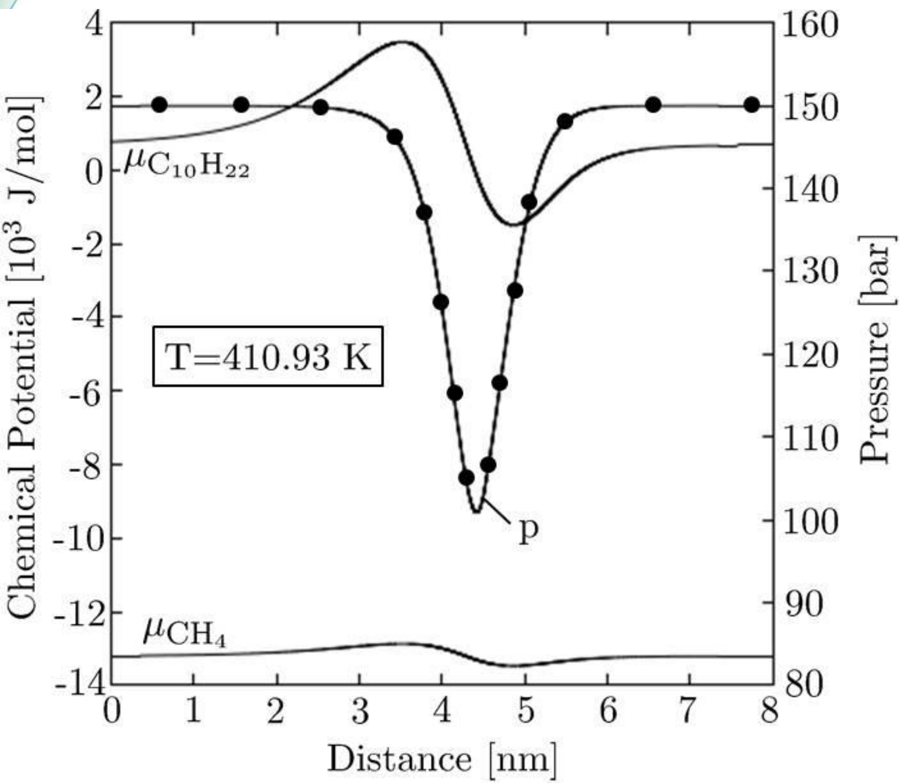


Classic two-phase theory & spray atomization



- **Classic scenario**
  - Injected liquid has significantly lower temperature than ambient gas
- **Isothermal interface regardless of temperature difference**
- **Interface: State of global thermal equil. → Helmholtz free energy is minimal !**
  - Mean-field equation $\left(\sum\sum\frac{1}{2}\kappa_{ij}\frac{d\varrho_i}{dz}\frac{\varrho_j}{dz}=\bar{\omega}(\varrho)-\bar{\omega}_s\right)$ only valid for minimal free energy!
  - Minimal energy state governs interfacial species distributions

Classic two-phase theory & spray atomization



- ☐ **Chem. pot./pressure determined by density, composition, gradient energy**
  - Distributions also governed by interfacial minimal Helmholtz free energy state

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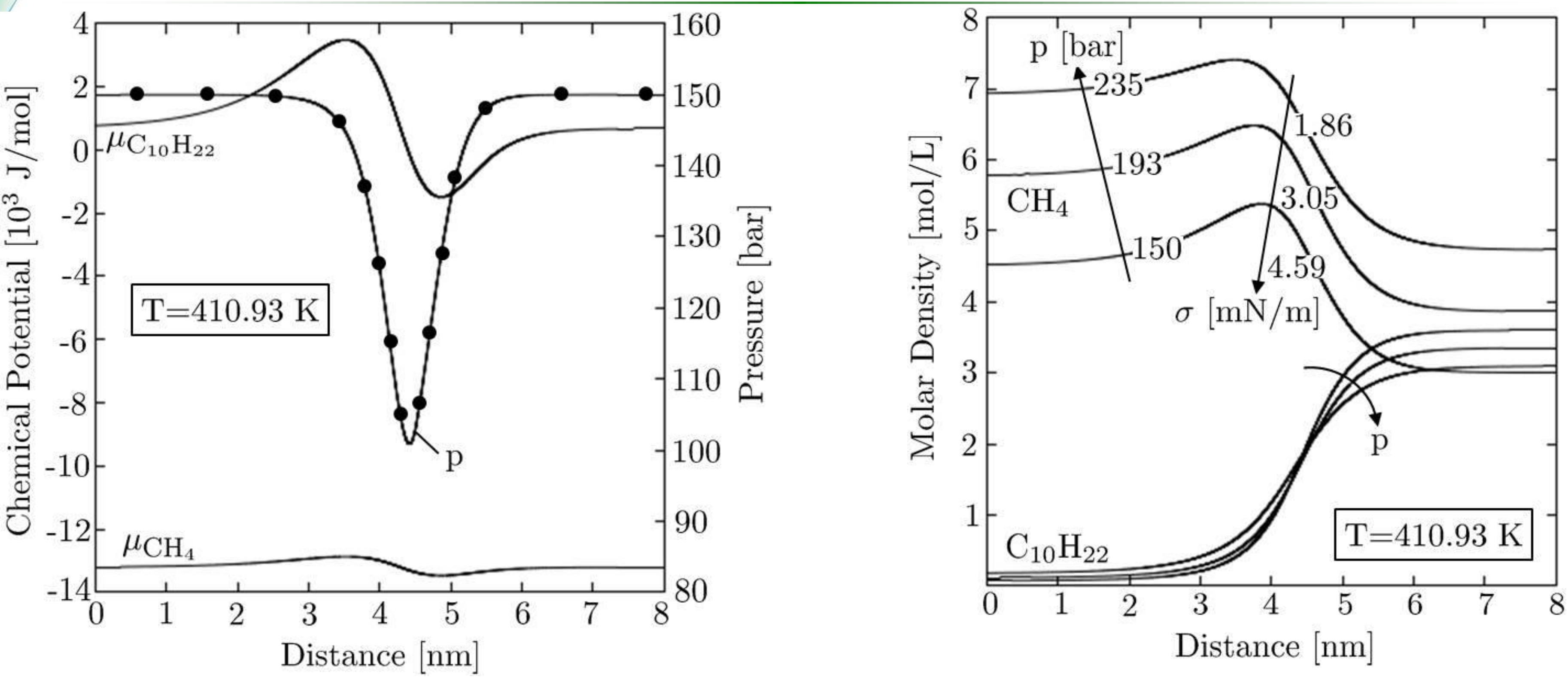
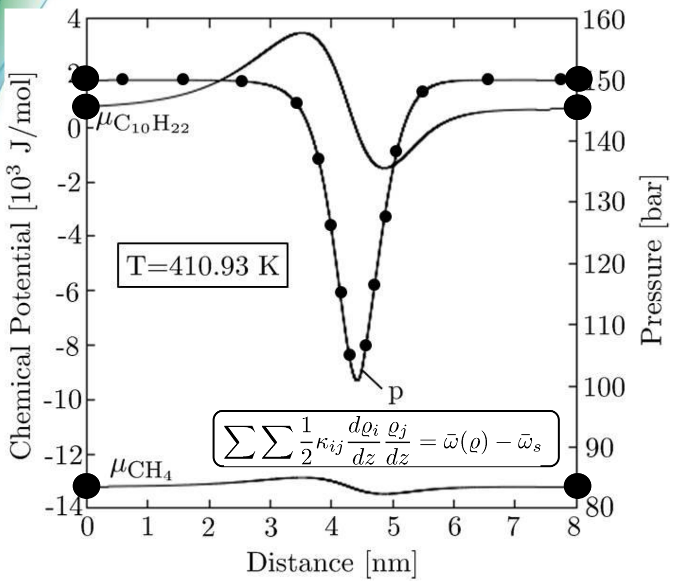


- **Chem. pot./pressure determined by density, composition, gradient energy**
  - Distributions also governed by interfacial minimal Helmholtz free energy state

- **Minimal free energy state maintained in the limit of low surface tension**
  - Fundamental property distributions remain valid $\left(\sum\sum\frac{1}{2}\kappa_{ij}\frac{d\varrho_i}{dz}\frac{\varrho_j}{dz}=\bar{\omega}(\varrho)-\bar{\omega}_s\right)$
  - Interactions with flow field remain negligible (separation of scales)

From molecular dynamics to classic two-phase theory and spray atomization



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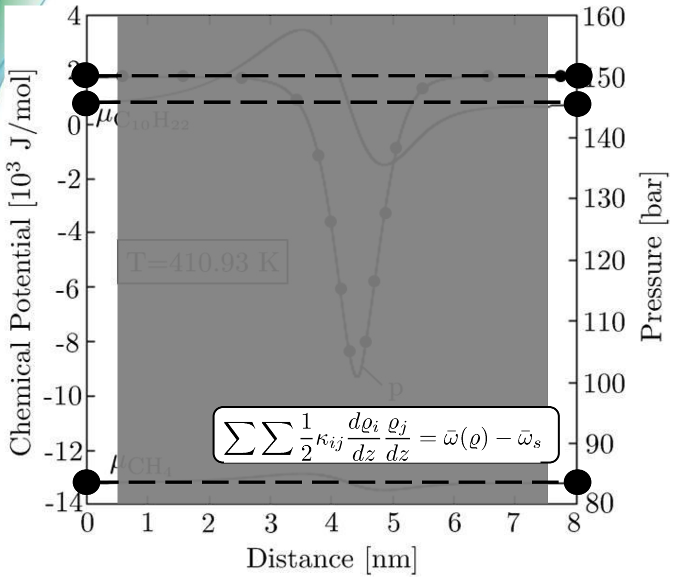

$$\sum\sum \frac{1}{2}\kappa_{ij}\frac{d\varrho_i}{dz}\frac{\varrho_j}{dz} = \bar{\omega}(\varrho) - \bar{\omega}_s$$

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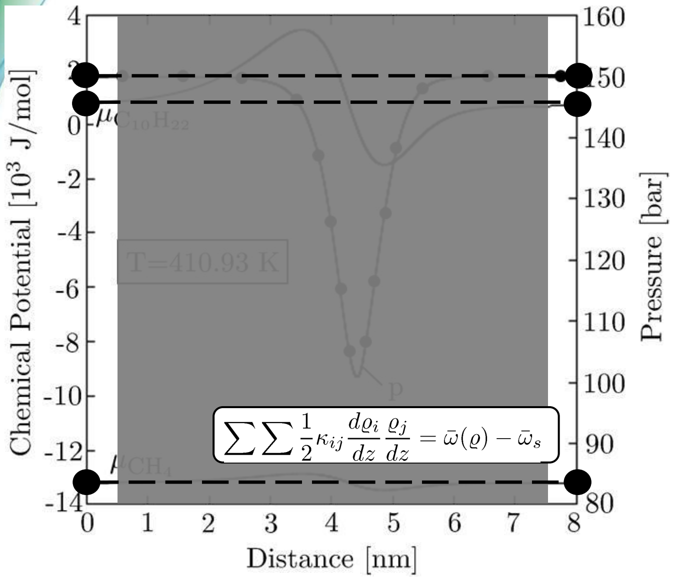

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❑ **Chemical potential/pressure distribution recovers vapor–liquid–equilibrium theory**

$$\begin{aligned} T^V &= T^L \\ p^V &= p^L \\ \mu_i^V &= \mu_i^L \end{aligned}$$

- Assumption in two-phase theory: Minimal free energy !
- Justifies calculations of true critical points of mixtures
- Justifies evaporation and heating laws, Spalding transfer number, D^2 – law, etc.

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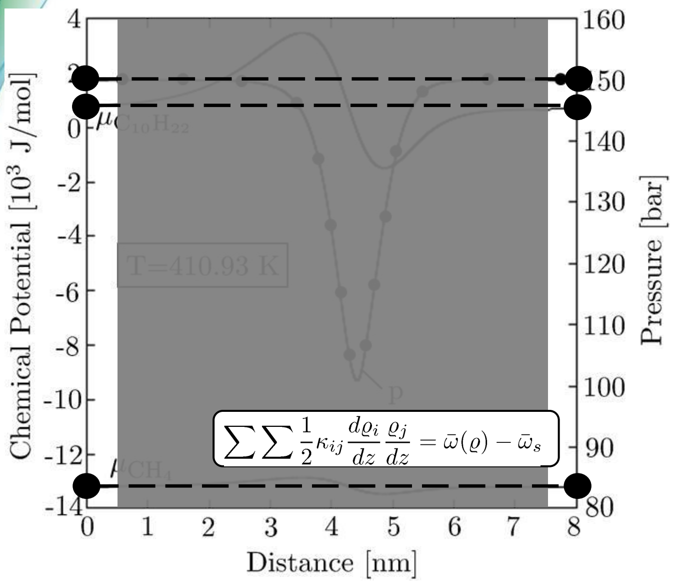

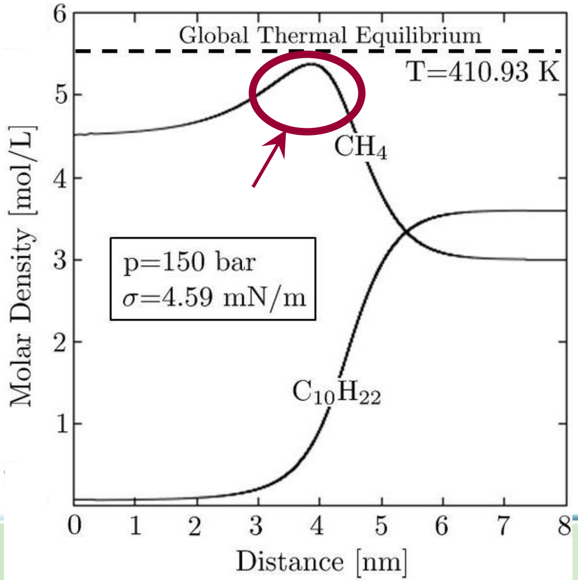

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❑ **Accumulation of methane within the interface**

- – Lowers interfacial Helmholtz free energy
- – Hence, species distribution maintained regardless of differences (T, X_i) between gas & liquid
- • Immiscible boundary, mixing is prohibited
- • Determines surface tension forces
- ➢ Must lead to breakup processes

➢ **Atomization, evaporation & drop formation**

Fundamental changes in interfacial molecular dynamics

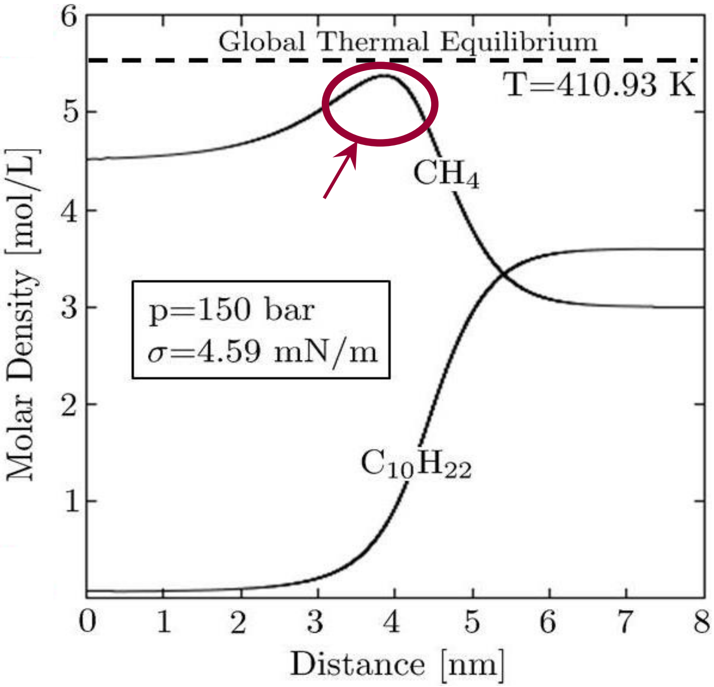
- **Molecular dynamics governed by minimizing Helmholtz free energy at equilibrium**

$$\sum\sum\frac{1}{2}\kappa_{ij}\frac{d\varrho_i}{dz}\frac{\varrho_j}{dz}=\bar{\omega}(\varrho)-\bar{\omega}_s \longrightarrow$$

- **Helmholtz free energy only minimized in isothermal systems**

$$dF \leq -SdT - pdV$$

- **Thermal equilibrium unsustainable if internal temperature gradients develop**



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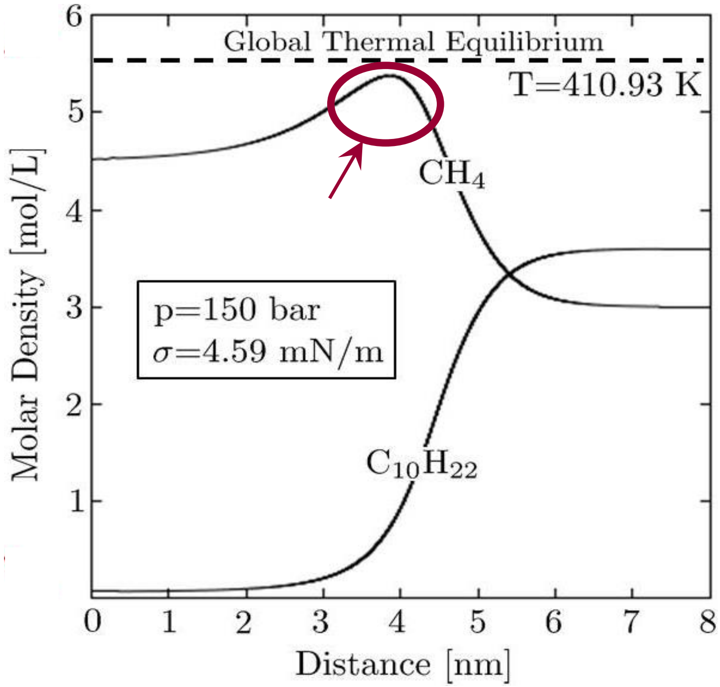


- **Fundamental thermodynamics: Under non-isothermal constraints, Helmholtz free energy no longer be minimized at equilibrium!**
  - Fundamental equation becomes invalid
  - Breakdown of classic two-phase relations
  - Fundamental dynamics which lead to sprays & drop dynamics no longer apply

$$\cancel{\sum\sum\frac{1}{2}\kappa_{ij}\frac{d\varrho_i}{dz}\frac{\varrho_j}{dz}=\bar{\omega}(\varrho)-\bar{\omega}_s}$$

Development of
interfacial temperature gradients

- ❑ **At what conditions becomes Fourier's law valid across the otherwise isothermal interface?**

$$\frac{\partial T}{\partial t} = \frac{k}{\varrho c_p} \nabla^2 T$$

- ❑ **Minimal length scale derivable in analogy to Fick's diffusion law relations**

$$q_{\max} = -\varrho c_p \Delta T \bar{v}$$

$\bar{v}$: Mean molecular particle speed

$$q_{\text{Fourier}} = -k \nabla T$$

$$\longrightarrow \boxed{\ell_{Th} \sim \frac{k}{\varrho c_p \bar{v}}}$$

- ❑ **Burnett & Super-Burnett extensions to Navier-Stokes equations**
  - ➢ Meaningful temperature differences develop over ~5 times this length scale

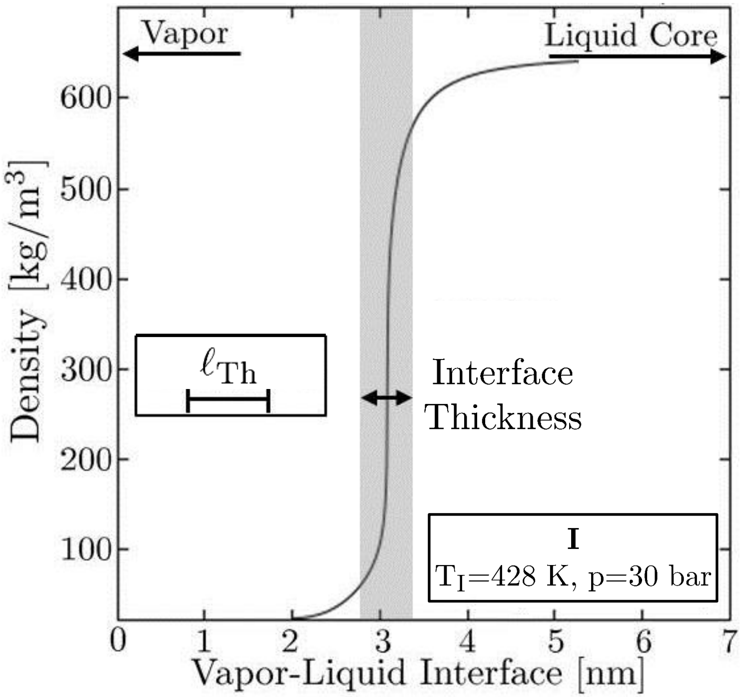
- ❑ **Required interface thickness for interfacial temperature gradients**

$$\longrightarrow \boxed{\Delta \ell \geq 5 \cdot \ell_{Th}}$$

Two-phase interface structures & collision lengths

- ❑ Careful comparison of imaging & simulation
- ❑ Liquid n-dodecane C_{12} (T=363 K) into gaseous nitrogen at different conditions

“Low” Pressure (30 bar)



Isothermal Interface
Classic two-phase theory valid

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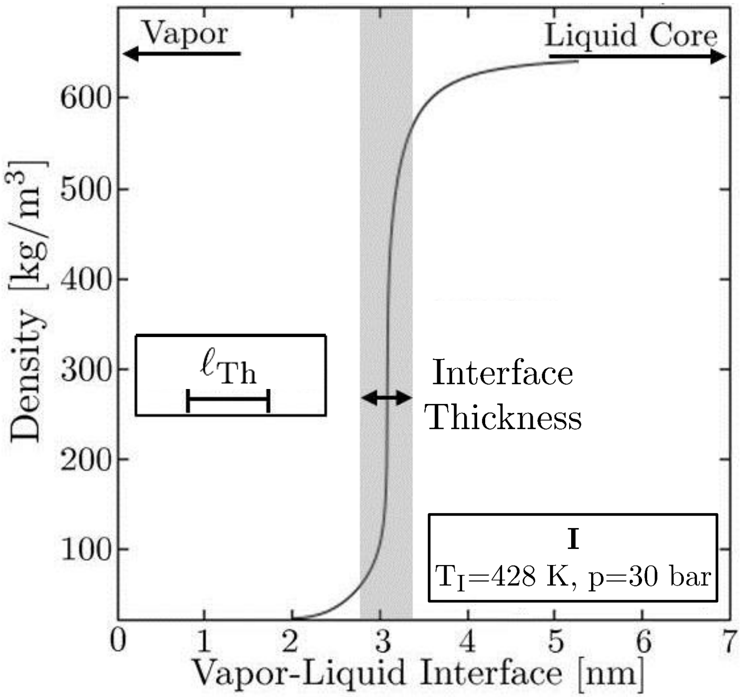
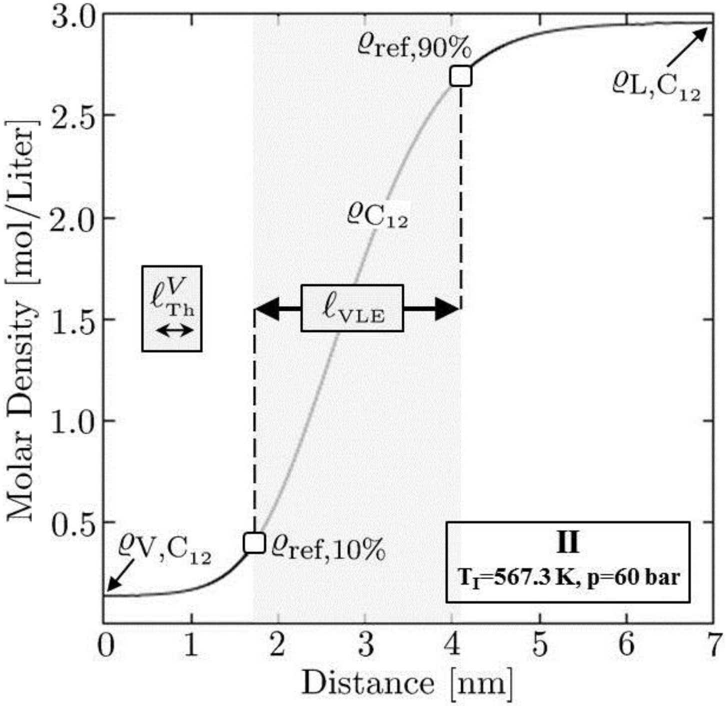


Isothermal Interface

Classic two-phase theory valid

"High" Pressure (60 bar)



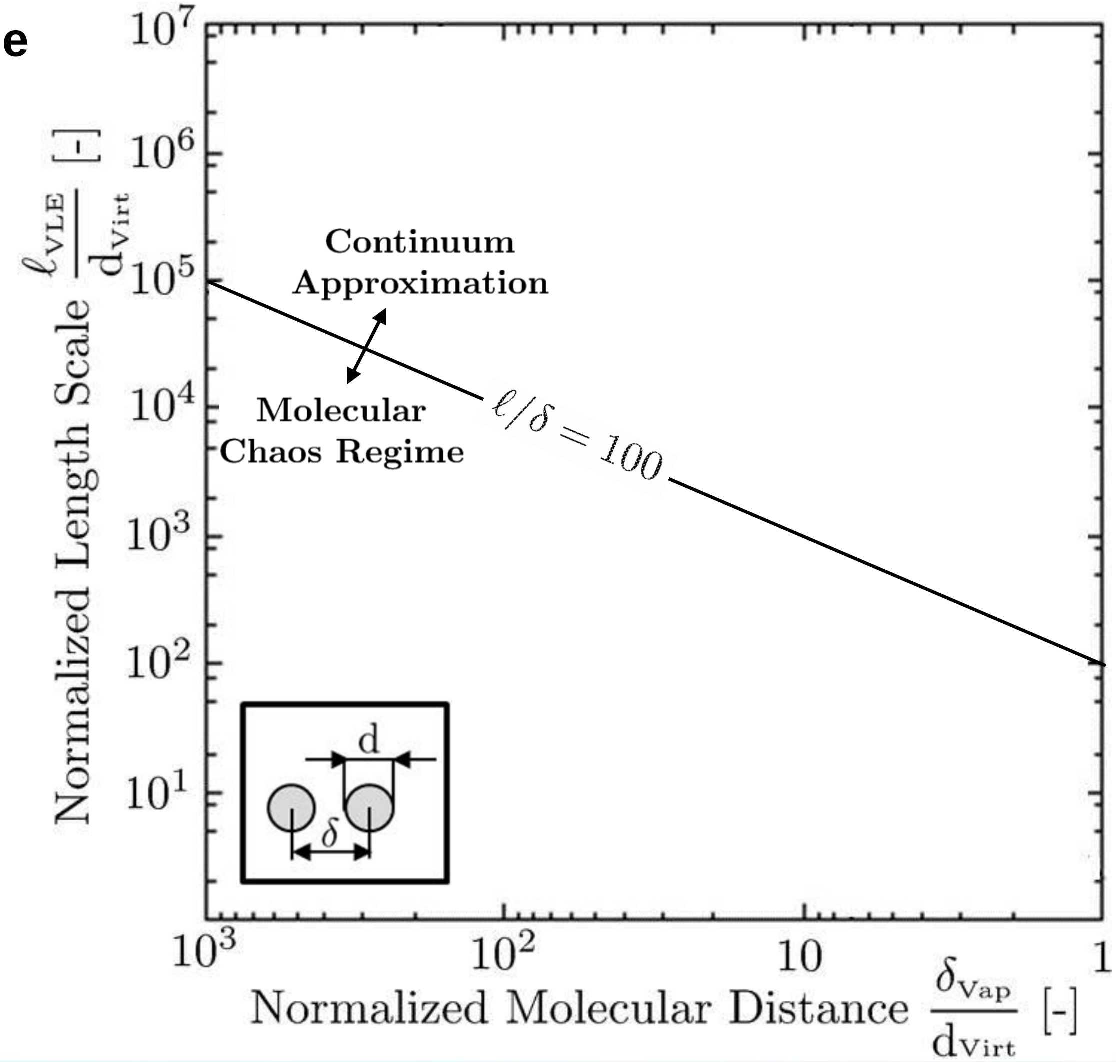
Non-isothermal Interface

Breakdown of spray atomization

Non-equilibrium nano-scale flow dynamics within fluid interfaces

- **Classic micro-scale flow regime diagram for pure gas flows**
  G.A. Bird, “Molecular gas dynamics and the direct simulation of gas flows,” *Oxford University Press*, New York, 1994

- **Modified for two-phase multi-component interfaces**



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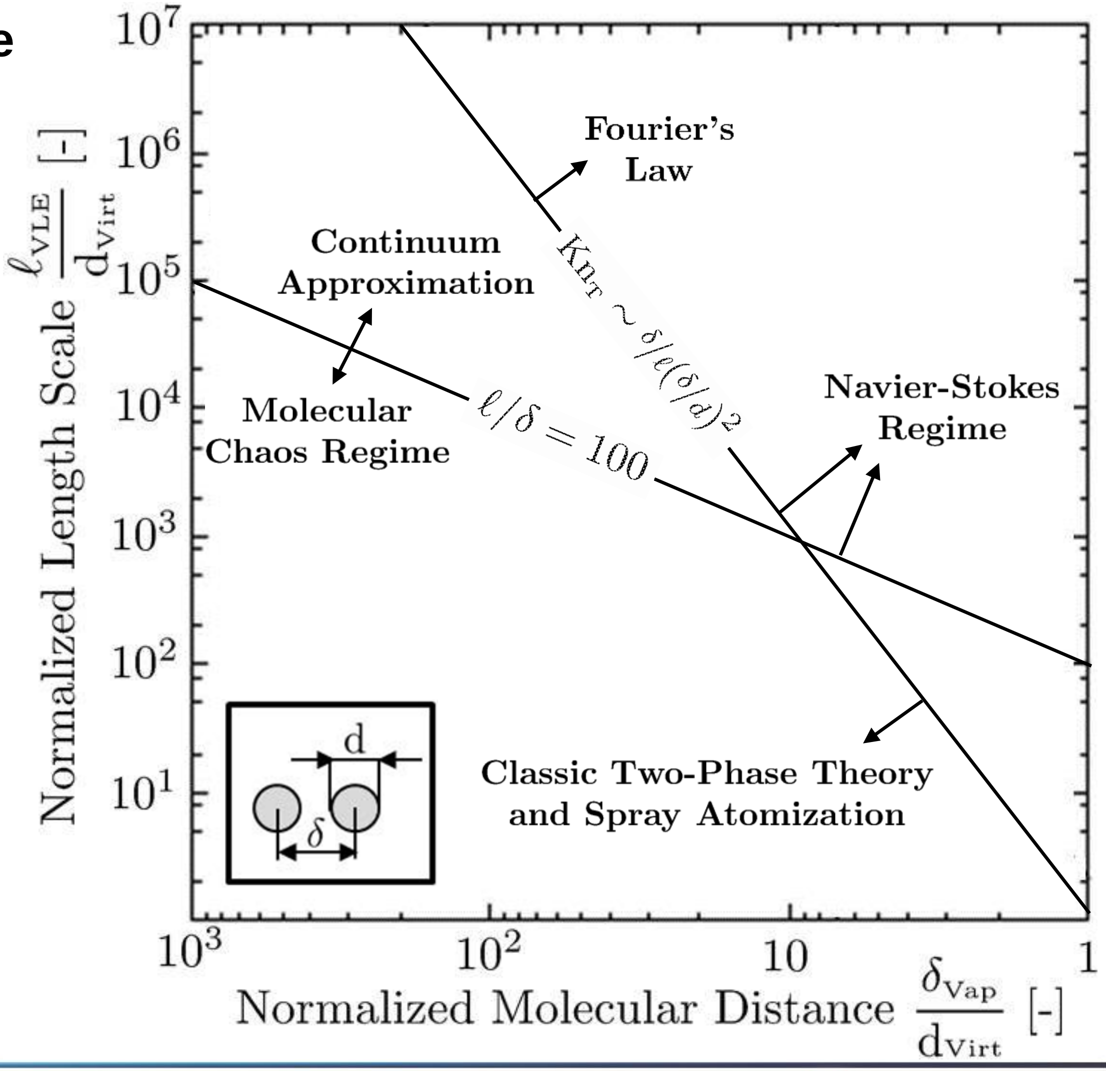

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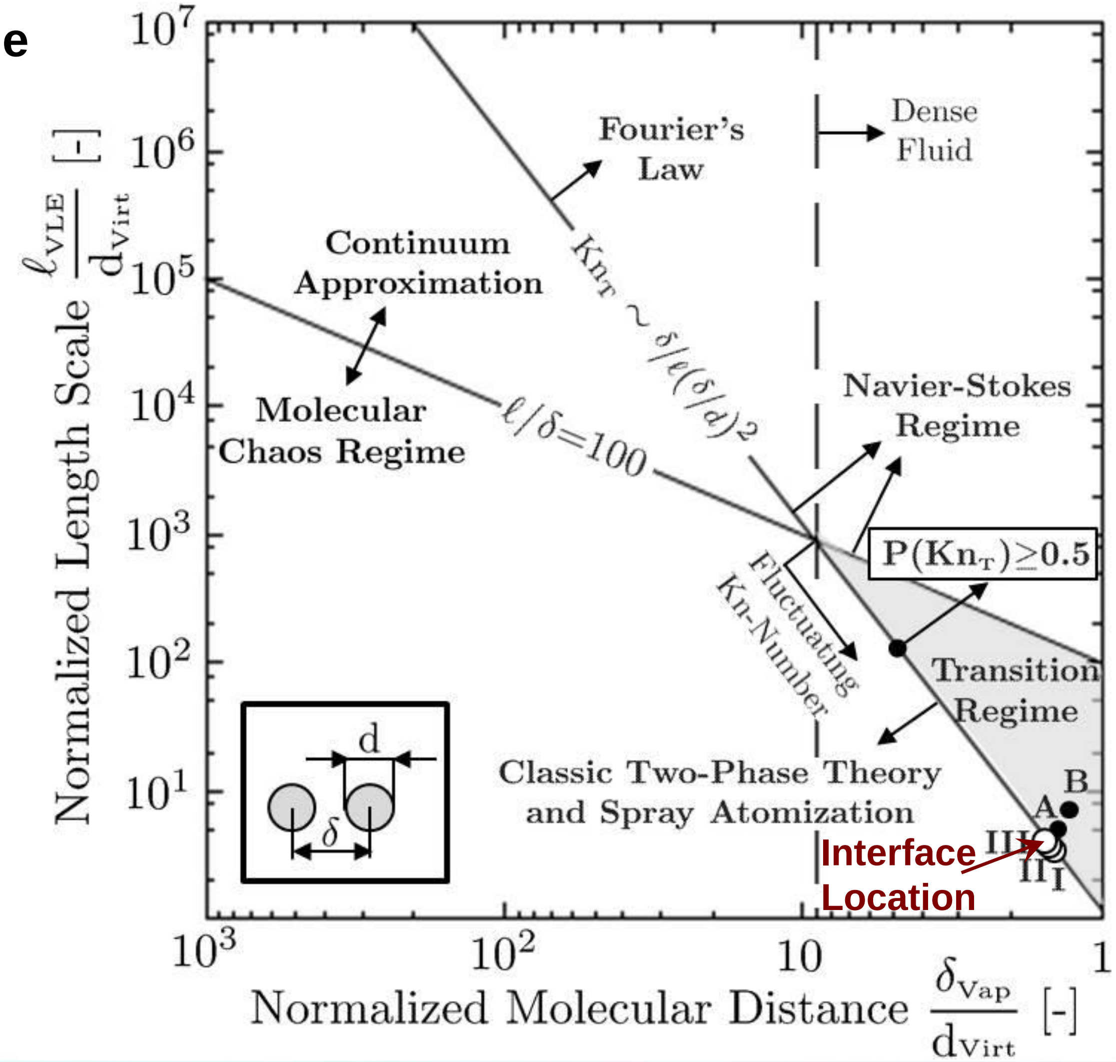
- **Modified for two-phase multi-component interfaces**
- **Non-isothermal interfaces**
  - Navier-Stokes invalid !
- **Transition regime**
  - Bridges classic two-phase & dense-fluid mixing dynamics



Time scale analysis
From two-phase relations to single-phase mixing

- ☐ **Navier-Stokes regime from micro-scale regime diagram**

$$\boxed{\frac{\ell}{d} \sim 500} \qquad \boxed{\ell \sim 250 \text{ nm}} \qquad \boxed{\Delta\tau \sim \frac{\ell^2}{D} \sim 10\ \mu s}$$

- ☐ **Transition time scale: Micro-seconds rather than nano-seconds**
  - Meaningful to liquid dynamics, 30% of liquid penetration in diesel spray

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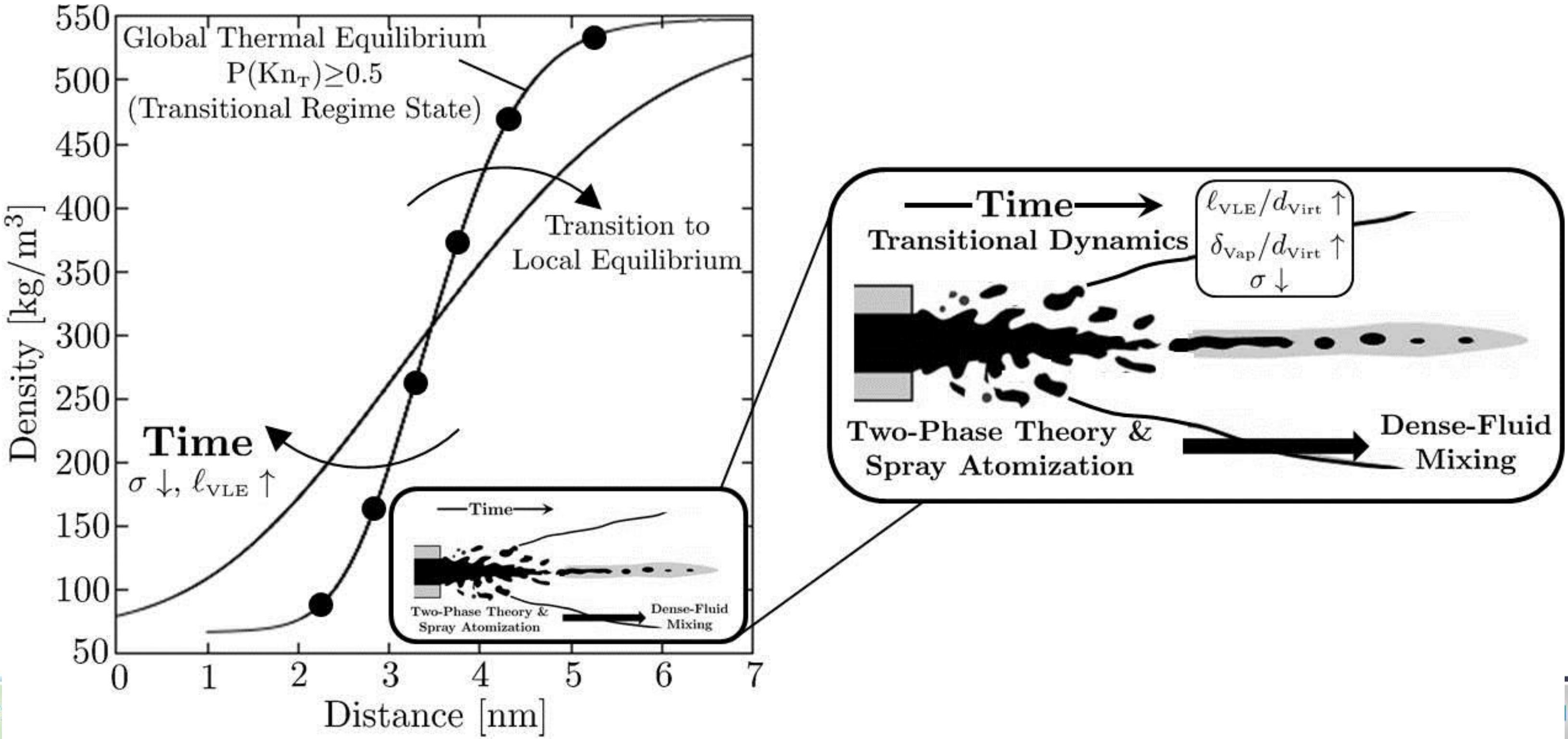
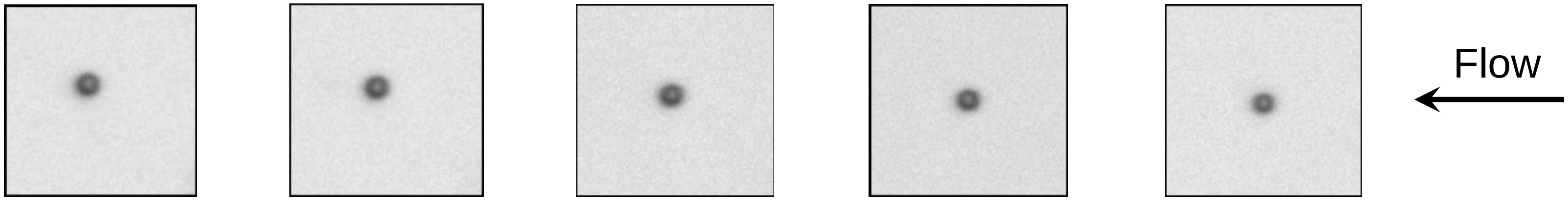


Imaging of transitional mixing dynamics
LC_{12} (363 K) into GN_2

Crua *et al*, “Transition from droplet evaporation to miscible mixing at diesel engine conditions,” ICLASS 2015, August 23-27, 2015

Classic Evaporation

p=62 bar
T=700 K



100 µs 133 µs 857 µs 1400 µs 1800 µs

- ❑ **At predicted “supercritical pressure” conditions:**
  - Sphere’s diameter vanishes over time → Classic evaporation

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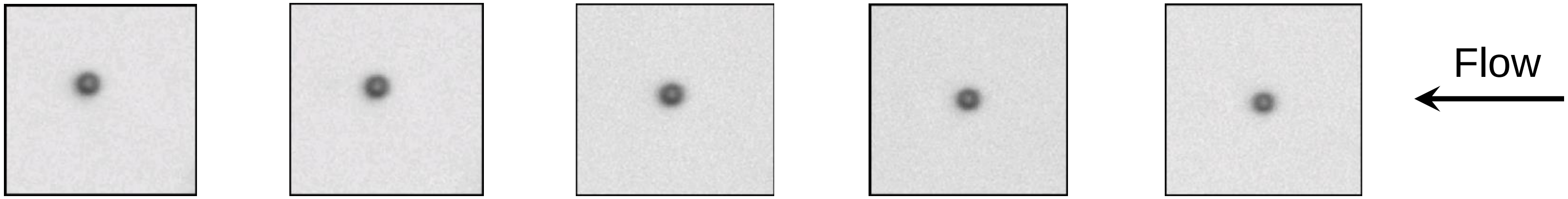

100 µs 133 µs 857 µs 1400 µs 1800 µs

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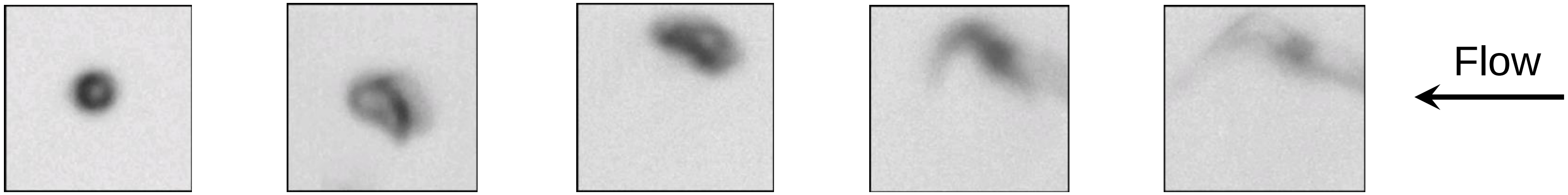
  - Sphere’s diameter vanishes over time → Classic evaporation

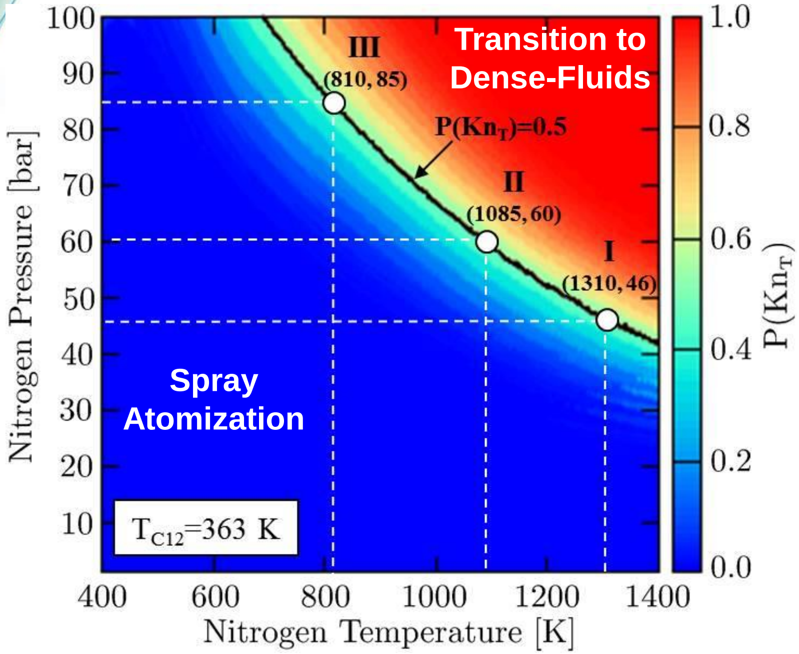
Transitional Mixing

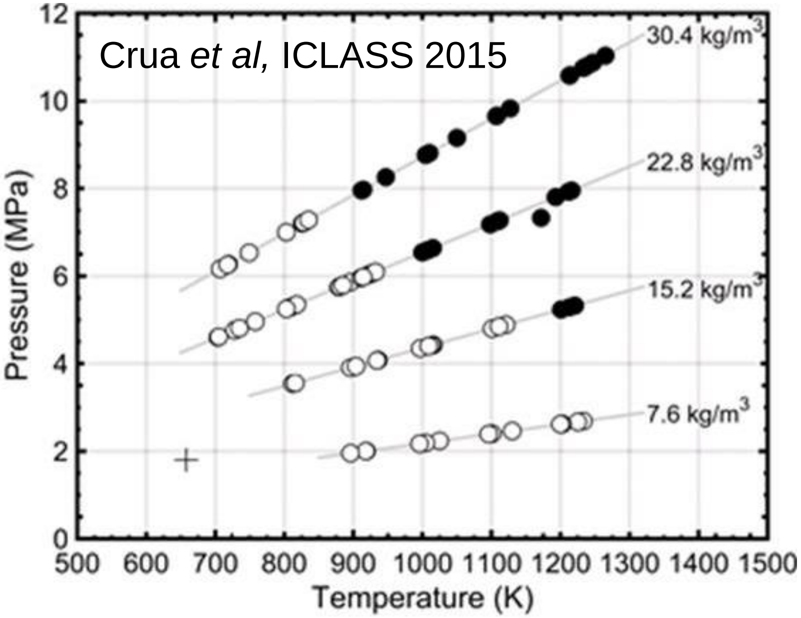
p=88 bar
T=1000 K



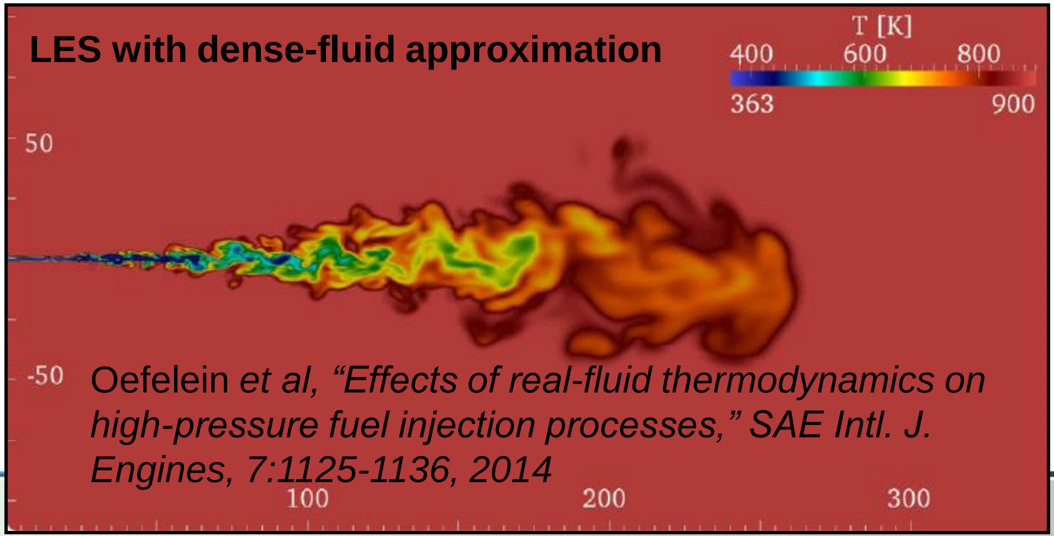
- **At predicted conditions:**
  - Initial spherical drop shape evidence of surface tension
  - Liquid deformation & mixing with flow evidence of vanishing interface

Regime diagram of liquid injection
Example: N-dodecane / nitrogen





Crua *et al*, ICLASS 2015

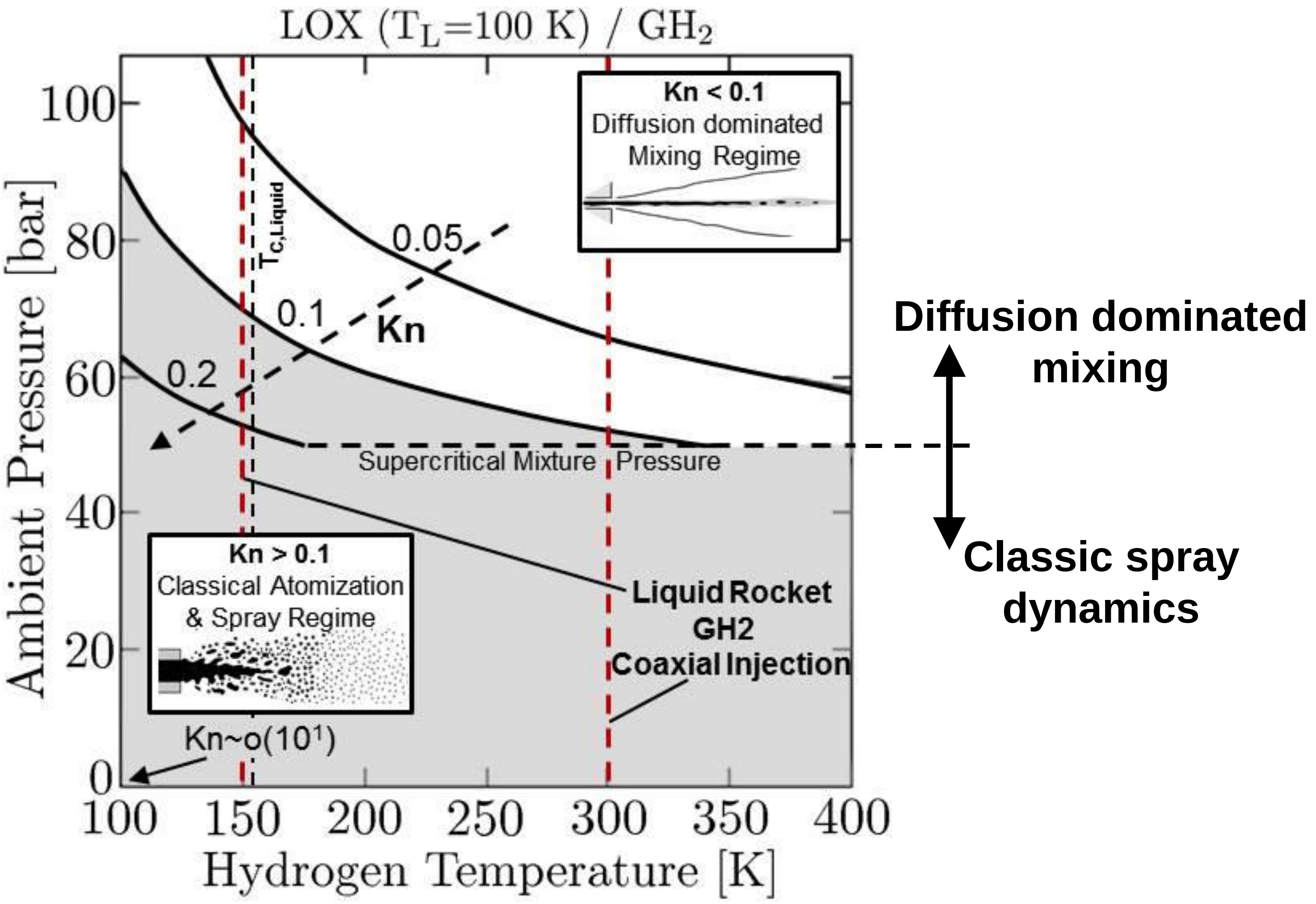


LES with dense-fluid approximation

Oefelein *et al, "Effects of real-fluid thermodynamics on high-pressure fuel injection processes," SAE Intl. J. Engines, 7:1125-1136, 2014*

- ☐ **Molecular interface simulations predict transition conditions**
- ☐ **Performed experiments validated predictions**
- ☐ **High-fidelity LES using dense-fluid approximation utilizes predictions**

Cryogenic LOX-H2:
Coincidentally, dense-fluids often exist beyond $p_{C,O2}$



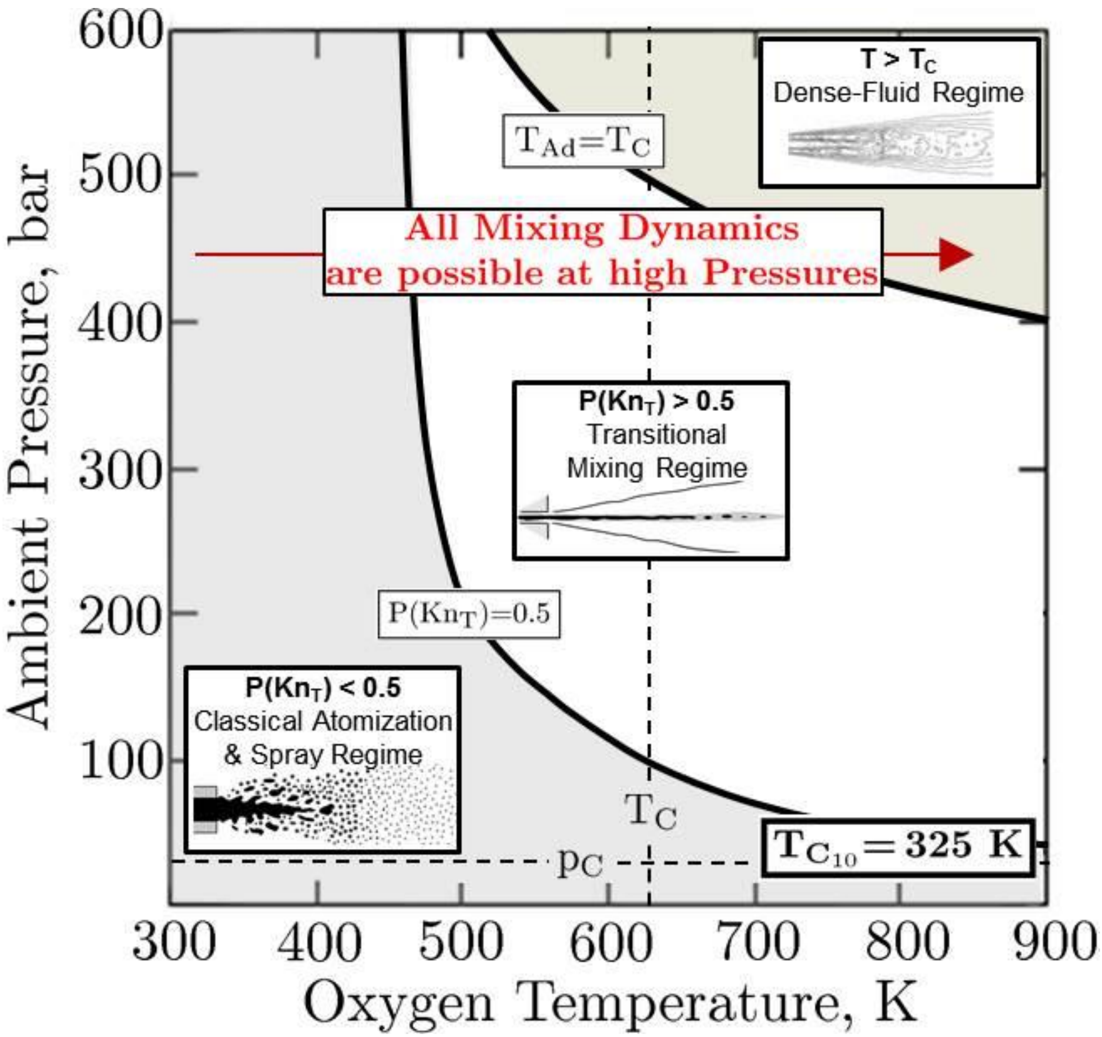
Diffusion dominated mixing

Classic spray dynamics

Hydrocarbon fuels: Complex mixing dynamics even at very high pressures

- **Dependent on gas temperature:** Injection may exhibit all forms of mixing dynamics

- **Liquid injection temperature:** Significantly alters locations of regime separators



Summary & Conclusions

I. Real-fluid framework for thermodynamics, transport & capillary effects applies at arbitrary mixture states

II. Molecular two-phase dynamics, governed by minimizing Helmholtz free energy, lead to classic two-phase theory & spray atomization

III. For non-isothermal interfaces, Helmholtz free energy, along with two-phase theory & spray atomization, becomes meaningless

IV. Non-equilibrium transitional dynamics to gas-liquid mixing layer seamlessly extend classic two-phase theory

V. Cryogenic conditions often result, coincidentally, in dense-fluid jets above chamber pressures comparable to pure liquid critical pressure

VI. Hydrocarbon fuels show complex dynamics even at very high pressures

COMBUSTION
RESEARCH
FACILITY

The breakdown of classic spray atomization at engine-relevant conditions

Thank you for your attention!

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U.S. DEPARTMENT OF ENERGY | Office of Science

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences

Acknowledgement: Eric W. Lemmon, NIST, Boulder, CO
Cyril Crua, University of Brighton, Brighton, UK