Heterogeneous nucleation in the long range Ising model

James B. Silva

Boston University

October 15, 2013
**What is Nucleation?**

Process that starts the decay of a metastable state into the stable state.
OUTLINE

Introduction
An introduction to the process of nucleation
Whats important about nucleation?

Quantifying nucleation in the real world
What is our current understanding of nucleation?
Does our understanding of nucleation work

Building a model for heterogeneous nucleation
The long range Ising model
Exploring heterogeneous nucleation in the Ising model

Unresolved questions in nucleation
Work to be done in nucleation

Summary
CLASSIFYING NUCLEATION PROCESSES

▶ Homogeneous Nucleation
  ▶ Nucleation which occurs in the bulk with no preferential location
▶ Heterogeneous Nucleation
  ▶ Nucleation which occurs in preferential locations

Figure: Homogeneous nucleation in the Ising model.

Figure: Liquid-Crystal nucleation in a charged sphere colloidal system.
Why care about nucleation?

- Nucleation is a key process in many important larger processes in nature and industrial applications.
  - Heterogeneous nucleation appears to be important process in atmospheric physics. Heterogeneous nucleation has an influence on the amount of precipitation. [kulmala13]
  - Ice formation causes cell damage. Heterogeneous nucleation used to model ice formation in cells [toner90]
  - Damage to turbines caused by steam with droplets of excess size. [bakhtar95]
  - Aerosols used in various industrial applications: Paint finishes on products
**CLASSICAL NUCLEATION THEORY (CNT)**

- Nucleation is viewed as growth on the surface of a droplet of the stable phase.

- Free energy given by competing sum of surface area and bulk contribution. Example (liquid-gas): Given density $\rho$, chemical potential $\mu$, and surface tension of liquid droplet $\gamma$. 
Spinodal is the limit of metastability of a metastable state.
Theoretical description of nucleation near the spinodal. [klein84]
- Nucleation is viewed as coalescence process of fractal-like percolation clusters.
Nucleation near the classical spinodal

- Percolation first popularized by Broadbent and Hammersley in the 1950s.
- Given sites are clustered based on some given bond probability.
Nucleation and Percolation: Defining Clusters

- Theory predicts a more disperse nucleating droplet as spinodal is approached. A breakdown of CNT.
- Percolating clusters defined with a bond probability.

\[ p = 1 - e^{-\beta J} \]
Nucleation and Percolation

- Resulting size of clusters scales as following

\[ \eta_{s,T} = \frac{g((T - T_c)s^\sigma)}{s^\tau} \]

- Example at Tc: Gas (White) : Liquid (Black)
  Largest Cluster (Yellow) : Second Largest Cluster (Cyan)
EVALUATION OF CNT WITH EXPERIMENTAL DATA

- Classical Nucleation Theory (CNT) appears to agree with detailed measurement in limited cases like the short range Ising model. [binder75] [ryu10]

- Classical Nucleation Theory (CNT) appears to have trouble in agreeing with experimental data in other nucleating systems like glass oxides and colloids. [granasy98]

- Dense spherical droplet of classical nucleation theory (CNT) does not agree particularly well with experiments in colloids. [wang12]
THE ISING MODEL

Ising model nucleation event.

- Define spin variable $\sigma = +1, -1$.
- Define a field $h_i$.
- Hamiltonian given by

$$H_{\text{Ising}} = \sum_{<i,j>} J_{i,j} \sigma_i \sigma_j - \sum_j h_j \sigma_j$$

- Ferromagnetic : $J_{i,j} > 0$
- Anti-Ferromagnetic : $J_{i,j} < 0$
A Model With Heterogeneous Nucleation: Fixing Spins in an Ising Model

- Build a model for heterogeneous nucleation by adding “impurities” in the form of fixed spins.
- Spins are randomly quenched into a given value.
- Example:
  +1 (White) : -1 (Black)
  -1 Fixed (Magenta) : Largest Cluster (Yellow)
A Model With Heterogeneous Nucleation: Fixing Spins in an Ising Model

- Similar model used by others as a model with heterogeneous nucleation in other applications:
  - To determine limit of stability with respect to fixed spins in the short range. [poole13]
  - Nucleation near spinodal for very small percentage of fixed spins. Crossover from homogeneous and heterogeneous with interaction range. [gulbahce04]
  - Used to model heterogeneous nucleation on a crevice in short range system. [curcio10][frenkel06]
PERCOLATION MAPPING IN LONG RANGE HETEROGENEOUS ISING SYSTEM

- How applicable is our understanding of nucleation near the spinodal to heterogeneous systems.
  - If current theory for spinodal nucleation is not applicable. What is the cause?
- Will begin by determining if percolating clusters result in same behavior as pure system
- How does heterogeneity change the interface between the droplet and rest of system
DEFINING PERCOLATING CLUSTERS IN THE LONG RANGE HETEROGENEOUS ISING SYSTEM.

▶ Loss of scaling as fixed spin density is increased.

Cluster Size Frequency $L=200 \, R=10 \, T=3.928 \, h=0$
DEFINING PERCOLATING CLUSTERS IN THE LONG RANGE HETEROGENEOUS ISING SYSTEM

- Scaling is restored for given fixed spin density if extra field is applied $h = -4x$.

Cluster Size Frequency $L=128$ $R=7$ $T=T_c$
DEFINING A MEASURE OF HETEROGENEITY:
DIFFERENTIATING HETEROGENOUS SYSTEMS

Figure: Nucleation probability. Figure: Coarse grained fixed spins.

- Fixed spin coarse graining is proportional to probability of nucleation occurring in a given site.
- Can differentiate heterogeneous systems and determine the relation of heterogeneity to properties of droplet-background interface.
Nucleating Droplet Interface

- Increased heterogeneity ⇒ more disperse droplet.
- Diminishing applicability of classical nucleation theory.

Local Spin Density $L=128 \ R=8$
WORK TO BE DONE IN NUCLEATION

- Role of self-averaging on nucleation free energy barriers in long range systems. [sear12]
  - Given a sample of a material of a given density of impurities. To what degree does heterogeneity account for different nucleation rates
- Role of geometry / impurity distribution on nucleation rates.
- Role of impurities on droplet growth.
Nucleation Summary

- Nucleation is an important process in areas such as atmospheric physics, biological physics, and various industrial applications.
- The long range Ising model is a valuable tool to test various nucleation theories such as CNT and spinodal nucleation.
- Preliminary results show a more disperse droplet which is indicative of a bigger departure from the applicability of classical nucleation theory.
- More work needs to be done on nucleating droplet growth and the effects of impurity distribution on energy barrier effects.
COLLABORATORS

- W. Klein
- H. Gould
- K. Liu
- N. Lubbers
REFERENCES I
Measuring Local Spin Density: Process

- Quench system into temperature $T$ and field $h$
- Flip field to $-h$ such that system is now in a metastable phase
- Allow system to reach stable state. Determine nucleation time through intervention technique.
- Save configuration of nucleating droplet at nucleation time.
- Determine center of nucleating drop and measure coarse grained density around given location.
Metastable state lifetime: Process

- Quench system into temperature $T$ and field $h$
- Flip field to $-h$ such that system is now in a metastable phase
- Allow system to reach stable state and measure time until droplet grows until $M_{\text{thresh}} = -0.8$. 
Metastable lifetime in long range heterogeneous Ising system

- Drop in nucleation energy barrier expected → drop in metastable lifetime.